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Zeolitic Diagenesis of Tuffs in the Miocene Chalk Hills Formation, Western Snake River Plain, Idaho

By RICHARD A. SHEPPARD

Silicic vitric tuffs south of Oreana are completely altered to clinoptilolite with or without smectite and opal-CT

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U.S. DEPARTMENT OF THE INTERIOR MANUEL LUJAN, JR., Secretary

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Zeolitic Diagenesis of Tuffs in the Miocene Chalk Hills Formation, Western Snake River Plain, Idaho

By Richard A. Sheppard

Abstract

Zeolites are common diagenetic alteration products of silicic, vitric tuffs in Cenozoic lacustrine deposits of the western United States, especially those that were deposited in highly saline and alkaline water. Such deposits have received much attention in the last 25 years because they contain economic or potentially economic phillipsite, chabazite, erionite, and clinoptilolite. In contrast, study of zeolites in relatively freshwater lacustrine deposits has been neglected. The Miocene Chalk Hills Formation of southwestern Idaho consists of about 100 m (meters) of mainly mudstone, siltstone, and interbedded vitric tuffs that were deposited in a chiefly freshwater lake. Throughout most of the outcrop belt, the tuffs are unaltered, but, locally, the tuffs are completely altered to clinoptilolite with or without smectite and opal-CT. This study was undertaken to delineate the pattern of alteration, to characterize the authigenic minerals and their paragenetic relationships, and to suggest a possible origin.

About 300 samples of fresh and diagenetically altered tuffs and tuffaceous sediments were collected throughout the formation and from an area of about 500 km² (square kilometers). A marker tuff, 1–13 m thick, is the thickest and most continuous tuff in the area, and this tuff was most extensively sampled and studied. The relative abundance of zeolites and other authigenic minerals in all samples was determined by X-ray powder diffraction. Selected samples were also studied by scanning electron microscopy and thinsection optical microscopy to ascertain the process of genesis and the paragenetic relationships of the minerals, and by X-ray fluorescence analysis to chemically characterize the zeolites and to provide data on the chemical changes during alteration.

The zeolite zone in the Chalk Hills Formation is south of Oreana, Idaho, and extends parallel to the southern margin of the Snake River Plain, northwestward from Birch Creek to Hart Creek, about 21 km. Fresh, rhyolitic, vitric tuffs that are about 1 cm (centimeter) to 13 m thick grade laterally into zeolitic tuffs. Original textures and sedimentary structures are

generally preserved in the tuffs of the zeolite zone. Fresh (unaltered) tuffs are light gray and soft, but altered tuffs are white, light yellow, or light brown and relatively hard, commonly forming ledges. Zeolitic tuff generally consists of clinoptilolite, smectite, and opal-CT. Although some tuffs are locally nearly monomineralic clinoptilolite, most contain 40-90 percent clinoptilolite. Trace to minor amounts of erionite are associated with the clinoptilolite at a few localities. Trace amounts of fibrous to filiform mordenite are commonly associated with the clinoptilolite, but the mordenite can be recognized only by scanning electron microscopy. Phillipsite was recognized at only one locality where it makes up about 70 percent of the basal centimeter of the clinoptilolite-rich marker tuff. Other authigenic minerals locally recognized in the zeolitic tuffs are quartz, calcite, and gypsum. The common paragenetic sequence (listed early to late) is glasssmectite—clinoptilolite—mordenite. Opal-CT crystallized after smectite and either before or after clinoptilolite. Erionite, quartz, and calcite generally crystallized later than the clinoptilolite, and quartz and calcite locally replaced all of the earlier authigenic silicate minerals.

The zeolites and associated silicate minerals undoubtedly formed during diagenesis by hydrolysis and dissolution of rhyolitic glass by pore water that was trapped in the tuffs during lacustrine sedimentation. Where tuffs are still unaltered, the original lake water probably was fresh. The zeolite zone probably coincides with a part of the lake that had a low to moderate salinity and a pH of 7–9.

INTRODUCTION

The Miocene Chalk Hills Formation is chiefly a lacustrine deposit that crops out discontinuously along the southwestern margin of the western Snake River Plain from Adrian, Oregon, southeastward to Bruneau, Idaho, a distance of about 140 km (fig. 1). Most of this area is in the northwestern part of Owyhee County, Idaho. The Snake River Plain is the only major expanse of relatively gentle topography in Idaho, and this part of the Snake River Plain

is characterized by badlands at the base of conspicuous northeast-trending benches. The Chalk Hills Formation crops out mainly in the badlands.

Zeolites are common alteration products of silicic, vitric tuffs in Cenozoic lacustrine deposits of the western United States, especially those that were deposited in highly saline and alkaline water (Hay, 1966). These deposits have received much attention in the last 25 years because they contain economic or potentially economic phillipsite, chabazite, erionite, and clinoptilolite. In contrast, study of zeolitic tuffs in relatively freshwater lacustrine deposits has been neglected. The Chalk Hills Formation contains at least 20 interbedded vitric tuffs that were deposited in a chiefly freshwater lake. Throughout most of the outcrop belt, the tuffs are unaltered, but, locally, the tuffs are completely altered to mainly clinoptilolite with or without smectite and opal-CT.

Previous Work

The Miocene Chalk Hills Formation, part of the Idaho Group of Miocene to Pleistocene age, was named by Malde and Powers (1962), but upper Cenozoic lacustrine deposits were recognized in the western Snake River Plain as early as the 1870's (Kimmel, 1979). Since 1962, numerous studies have been conducted on the stratigraphy, paleontology, and depositional environment of the formation, and these have been reviewed by Kimmel (1979, 1982). Swiryd-czuk and others (1982) used chemical analyses and field studies to attempt correlation of tuffs in the Chalk Hills Formation throughout the Snake River Plain. Ekren and

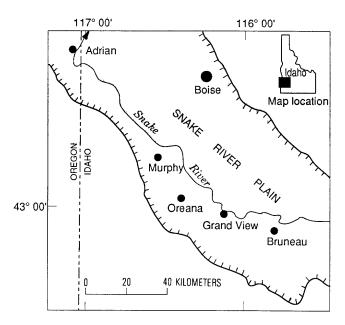


Figure 1. Map showing the margins (hachured lines) of the western Snake River Plain.

others (1981) mapped the geology of the western part of Owyhee County, Idaho, which includes the present study area

Clinoptilolite was first discovered in the Chalk Hills Formation by T.H. Eyde in 1962 at the so-called "Castle Creek zeolite deposit" during a nationwide exploration program for zeolites by the Union Carbide Corporation (Mumpton, 1983). The Castle Creek zeolite deposit is about 2.5 km west of Castle Creek and about 9 km south of Oreana, Idaho (fig. 2; loc. 1, fig. 3). Zeolites in the Chalk Hills Formation have not received much study, and published information concerning them is meager. Regis (1978) investigated the mineralogy and properties of bentonite from the Ben-Jel mine, located in the Chalk Hills Formation about 1.5 km southeast of the Castle Creek zeolite deposit (fig. 2; loc. 2, fig. 3), and found that some of the bentonite contains major amounts of clinoptilolite. Also, Ekren and others (1981) reported that tuffaceous material in the formation is commonly altered to zeolites and montmorillonite, but these authors did not identify the zeolites or give the localities. Two brief reports by Sheppard and Gude (1982, 1983) presented mineralogical, chemical, cationexchange, and gas-adsorption data for clinoptilolite-rich tuff from the Castle Creek deposit.

In 1975, the Double Eagle Petroleum and Mining Company (Casper, Wyo.) filed claims on a clinoptiloliterich tuff at the Castle Creek deposit. Since then, this company has mined, crushed, and stockpiled clinoptiloliterich tuff but has shipped only a minor tonnage for use chiefly in agricultural experiments and tests. Additional claims were made in 1987 on a clinoptilolite-rich tuff east of Castle Creek by Teague Mineral Products (Adrian, Ore.).

Scope of Present Investigation

After a brief reconnaissance of the alteration of the Chalk Hills Formation throughout the outcrop belt of the formation in 1984, this present investigation concentrated on the extensively altered area south of Oreana, Idaho (fig. 3). About 300 samples of fresh and diagenetically altered tuffs and tuffaceous sediments from that area were collected and studied during 1985-88. The tuffaceous rocks of the Chalk Hills Formation were chosen for detailed study for the following reasons: (1) the formation contains numerous tuffs that are both fresh (unaltered) and altered to a variety of authigenic silicate minerals, (2) the formation was subjected to relatively shallow burial and shows only slight deformation, and (3) the formation is well exposed, and individual tuffs can be traced throughout much of the extent of the formation. Objectives of this investigation were to delineate the pattern of zeolite alteration, to characterize the authigenic minerals and their paragenetic relationships, and to suggest a possible origin for the zeolites and associated silicate minerals.



Figure 2. View eastward of Castle Creek zeolite deposit, Idaho. Pit in foreground is in the clinoptilolite-rich marker tuff that was excavated by the Double Eagle Petroleum and Mining Company. Disturbed area in upper right (arrow) is the Ben-Jel bentonite mine. Castle Creek is just beyond the Ben-Jel mine.

Laboratory Methods

The mineralogy of bulk samples was determined by X-ray diffractometer analysis. Samples were first ground to a powder and then packed in aluminum sample holders, exposed to copper radiation, and scanned at 2° 20 per minute over the range of 2°–40° 20. Relative abundances of the minerals were visually estimated from the diffractometer patterns following the procedure outlined by Sheppard and Gude (1968, p. 2–3). Estimates are probably less reliable for samples containing glass or opal-CT because these materials yield rather poor X-ray records.

Textures and paragenetic relationships of the diagenetic minerals were examined in thin sections and immersion oil mounts by transmitted-light microscopy and by scanning electron microscopy of freshly broken fragments. Selected samples to be studied by scanning electron microscopy were coated with a film of gold (about 300 Å (angstrom) thick) just prior to examination.

The chemical compositions of bulk samples and pure separates of clinoptilolite and glass shards were determined by standard X-ray fluorescence analyses. The H_2O content

was determined by weight loss on ignition at 900 °C. An energy-dispersive X-ray unit, attached to the scanning electron microscope, was used to determine the Si:Al ratios of selected zeolites. The pure clinoptilolite and glass-shard concentrates were prepared by crushing the tuff and then disaggregating it in an ultrasonic bath. Clinoptilolite and the glass shards were then concentrated by repeated centrifuging in a heavy-liquid mixture of bromoform and acetone.

Acknowledgments

Grateful appreciation is expressed to those in the U.S. Geological Survey who provided technical assistance during this investigation. Arthur J. Gude, 3d, helped during the field reconnaissance of the western Snake River Plain in 1984 and performed the initial X-ray diffractometer analyses. Gary L. Skipp assisted in the field in 1985 and 1986 and performed X-ray diffractometer analyses and mineral separations in the laboratory. The guidance and patience of James M. Nishi during my use of the scanning electron microscope is greatly appreciated. I also thank

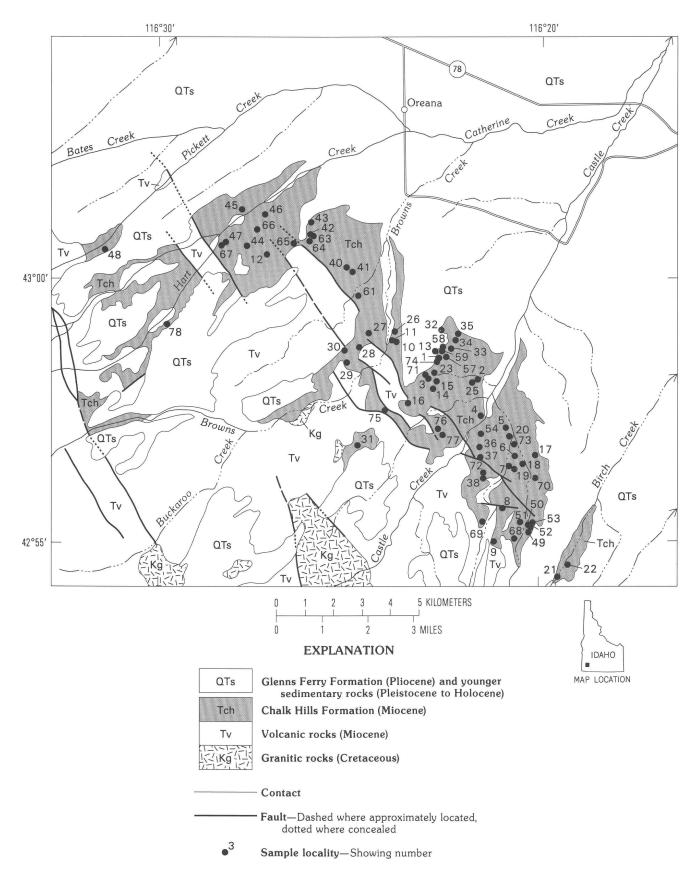


Figure 3. Generalized geologic map showing sample localities in the Chalk Hills Formation south of Oreana, Owyhee County, Idaho; modified from Ekren and others (1981). Mineralogic composition of samples, as estimated from X-ray diffractometer patterns, is given in the appendix.

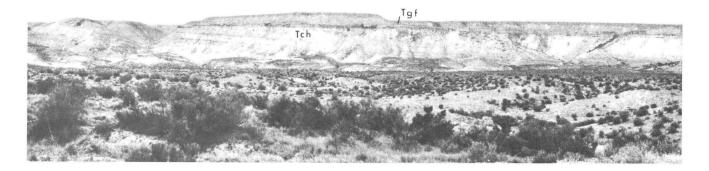


Figure 4. View west to gently dipping Chalk Hills Formation (Tch), about 9 km south of Oreana, Idaho. The light-colored Chalk Hills Formation crops out in the lower part of the conspicuous bench and in badlands at the base of the bench. The Glenns Ferry Formation (Tgf) unconformably overlies the Chalk Hills Formation. A veneer of Quaternary gravel caps the bench.

Barbara A. Lockett for preparing excellent thin sections from difficult materials.

GEOLOGIC SETTING

The western Snake River Plain (fig. 1) is an intermontane rift basin that received several thousand meters of volcanic and sedimentary deposits in Miocene and later time. The western Snake River Plain trends northwest, in contrast to the northeast-trending eastern Snake River Plain. Highlands south of the western Snake River Plain near the study area are underlain chiefly by Cretaceous granitic rocks and Miocene rhyolitic ash-flow tuffs (Ekren and others, 1981). The upper Miocene Chalk Hills Formation unconformably overlies silicic ash-flow tuffs of Miocene age and is unconformably overlain by sedimentary rocks of the Glenns Ferry Formation of Pliocene age. Throughout most of the study area south of Oreana, the basal part of the Glenns Ferry Formation is a brown, conspicuous, ledgeforming, calcareous oolite (Swirydczuk and others, 1979). Similar to the Chalk Hills Formation, the Glenns Ferry Formation contains numerous silicic, vitric tuffs. Unlike the Chalk Hills Formation, however, the tuffs in the Glenns Ferry Formation are unaltered or are partly altered to smectite throughout the region.

Most of the Chalk Hills Formation is lacustrine, but fluviatile, flood-plain, and beach deposits have also been recognized (Kimmel, 1979; Middleton and others, 1985). The formation resulted from the interplay of alluvial and lacustrine depositional processes in an active volcanotectonic setting. Erosion of the Chalk Hills Formation generally has resulted in badlands. South of Oreana, the formation crops out in badlands at the base of conspicuous northeast-trending benches that are capped by Quaternary gravel (fig. 4). The formation commonly dips about 3°–10°

northeastward (basinward) and is cut by numerous north-west-trending faults.

STRATIGRAPHY AND LITHOLOGY OF THE CHALK HILLS FORMATION

The Chalk Hills Formation is about 100 m thick and consists chiefly of mudstone, siltstone, sandstone, diatomite, and numerous thin volcanic ash beds or tuffs. For convenience throughout the remainder of this report, the volcanic ashes are referred to as tuffs even though they are relatively unconsolidated and unaltered. Along the southwestern margin of the Snake River Plain south of Oreana, the thickness of the formation is commonly less than 50 m. This reduced thickness is due to nondeposition on topographic highs of Miocene volcanic rocks and to slight basinward tilting of the Chalk Hills Formation and erosion, particularly at the extreme margin of the basin, prior to deposition of the overlying Glenns Ferry Formation. In addition to the numerous discrete tuffs, most epiclastic rocks in the formation have a tuffaceous component, whether the rock is mudstone or sandstone. Fossil fish, mammals, mollusks, ostracodes, and plants have been collected by others from the Chalk Hills Formation, and their significance has been discussed by Malde and Powers (1962) and Kimmel (1979).

A generalized composite columnar section of the Chalk Hills Formation (fig. 5) was compiled from several partial measured sections from Hart Creek southeast to Birch Creek (fig. 3). Tuffaceous mudstone and siltstone are the predominant lithologies, but tuffaceous sandstone is especially common in the lower part of the section, particularly near the basin margin. Locally, such as at the head of Castle Creek and its tributaries, most of the exposed formation consists of sandstone. The thickest and most continuous tuff in the area studied is in the lower half of the

formation and is termed the "marker tuff" (figs. 5, 6). The marker tuff is 1–13 m thick and averages about 5 m thick. This marker tuff was most extensively sampled and studied.

Sandstone

Sandstone is generally yellowish gray or light brown and is highly variable in grain size, thickness, and induration. Most commonly, sandstone is medium to coarse grained and medium to thick bedded. Crossbedding and ripple marks are locally common. Dips of foresets near the basin margin at Castle Creek and its tributaries indicate a northeast (basinward) transport direction. Cementation is common, especially where interbedded tuffs are zeolitic. Cements are chiefly clinoptilolite and smectite, but opal-CT, gypsum, and calcite have also been recognized.

The framework constituents of sandstone consist of varying amounts of mineral grains, rock fragments, and vitroclastic particles. Sorting commonly is poor, and the clasts are generally angular to subrounded. The detrital mineral grains are quartz and feldspar with lesser amounts of biotite, muscovite, hornblende, zircon, and epidote. Feldspar and quartz are commonly present in nearly equal amounts, and sodic plagioclase is more abundant than alkali feldspar. Rock fragments are mostly volcanic and granitic. The tuffaceous component commonly makes up 20–60 percent of the framework and varies from fresh to completely altered.

Siltstone and Mudstone

Siltstone and mudstone are the predominant rocks in the Chalk Hills Formation. Both are medium to thick bedded, yellowish brown to grayish brown, and have an earthy luster. These fine-grained rocks break with a conchoidal to subconchoidal fracture and, where weathered, have a punky "popcorn" coating. Most mudstones contain, in addition to clay minerals, detrital sand grains, diatoms, vitroclastic particles, and authigenic minerals. Angular to subrounded detrital mineral grains commonly make up 10-40 percent of mudstones and are the same suite that is in the sandstones. Locally, diatoms and vitroclastic grains are represented by pseudomorphs of clinoptilolite. Authigenic minerals include clinoptilolite, opal-CT, potassium feldspar, gypsum, and calcite. Near-vertical joints in the fine-grained rocks are locally filled with gypsum or starkeyite $(MgSO_4 \cdot 4H_2O)$ (fig. 7).

Diatomite

Diatomite is a light-gray, fine-grained, lightweight, porous rock that breaks with a blocky to subconchoidal fracture. It is commonly medium to thick bedded and shows

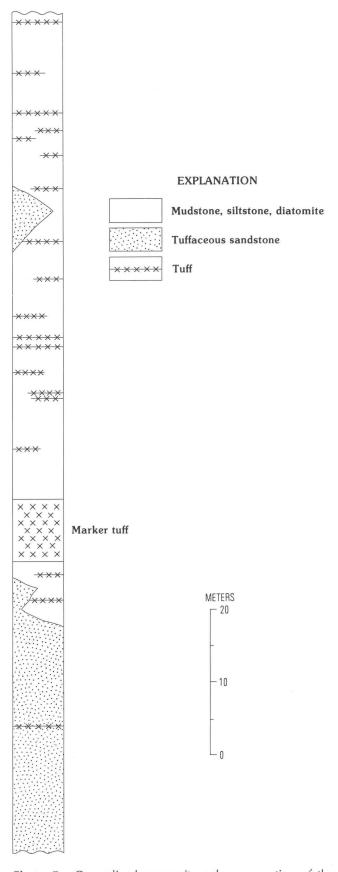


Figure 5. Generalized composite columnar section of the Chalk Hills Formation, Idaho.

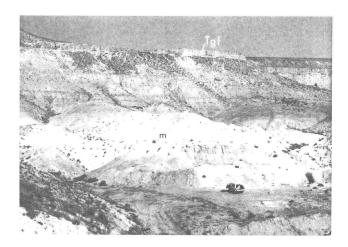


Figure 6. Excellent exposure of most of the Chalk Hills Formation (Tch) west of Castle Creek, about 9 km south of Oreana, Idaho. Formation consists chiefly of yellowish-brown and grayish-brown mudstone and siltstone. White unit in lower part of the formation is the zeolitic marker tuff (m) where it has been excavated for clinoptilolite. The basal oolite of the Glenns Ferry Formation (Tgf) unconformably overlies the Chalk Hills Formation.

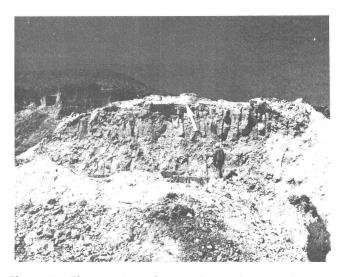


Figure 7. Closeup view of excavation in figure 6, showing contact (arrow) between the white, clinoptilolite-rich marker tuff and the underlying tuffaceous mudstone in the Chalk Hills Formation. Near-vertical joints in the mudstone are locally filled with gypsum or starkeyite. The cliff-forming basal oolite of the Glenns Ferry Formation can be seen in the background.

no sedimentary structures. In the field, diatomite superficially resembles altered fine-grained tuff. All the examined diatomites in the Chalk Hills Formation contain as much as 50 percent nondiatom constituents, including clay minerals, vitroclastic particles, and angular clastic grains of quartz and sodic plagioclase. Because of the amorphous character of the diatom frustules and the vitroclastic material, X-ray diffractometer patterns of diatomite tend to show less than the amount of these constituents in diatomite.

Tuff

Silicic, vitric tuffs in the Chalk Hills Formation make up about 10–12 percent of the stratigraphic section and are the most conspicuous and continuous strata. At least 20 tuffs were recognized, and they are about 1 cm to 13 m thick, although most are less than 0.5 m thick. The thicker tuffs are generally more continuous than the thinner ones. Thin tuffs are commonly single beds, but most tuffs that are thicker than 25 cm are multiple bedded (fig. 8). Individual beds of either single-bedded or multiple-bedded tuffs are commonly graded, being coarser at the base. Some thin tuffs and the lower part of thick tuffs are the result of ash falls directly into the lake. Other tuffs (particularly the upper part of thick tuffs) consist of reworked vitroclastic material mixed with epiclastic grains derived from highlands south of the depositional basin.

Two or more thin (less than 25 cm thick) basaltic tuffs are locally present in the upper part of the Chalk Hills Formation. The pattern of alteration of these basaltic tuffs is

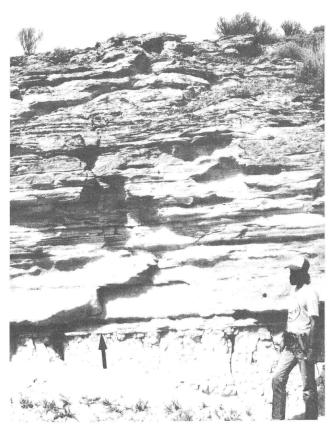


Figure 8. Natural exposure of lower part of the marker tuff east of Hart Creek, about 8 km southwest of Oreana, Idaho. The tuff is gray, friable, and multiple bedded. Silicic, vitric shards are unaltered. The contact (arrow) with underlying brown mudstone is sharp.

similar to that of the silicic ones, although the detailed diagenetic mineralogy differs. The alteration of the mafic tuffs will be the subject of another report and is not discussed further herein.

Fresh silicic tuff is light to dark gray and has a characteristic vitreous luster. It is very friable and easily disaggregated with the fingers. Natural exposures are poor; therefore, thin fresh tuffs could easily be overlooked. The lower contact of a tuff generally is sharp (fig. 8), whereas the upper contact is commonly gradational into the overlying rock, regardless of lithology. Ripple marks, laminations, and contorted bedding are present but rare.

The silicic tuffs are vitric but contain varying amounts of crystal and rock fragments, and, locally, diatoms. Most are fine to medium grained. The vitric particles consist of platy bubble-wall shards and pumice shards, but the platy bubble-wall type predominates. The index of refraction of unaltered shards ranges from 1.496 to 1.503. Angular crystal and rock fragments commonly make up 1–10 percent of the tuffs. Pyrogenic crystals consist of quartz, sodic plagioclase, sanidine, biotite, hornblende, zircon, and magnetite. The rock fragments are spherulitic and hyalopilitic lavas, ash-flow tuff, granite, and chert. Mineral grains of epiclastic origin are microcline, muscovite, and epidote.

Chemical analyses of unaltered shards separated from the marker tuff and a tuff stratigraphically above the marker tuff indicate a rhyolitic composition (table 1). Although the shards do not contain any authigenic minerals, the high (greater than 5 percent) $\rm H_2O$ content is evidence that even the freshest shards are hydrated. All other silicic tuffs are presumed to be rhyolitic on the basis of the low index of refraction of the glass shards and the coexisting pyrogenic minerals as described above.

DIAGENETIC ZONES

Distribution and Field Description

Throughout the extent of the Chalk Hills Formation, the tuffaceous rocks are unaltered or partly altered to smectite with or without opal-CT and are herein considered to be in the fresh-glass zone. South of Oreana, from Hart Creek southeastward to Birch Creek (fig. 3), a distance of about 21 km, some or all tuffaceous beds are altered to clinoptilolite and are considered to be in the zeolite zone, which is generally 1–3 km wide and roughly parallel to the southwestern margin of the Snake River Plain (fig. 9). The zeolite zone is gradational, interfingers into the fresh-glass zone, and pinches out to the northwest and southeast. Near marginal parts of the basin, such as along an eastern tributary to Castle Creek (fig. 10), the gradation from unaltered tuff to completely altered zeolitic tuff in a

Table 1. Chemical analyses of unaltered shards separated from tuffs of the Chalk Hills Formation, Owyhee County, Idaho

[Values are in percent. Chemical analyses by X-ray fluorescence, $\rm H_2O$ determined by loss on ignition at 900 °C. Analysts: J.E. Taggart, A.J. Bartel, and K.C. Stewart. <, less than]

Sample	1	2
SiO ₂	72.3	72.1
Al_2O_3	11.3	11.5
Fe ₂ O ₃	1.84	1.69
MgO	0.11	0.21
CaO	0.55	0.57
Na ₂ O	1.94	2.42
K ₂ O	5.54	5.58
TiO ₂	0.19	0.20
P ₂ O ₅	< 0.05	< 0.05
MnO	0.03	0.04
H ₂ O	5.93	5.37
Total	99.73	99.68

SAMPLE DESCRIPTIONS

- 1. Marker tuff, Hart Creek; NW1/4SE1/4sec. 4, T. 5 S., R. 1 W.; sample No. CC-46D.
- 2. Tuff in the upper part of the formation, between Browns and Castle Creeks; NW¹/₄SE¹/₄ sec. 18, T. 5 S., R. 1 E.; sample No. CC-32.

basinward direction is well exposed and has occurred over a distance of about 300 m. The zeolite zone is thickest between Browns Creek and Castle Creek (fig. 9) where the entire Chalk Hills Formation is zeolitic. To the northwest and southeast, the zeolite zone thins and is restricted to the marker tuff and (or) stratigraphically lower tuffaceous beds (fig. 11).

Tuffs of the fresh-glass zone are typically light gray and friable; shards have a distinct vitreous luster. Unlike tuffs of the fresh-glass zone, zeolitic tuffs are resistant and ledge forming. The zeolitic tuffs are white to light yellow or brown, relatively hard, and dull or earthy. Even those zeolitic parts of otherwise fresh tuffs stand out in a natural exposure. Original textures and sedimentary structures are generally preserved in the zeolitic tuffs. Tuffs of the zeolite zone break with a platy, hackly, or conchoidal fracture (fig. 12). Tuffs of the fresh-glass zone generally can be distinguished from altered tuffs in the field, but the zeolite content in altered tuff cannot be predicted without the aid of X-ray diffractometry. Altered tuff consisting mainly of smectite and opal-CT superficially resembles clinoptiloliterich tuff in the field.

Petrology of Tuffs in the Zeolite Zone

The mineralogy of the altered tuffs was determined mainly by study of X-ray diffractometer powder patterns of bulk samples (see appendix), supplemented by thin-section

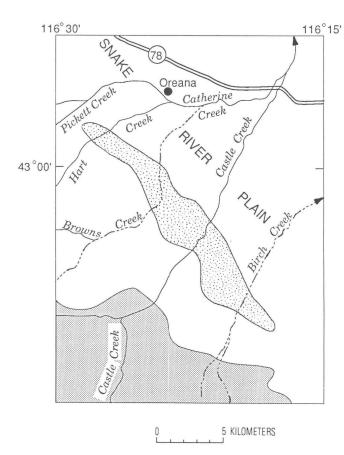


Figure 9. Southern part of the western Snake River Plain showing the distribution of the zeolite zone (stippled) in the Chalk Hills Formation, south of Oreana, Idaho. The unpatterned area is the fresh-glass zone. The shaded area is highlands along the southern margin of the Snake River Plain. These highlands are underlain chiefly by Cretaceous granitic rocks and Miocene volcanic rocks.

study and scanning electron microscopy. Thin sections and scanning electron microscopy were especially useful for determining the age relationships of the authigenic minerals, but these techniques were not used until the mineralogy was known by X-ray methods.

Silicic, vitric tuffs in the zeolite zone consist mainly of variable amounts of authigenic zeolites, smectite, and opal-CT. Clinoptilolite is by far the most abundant zeolite in the Chalk Hills Formation, but minor or trace amounts of erionite, mordenite, and phillipsite have been recognized locally. These diagenetic minerals locally coexist with relict glass. Near the boundary between the fresh-glass zone and the zeolite zone, the relict shards show obvious dissolution features, including pitting and corrosion (fig. 13). Even where no relict glass remains, the vitroclastic texture is generally preserved in the tuffs.

Typical pseudomorphs after bubble-wall shards consist of a thin (2–10 μ m, micrometer) marginal layer of smectite that is succeeded inwardly by crystals of one or more zeolites (fig. 14). The pseudomorphs may be solid or hollow, and commonly both types can be recognized in the



Figure 10. View north, along the eastern tributary of Castle Creek, of the lower part of the Chalk Hills Formation, Idaho, showing basinward gradation of fresh tuff (foreground) to zeolitic tuff (left of standing figure in middle ground). The fresh tuff in the foreground is gray and friable, but altered tuff left of the figure is light yellow and resistant.

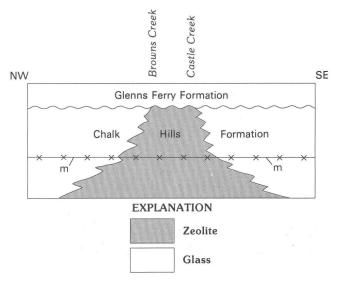


Figure 11. Diagrammatic northwest-southeast cross section showing the interfingering relationship between the fresh-glass zone and the zeolite zone in tuffaceous rocks of the Chalk Hills Formation, Idaho. Between Browns Creek and Castle Creek, the entire thickness of the formation is zeolitic, but to the northwest and southeast, the boundary between the zones is at the marker tuff (m) or stratigraphically lower beds. The Glenns Ferry Formation unconformably overlies both the fresh-glass zone and the zeolite zone. Tuffaceous rocks in the Glenns Ferry Formation are mostly unaltered and lack zeolites.

same specimen. Hollow ones are more common than solid ones and can be recognized even with a hand lens. The larger pseudomorphs generally are the hollow ones. The most common hollow pseudomorphs in tuffs of the Chalk Hills Formation consist of the marginal layer of smectite succeeded inwardly by a single layer of clinoptilolite

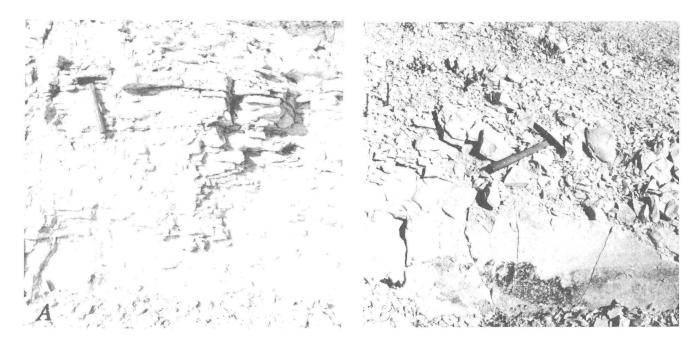


Figure 12. Clinoptilolite-rich marker tuff in exploration pits west of Castle Creek, Idaho. *A*, hackly to platy fracture in upper part of the tuff. *B*, conchoidal fracture in lower part of the tuff.

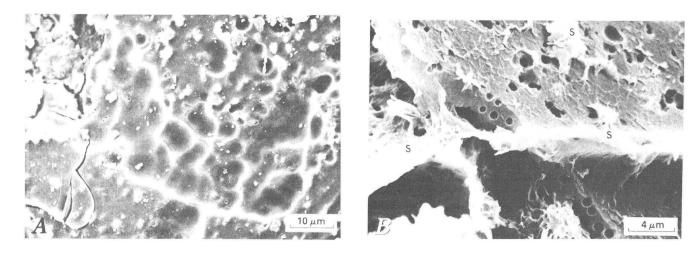


Figure 13. Scanning electron micrographs showing dissolution features of silicic, vitric shards in the marker tuff, Chalk Hills Formation, Idaho. *A*, corroded surface of platy shard. *B*, crenulated authigenic smectite (S) that grew on pitted shard.

crystals oriented quasi-perpendicular to the shard wall (fig. 15). Hollow pseudomorphs are convincing evidence that at least some of the clinoptilolite and coexisting zeolites crystallized in cavities from which the glass had already been dissolved.

DIAGENETIC MINERALS

Clinoptilolite

Clinoptilolite occurs in altered tuffs as well-formed platy or blocky crystals that are $1-30 \, \mu m$ long, but most are

 $1-15~\mu m$ long (figs. 16, 17). Although some tuffs contain only a trace of clinoptilolite and some tuffs are nearly monomineralic, most tuffs of the zeolite zone contain 40–90 percent clinoptilolite. The tuffs seem consistently rich in this zeolite between Browns Creek and Castle Creek (fig. 3). The marker tuff, in particular, is consistently rich in clinoptilolite and thick (generally 5–13 m thick) in this area.

X-ray fluorescence analyses of three pure separates of clinoptilolite from the marker tuff (table 2) show that the Si:(Al+Fe $^{3+}$) ratio is about 3.8–4.1 and that the exchangeable cations are variable. Additional energy-dispersive X-ray analyses of clinoptilolite from other tuffs in the Chalk Hills Formation have an average Si:Al ratio of

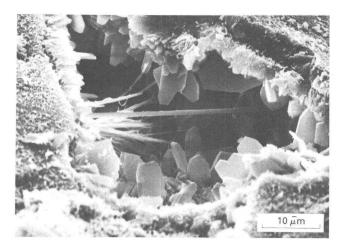


Figure 14. Scanning electron micrograph of shard pseudomorph consisting of a marginal layer of smectite (S) that is succeeded inwardly by platy clinoptilolite and then fibrous mordenite, Chalk Hills Formation, Idaho.

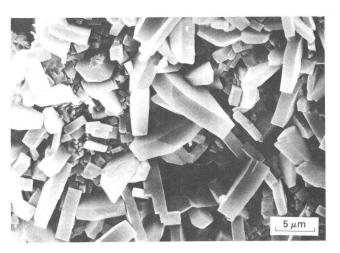


Figure 16. Scanning electron micrograph of well-formed platy to blocky clinoptilolite from the marker tuff, Chalk Hills Formation, Idaho.

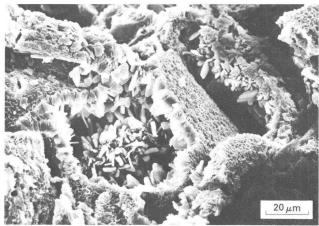
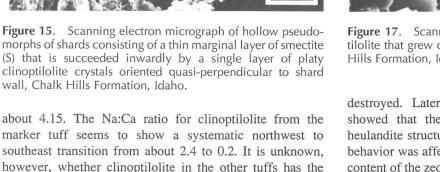


Figure 15. Scanning electron micrograph of hollow pseudomorphs of shards consisting of a thin marginal layer of smectite (S) that is succeeded inwardly by a single layer of platy clinoptilolite crystals oriented quasi-perpendicular to shard wall, Chalk Hills Formation, Idaho.



reflects postcrystallization cation exchange. Thermal stability has been used as a simple way to distinguish clinoptilolite from heulandite; both are members of the heulandite structural group. Mumpton (1960) suggested overnight heating in an oven at 450 °C, followed by examination by X-ray diffraction, as a means to differentiate the zeolites. Under these conditions, Mumpton found that the X-ray diffractometer pattern for clinoptilolite was essentially unaffected but that the pattern for heulandite was

same apparent transition and whether the present Na:Ca

ratio represents the ratio at the time of crystallization or



Figure 17. Scanning electron micrograph of blocky clinoptilolite that grew on star-shaped aggregates of opal-CT, Chalk Hills Formation, Idaho.

destroyed. Later workers (Alietti, 1972; Boles, 1972) showed that the thermal behavior of members of the heulandite structural group was more complex and that the behavior was affected by the Si:Al ratio and divalent cation content of the zeolite. Thus, Alietti (1972) proposed that the overnight heating test of Mumpton be increased to 550 °C. The three analyzed clinoptilolite samples of table 2 were heated in an oven at 550 °C for 16 hours. After cooling to room temperature, the samples were examined by X-ray diffraction. The diffractometer pattern for all samples persisted, but the peak intensities were diminished. For example, the (020) reflection showed about a 30-80 percent reduction in intensity after heating. The sample showing the most reduction is also the most calcic, and this thermal behavior is consistent with the findings of Alietti (1972) and Boles (1972).

Table 2. Chemical analyses and unit-cell contents of clinoptilolite separated from the marker tuff of the Chalk Hills Formation, Owyhee County, Idaho

[Chemical analyses by X-ray fluorescence; $\rm H_2O$ determined by loss on ignition at 900 °C. Analysts: J.E. Taggart, A.J. Bartel, and K.C. Stewart. <, less than . Unit-cell contents based on 72 oxygen atoms (Ti, P, and Mn were omitted in calculation of the unit cell)]

Sample	1	2	3
	Chemical anal	lyses	
SiO ₂	62.6	63.9	63.3
Al ₂ O ₃	13.4	12.3	12.8
Fe ₂ O ₃	0.70	1.37	0.91
MgO	1.04	1.02	1.21
CaO	1.96	2.39	4.08
Na ₂ O	2.59	1.32	0.36
K ₂ O	2.35	2.78	1.71
TiO ₂	0.38	0.19	0.17
P ₂ O ₅	0.06	< 0.05	< 0.05
MnO	< 0.02	< 0.02	< 0.02
H ₂ O	14.0	14.5	15.9
Total	99.08	99.77	100.44
	Unit-cell cont	ents	
Si	28.65	29.09	28.87
Al	7.22	6.60	6.88
Fe ³⁺	0.24	0.47	0.31
Mg	0.71	0.69	0.82
Ca	0.96	1.16	1.99
Na	2.30	1.16	0.32
K	1.37	1.61	1.00
H ₂ O	21.36	22.01	24.18
Si:(Al+Fe ³⁺)	3.84	4.11	4.02

SAMPLE DESCRIPTIONS

- 1. Between Hart and Browns Creeks; SE¹/₄SE¹/₄ sec. 3, T. 5 S., R. 1 W.; sample No. CC–42C.
- 2. Browns Creek; NE¹/₄NE¹/₄ sec. 24, T. 5 S., R. 1 W.; sample No. CC-10B.
- 3. Birch Creek; NW¼NE¼ sec. 15, T. 6 S., R. 1 E.; sample No. CC-21-5B.

Erionite

Trace to minor amounts of fibrous erionite occur with clinoptilolite at several localities in tuffs of the Chalk Hills Formation (see appendix). In addition to the erionite identified by X-ray diffraction, some was recognized by scanning electron microscopy in tuffs where the abundance is so low (less than 1 percent) that the erionite could not be detected by X-ray diffraction of the bulk samples. Most erionite in the tuffs of lacustrine deposits occurs as acicular or prismatic crystals or compact bundles of such crystals, but the erionite from the Chalk Hills Formation occurs as threadlike fibers that are 10–70 µm long and about 0.1–0.2 µm wide. The erionite fibers commonly occur in loose clusters (figs. 18, 19). Where associated with clinoptilolite,

the erionite generally formed later than the clinoptilolite. Energy-dispersive X-ray analyses show that the Si:Al ratio of the erionite is about 3.2–3.8, which is about the same range determined for erionite from a variety of lacustrine deposits throughout the world (Gude and Sheppard, 1981).

Mordenite

Thin fibers or filaments of presumed mordenite are commonly associated with clinoptilolite in the tuffs, but the abundance is so low that the mordenite cannot be recognized in X-ray diffractometer patterns of bulk samples. The morphology, size, and mode of occurrence are very similar to mordenite from a variety of clinoptilolite deposits examined by scanning electron microscopy by Mumpton and Ormsby (1976). Individual mordenite fibers from the Chalk Hills tuffs are 2-40 µm long and about 0.1–1.2 µm wide. The mordenite fibers commonly lie across clinoptilolite crystals or are interstitial to them (figs. 20, 21), indicating that the mordenite postdates the clinoptilolite. Energy-dispersive X-ray analyses yield Si:Al ratios of about 4.6–5.2 for the mordenite. This range of Si:Al ratios is well within the range of 4.2-5.9 that was determined for mordenites from a variety of localities and rock types by Passaglia (1975). The Chalk Hills mordenite cannot be distinguished from erionite solely on the basis of morphology. The combination of morphology and Si:Al ratios, however, gives credence to the identification in the absence of X-ray diffraction data.

Phillipsite

Phillipsite was recognized only at one locality (10, fig. 3), just east of Browns Creek, where it locally makes up as much as 70 percent of the basal centimeter of the

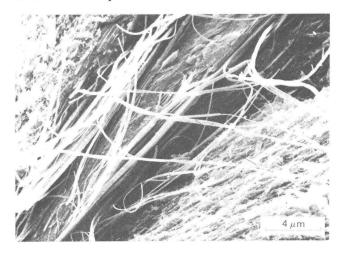


Figure 18. Scanning electron micrograph of a cluster of threadlike fibers of erionite, Chalk Hills Formation, Idaho.

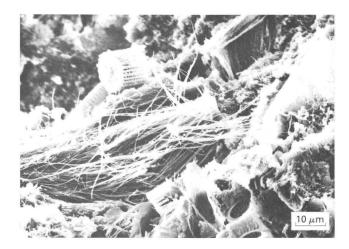


Figure 19. Scanning electron micrograph of a cluster of threadlike fibers of erionite that grew among corroded diatom tests in a tuff from the upper part of the Chalk Hills Formation, Idaho.

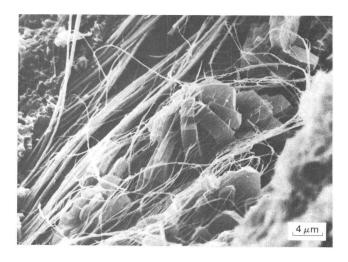


Figure 20. Scanning electron micrograph of mordenite fibers that are draped over blocky to platy crystals of clinoptilolite, Chalk Hills Formation, Idaho.

otherwise clinoptilolite-rich marker tuff. The phillipsite occurs as malformed length-slow spherulites that are 60– $300~\mu m$ in their longest dimension. The mean index of refraction is 1.475 and is consistent with a relatively siliceous and alkali-rich phillipsite (Galli and Loschi Ghittoni, 1972). This index is only slightly higher than that for a suite of phillipsites from silicic tuffs in saline, alkaline-lake deposits (Sheppard and Fitzpatrick, 1989). These saline, alkaline-lake phillipsites have Si:Al ratios of about 3.1–3.4; thus, the Si:Al ratio of the Chalk Hills phillipsite is probably only slightly lower than this range. Where phillipsite is associated with clinoptilolite and erionite in the basal marker tuff, the crystallization of phillipsite seems to have preceded that of the other zeolites.

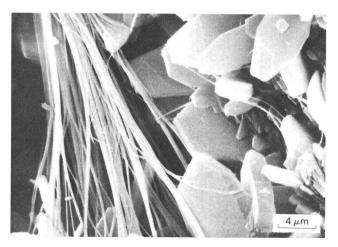


Figure 21. Scanning electron micrograph of mordenite fibers that are interstitial to and draped across platy crystals of clinoptilolite, Chalk Hills Formation, Idaho.

Smectite

Smectite is a nearly ubiquitous authigenic mineral in the tuffaceous rocks of the Chalk Hills Formation. The smectite content in completely altered tuffs of the zeolite zone commonly ranges from a trace to about 30 percent. Even in the fresh-glass zone, smectite is locally common and, at some localities, is associated with opal-CT (see appendix). Where smectite is the major constituent of a tuff, a punky "popcorn" coating is typical of weathered natural exposures. Scanning electron micrographs of smectite show the characteristic crenulated to flaky morphology (fig. 22). The smectite has a first-order basal spacing of about 14.0 Å for oriented, air-dried samples and of 17.0-17.3 Å for oriented, glycolated samples. The (060) reflection is 1.49–1.50 Å and is typical for dioctrahedral smectite (montmorillonite). Where associated with other authigenic silicate minerals in the tuffs, the smectite is the earliest mineral to have crystallized.

Opal-CT

Opal-CT is difficult to recognize in thin sections of altered tuffs because of its isotropic and finely crystalline character. Most identifications of opal-CT in the Chalk Hills Formation are based on X-ray diffractometer data or scanning electron microscopy. The opal-CT has relatively broad diffractometer peaks at the following d spacings: 4.23–4.29 Å, 4.07–4.09 Å, and 2.49–2.50 Å. The peak near 4 Å has, by far, the strongest intensity. Scanning electron micrographs show that opal-CT occurs as clusters of minute (less than 1 μ m) crystals. The morphology of the clusters is commonly ragged blades (fig. 23), lepispheres, mammillary coatings (fig. 24), or star-shaped aggregates (fig. 17). The abundance of opal-CT in the tuffs ranges from a trace to

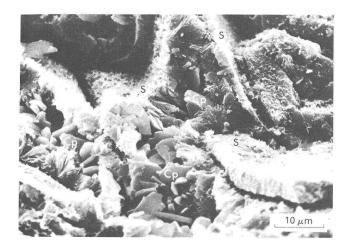


Figure 22. Scanning electron micrograph of pseudomorph of a shard that consists of marginal smectite (S) followed inwardly by platy clinoptilolite (Cp), Chalk Hills Formation, Idaho. The smectite shows the typical crenulated texture.

nearly 100 percent, but most tuffs contain less than 30 percent. Where associated with smectite and (or) clinoptilolite, opal-CT crystallized after smectite and either before or after clinoptilolite.

Quartz

Authigenic quartz is relatively rare in the altered tuffs of the Chalk Hills Formation. Most of the quartz identified by X-ray diffraction of bulk samples (see appendix) is epiclastic or pyrogenic. The authigenic quartz occurs in irregular patches of nearly equidimensional, minute (less than 2 μm in diameter) anhedral grains or of aggregates of fibers that commonly are 10–30 μm long. Both types of

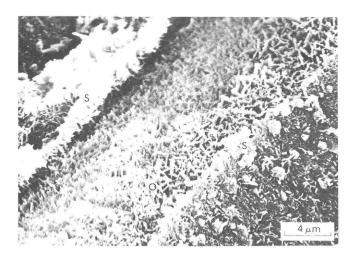


Figure 23. Scanning electron micrograph of a pseudomorph of a shard showing marginal smectite (S) followed inwardly by ragged blades of opal-CT (O), Chalk Hills Formation, Idaho.

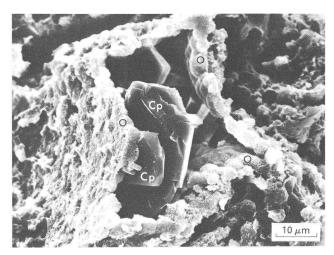


Figure 24. Scanning electron micrograph of a pseudomorph of a shard showing marginal mammillary opal-CT (O) followed inwardly by blocky clinoptilolite (Cp), Chalk Hills Formation, Idaho.

quartz have wavy extinction. The patches of quartz have replaced authigenic smectite and clinoptilolite and commonly have obliterated the vitroclastic texture.

Potassium Feldspar

Potassium feldspar of probable authigenic origin occurs in tuff and tuffaceous siltstone and mudstone of the Chalk Hills Formation. The feldspar was identified on the basis of its distinctive X-ray diffractometer pattern (Sheppard and Gude, 1969) in bulk samples where it makes up as much as 20 percent of the rock. The relationship of this authigenic potassium feldspar to other diagenetic minerals is unknown because the feldspar was not recognized in thin sections or in scanning electron micrographs. X-ray diffractometry (see appendix) showed that the feldspar occurs from Browns Creek to Birch Creek but is most common between Browns Creek and Castle Creek (fig. 3). Studies of authigenic potassium feldspar in lacustrine tuffs from other deposits generally indicate that the feldspar is finely crystalline (less than 10 µm in size), monoclinic, and pure or nearly pure potassium end member (Sheppard and Gude, 1969). Such authigenic feldspar crystallized after the coexisting authigenic zeolites and commonly from the precursor zeolites.

Other Authigenic Minerals

Zeolitic tuffaceous rocks in the Chalk Hills Formation locally contain authigenic calcite or gypsum. Thin-section and scanning electron microscopy studies show that these constituents postdate the zeolites. Luster mottling by calcite is locally common in the lower part of the zeolitic marker

tuff. The calcite crystals are as much as 8 cm long, and they have replaced authigenic smectite, clinoptilolite, and opal-CT. Gypsum, in trace amounts, was recognized by X-ray diffraction in certain tuffs and tuffaceous mudstones. Gypsum also occurs locally in zeolitic tuff as small spherical concretions (as much as 2 cm in diameter) and as veins of fibrous satin spar.

PARAGENESIS

The relative age relationships of diagenetic minerals in the tuffaceous rocks of the Chalk Hills Formation were determined by the sequence of filling of voids and oncehollow pseudomorphs of vitric particles and by replacement. The mineral in the interior of a void or pseudomorph presumably formed later than the mineral(s) nearer the periphery. The following sequences of crystallization were compiled from thin-section and scanning electron microscopy examination (the earliest mineral is listed first):

Smectite—clinoptilolite

Smectite—clinoptilolite—mordenite

Smectite—clinoptilolite—opal-CT

Smectite—clinoptilolite—calcite

Smectite—clinoptilolite—quartz—calcite

Smectite—phillipsite—clinoptilolite±erionite

Smectite-erionite

Smectite—clinoptilolite—erionite

Smectite—clinoptilolite—quartz

Opal-CT—clinoptilolite

Smectite—opal-CT—clinoptilolite

The first three sequences listed above are the common ones observed in tuffs of the zeolite zone. Smectite consistently crystallized prior to any zeolite. At the only occurrence of phillipsite in the Chalk Hills Formation, the phillipsite formed earlier than the coexisting zeolites. Except for this phillipsite occurrence, clinoptilolite is the zeolite that generally succeeded smectite. Mordenite and erionite postdated clinoptilolite, but the paragenetic relationship between mordenite and erionite is unknown because they have not been observed to coexist in the tuffaceous rocks studied. Erionite rarely crystallized on clinoptilolite crystals that had been affected by partial dissolution (fig. 25). Both quartz and calcite are late diagenetic minerals and locally have replaced smectite, clinoptilolite, and opal-CT. Calcite seems to have been the latest mineral to form because it locally has replaced authigenic quartz.

GENESIS OF DIAGENETIC SILICATE MINERALS

Factors that may control the formation and distribution of zeolites and other diagenetic silicate minerals

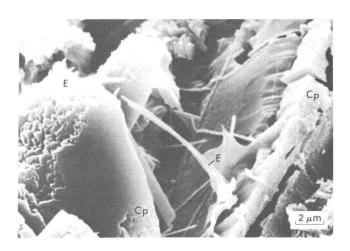


Figure 25. Scanning electron micrograph of fibrous erionite (E) that grew on corroded clinoptilolite (Cp), Chalk Hills Formation, Idaho. Energy-dispersive X-ray analyses indicated that the Si:Al ratios of clinoptilolite and erionite are 4.1–4.2 and 3.2–3.8, respectively.

in sedimentary rocks were summarized by Hay (1966, 1986) as composition, permeability, and age of the host rock, and metastable crystallization, temperature, pressure, and chemistry of the pore water. Inasmuch as all the diagenetic silicate minerals in the Chalk Hills Formation occur within a given tuff, differences in the composition, permeability, and age of the host rock cannot explain their distribution pattern. The Chalk Hills Formation was subjected to only shallow burial; therefore, the temperature and pressure during diagenesis were relatively low. Besides, the difference, if any, in the depth of burial between the fresh-glass zone and the zeolite zone was seemingly not great enough to cause the observed pattern. The present diagenetic silicate mineralogy of the tuffs and the pattern of distribution of the diagenetic zones probably reflect differences in the chemistry of pore water during diagenesis.

Experimental work by others and theoretical considerations indicate that the activity ratio of alkali ions to hydrogen ions and the activity of silica are the major chemical parameters of the pore water that control whether clay minerals or zeolites will crystallize under those conditions approximating surface temperatures and pressures (Hemley, 1959; Garrels and Christ, 1965; Hess, 1966, Barth-Wirsching and Höller, 1989). Zeolites are favored over clay minerals by relatively high alkali ion to hydrogen ion activity ratios and by relatively high silica activities. Such conditions in nature are especially common in the depositional environment of a saline, alkaline lake (Surdam and Sheppard, 1978).

The zeolites and associated authigenic silicate minerals in the tuffaceous rocks of the lacustrine Chalk Hills Formation undoubtedly formed during diagenesis by hydrolysis and dissolution of silicic glass by interstitial water. The pattern of alteration and the lateral gradation of unaltered (fresh) glass to an assemblage of smectite,

clinoptilolite, and opal-CT strongly suggest that the interstitial water causing the alteration was the connate water trapped in the tuffaceous rocks during lacustrine deposition and that the lake water was laterally chemically zoned.

Although regional studies by Kimmel (1979) and sedimentologic studies by Middleton and others (1985) indicate that the Chalk Hills Formation and the overlying Glenns Ferry Formation were deposited in large, essentially freshwater lakes, the lake water coinciding with the distribution of the zeolite zone in the Chalk Hills Formation was probably characterized by low to moderate salinity and a pH of 7–9. Zeolites have not been recognized in the overlying Glenns Ferry Formation where freshwater conditions remained throughout that depositional interval. Neither evaporite minerals or molds of evaporite minerals, indicative of high salinities, nor Magadi-type chert, indicative of high salinities and high pH (9–11), have been recognized anywhere in the Chalk Hills Formation.

Diatomites and diatomaceous sediments are common in the fresh-glass zone of the Chalk Hills Formation but are absent or rare in the zeolite zone. Mudstone, siltstone, and tuff from the zeolite zone are either barren of diatoms or contain highly corroded tests. Zeolitic tuffaceous rocks locally contain diatoms that have been replaced by clinoptilolite. Diatom-bearing mudstone was collected from the fresh-glass zone near the zeolite zone, and its presence there suggests deposition in lake water that was essentially fresh but slightly saline and alkaline (J.P. Bradbury, written commun., 1986).

Ostracodes provide information on the chemical composition of the ancient lakes where they lived (Forester, 1986). Clinoptilolite-bearing tuffaceous sandstone from one locality (NE½SW½ sec. 33, T. 5 S., R. 1 E.) in the zeolite zone of the Chalk Hills Formation contains a diverse assemblage of ostracode species that suggest a lake-water salinity greater than 300 mg/L (milligrams per liter) but less than 3 g/L (grams per liter) and a pH of about 8–9 (R.M. Forester, written commun., 1988).

Thus, the indirect evidence from the Chalk Hills Formation suggests that the lake water trapped in the tuffaceous rocks that now contain clinoptilolite was of low to moderate salinity and alkalinity.

Formation of Zeolites from Silicic Glass

Solution of silicic glass by the lake water trapped during deposition of the tuffaceous rocks provided the materials necessary for the crystallization of zeolites. The initial alteration of rhyolitic glass in the tuffaceous rocks by hydrolysis to form the early smectite probably raised the pH, activity of SiO₂, and (Na⁺+K⁺):H⁺ activity ratio of the pore water to levels where clinoptilolite crystallized. The increased pH and salinity of the pore water also caused

continual and rapid dissolution of the vitric material (Mariner and Surdam, 1970). Assuming a relatively closed system during the alteration, the crystallization of smectite and clinoptilolite resulted in an excess of SiO₂ that crystallized chiefly as opal-CT. Dissolution of diatoms also provided additional SiO₂ to the interstitial fluid.

During the later stages of diagenesis, slight changes in the pH and cation ratios of the pore water probably were responsible for the local minor to trace amounts of mordenite and erionite that crystallized after clinoptilolite. Experimental work by Mariner and Surdam (1970) indicated that the Si:Al ratio of the zeolite is controlled by the Si:Al ratio of the solution from which the zeolite crystallized. Their experiments showed that the Si:Al ratio of the solution decreased as the pH increased. Thus, relatively aluminous zeolites, such as erionite and phillipsite, are favored over siliceous zeolites, such as clinoptilolite and mordenite, as the pH is increased. Relatively high Na:K ratios favor mordenite over clinoptilolite (Hawkins, 1981; Barth-Wirsching and Höller, 1989). As scanning electron micrographs (fig. 25) have shown, clinoptilolite in tuff of the Chalk Hills Formation was only rarely corroded prior to the crystallization of erionite. Although mordenite also postdated the clinoptilolite in the tuffs, there is no evidence of dissolution of clinoptilolite prior to crystallization of mordenite.

Chemical Changes During Zeolitic Diagenesis

Chemical analyses of bulk samples of clinoptiloliterich marker tuff from the Chalk Hills Formation are given in table 3, along with a chemical analysis of an unaltered (fresh) bulk sample from the same tuff in the fresh-glass zone. In addition to the obvious and significant increase in the H₂O content, other approximate changes in chemical composition from fresh glass to zeolitic tuff were inferred by assuming that the Al₂O₃ content remained constant during diagenetic alteration, although this assumption has not been rigorously tested. Gains and losses of chemical components for the clinoptilolite-rich tuffs given in table 3 were determined by (1) recalculation of all the analyses to the anhydrous form and normalizing to 100 percent, (2) then, recalculation of these normalized analyses of the clinoptilolite-rich tuffs, assuming constant Al₂O₃ content of the fresh glass, and (3) finally, comparison of the Al₂O₃constant analyses with the anhydrous analysis of the fresh glass.

The clinoptilolite-rich tuffs differ significantly in composition from the fresh glass. Relative to the unaltered glass, all the clinoptilolite-rich tuffs show consistent gains of MgO and CaO and losses of ${\rm SiO_2}$ and ${\rm K_2O}$. Two of the three zeolitic tuffs also show losses of ${\rm Fe_2O_3}$ and ${\rm Na_2O}$. The most obvious chemical changes in the clinoptilolite-rich

Table 3. Chemical analyses and chemical comparison of unaltered and clinoptilolite-rich marker tuff from the Chalk Hills Formation, Owyhee County, Idaho

 $[H_2O$ determined by loss on ignition at 900 °C. A, uncorrected analysis in weight percent; B, analysis recalculated to anhydrous and normalized to 100 percent; C, gains (+) and losses (-) in weight percent assuming the constant Al_2O_3 value of the unaltered tuff (analysis 1B). Analyses by X-ray fluorescence by A.J. Bartel, K.C. Stewart, and J.E. Taggart]

Sample		1		2			3		4			
	A	В	A	В	С	A	В	С	A	В	С	
SiO ₂	71.7	76.90	62.8	73.72	-18.70	62.4	73.29	-14.83	62.0	74.33	-17.64	
Al_2O_3	11.4	12.23	13.2	15.49		12.3	14.44		12.8	15.34		
Fe ₂ O ₃	1.87	2.00	0.66	0.77	-1.39	2.77	3.25	+0.75	0.84	1.01	-1.20	
MgO	0.12	0.13	1.01	1.18	+0.80	1.37	1.61	+1.23	1.26	1.51	+1.07	
CaO	0.55	0.59	1.60	1.88	+0.89	3.36	3.94	+2.75	4.01	4.81	+3.24	
Na ₂ O	1.92	2.06	3.11	3.65	+0.82	1.70	2.00	-0.37	0.61	0.73	-1.48	
K ₂ O	5.46	5.86	2.36	2.77	-3.67	0.98	1.15	-4.89	1.70	2.04	-4.23	
TiO ₂	0.19	0.20	0.46	0.54	+0.23	0.21	0.25	+0.01	0.19	0.23	-0.02	
MnO	0.03	0.03	< 0.02		-0.03	0.06	0.07	+0.03	< 0.02		-0.03	
H ₂ O	6.37		14.4			14.9			15.7			
Total	99.61	100.00	99.60	100.00		100.05	100.00		99.11	100.00		

SAMPLE DESCRIPTIONS

- 1. Unaltered tuff, Hart Creek; NW1/4SE1/4 sec. 4, T. 5 S., R. 1 W.; sample No. CC-46D.
- 2. Clinoptilolite-rich tuff, between Hart and Browns Creeks; SE1/4SE1/4 sec. 3, T. 5 S., R. 1 W.; sample No. CC-42C.
- 3. Clinoptilolite-rich tuff, Castle Creek; SW1/4SE1/4 sec. 32, T. 5 S., R. 1 E.; sample No. CC-37A.
- 4. Clinoptilolite-rich tuff, Birch Creek; NW¼NE¼ sec. 15, T. 6 S., R. 1 E.; sample No. CC-21-5B.

tuffs are the gains of alkaline earth elements and the losses of alkalis and silica. Similar gains and losses have been noted for the diagenetic alteration of silicic glass to mainly clinoptilolite in the John Day Formation (middle Tertiary) of Oregon (Hay, 1963) and the Ricardo Formation (Miocene and Pliocene) of California (Sheppard and Gude, 1965).

Age of Zeolitic Diagenesis

Zeolites can form relatively rapidly from silicic vitric material in a saline, alkaline lake. Their formation from rhyolitic glass probably is facilitated by the rapid rate of solution and high solubility of the glass at a pH above 9 (Surdam and Sheppard, 1978). Rhyolitic tuffs near the surface of Teels Marsh, a saline, alkaline playa in western Nevada, are altered to zeolites. Studies of Teels Marsh by Taylor and Surdam (1981) indicated that alteration of the uppermost tuff at a depth of about 30 cm took 1,000 years or less. In the highly saline, alkaline diagenetic environments of Searles Lake, California, silicic ash altered to zeolites in less than 9,000 years (Hay and Guldman, 1987).

The age of the zeolitic alteration of tuffs in the Chalk Hills Formation is unknown, but the zeolitization probably was not as rapid as that given for the above lacustrine deposits because both the salinity and pH of the pore water in the zeolite zone of the Chalk Hills Formation were lower than that in these examples. Nevertheless, indirect evidence suggests that zeolitization of the Chalk Hills tuffs was completed prior to deposition of the unconformably over-

lying Glenns Ferry Formation. Some of the calcareous ooids in the basal oolite of the Glenns Ferry Formation contain nuclei of subrounded, clinoptilolite-rich tuff. Although the source of the zeolitic nuclei is uncertain, a probable source is zeolitic tuff in the Chalk Hills Formation. Inasmuch as the unconformity separating the two formations represents more than a million years (Swirydczuk and others, 1982), ample time was available for zeolitization, tilting, and erosion of the marginal part of the Chalk Hills Formation prior to deposition of the Glenns Ferry Formation.

CONCLUSIONS

A significant deposit of clinoptilolite, having economic potential, formed during diagenesis of rhyolitic, vitric tuffs in the lacustrine Miocene Chalk Hills Formation of southwestern Idaho. The marker tuff, in particular, is 5–13 m thick between Browns and Castle Creeks and generally consists of 70 to nearly 100 percent clinoptilolite. Authigenic smectite and opal-CT commonly coexist with the clinoptilolite. About 35 million short tons of clinoptilolite-rich marker tuff are still preserved between Hart and Birch Creeks.

The salinity and pH of the Chalk Hills lake water were probably lower than that of typical Cenozoic saline, alkaline-lake deposits of the western United States (Surdam and Sheppard, 1978). As a result, abundant clinoptilolite formed rather than chabazite, erionite, phillipsite, and (or) analcime. Experimental work by Mariner and Surdam

(1970) and Barth-Wirsching and Holler (1989) showed that a relatively low alkalinity (pH=7–9) should favor the crystallization of clinoptilolite rather than a zeolite of lower Si:Al ratio, such as phillipsite. The assemblage of clinoptilolite and associated minerals in the Chalk Hills tuffs and their inferred origin are similar to the alteration recently described by Hay and Guldman (1987) for vitric, silicic tuffs in the lower part of Searles Lake, California.

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APPENDIX

Mineralogic composition of tuffaceous rocks of the Chalk Hills Formation, Owyhee County, Idaho, as estimated from X–ray diffractometer patterns of bulk samples

[---, looked for but not found; Tr, trace. Mica includes pyrogenic biotite, detrital muscovite and biotite, and authigenic illite. Quartz includes pyrogenic, detrital, and authigenic varieties. Other: chiefly hornblende; sample CC-10A contains 7 parts phillipsite and a trace of erionite; samples CC-10-3D, CC-13A, CC-13B, and CC-58C contain a trace of erionite; sample CC-33J contains one part erionite; sample CC-60B contains 5 parts apatite. Sample location column shows position of sample in meters above (+) or below (-) base of marker tuff (M); U, above marker tuff but exact stratigraphic position unknown; L, below marker tuff but exact stratigraphic position unknown; Uk, stratigraphic position unknown; D, diatomite; Ms, mudstone; Ss, sandstone; St, siltstone, T, tuff. X-ray analyses in parts of 10]

Locality No. (fig. 3)	Sample No.	Sample location	Glass	Mica	Smectite	Clinop- tilolite	Potassium feldspar	Quartz	Opal- CT	Plagio- clase	Calcite	Gypsum	Other
1	CC-1A	–3.05, Ms		Tr	3	4	Tr			1			
	-1D	+0.01, M				3						7	
	-1E	+2.44, M				8			2				
	-1F	+5.26, M			1	9							
	-1G	+8.76, M				10						Tr	
	-1H	+10.82, M			1	9							
	-1N	+15.36, T			1	9							
	-1P	+22.38, T		Tr	1	3	1	1	4				
	-1I	+26.06, St		1	2	4	2	1				Tr	
	-1Q	+29.30, T			1	9							
	-1R	+30.58, T			1	9							
	-1S	+37.70, Ms		Tr	2	3	2		2		Tr	1	
	-1T	+46.10, Ss		1	1	3		2		3			
	−1 U	+61.22, T	10							Tr			
	-1 V	+66.26, T		Tr	Tr	3	2		5				
	-1 W	+73.82, T		Tr	2	3		3	2	Tr			
2	CC-55D	M, lower part		Tr	5				5				
	-55F	M, middle part		Tr	9				1				
	-55E	M, upper part			4	2			4				
3	CC-3A	L, T		Tr	1	3	1	5		Tr			
	–3B	L, Ss		Tr	3	4		2		1			
4	CC-4A	L, T			2	3		5		Tr			
	–4B	L, T			1	3		6		Tr			
	-56B	L, Ss		Tr	1	4		4		1			
5	CC-5	M, upper part		Tr	Tr	6			4				
6	CC-6A	L, Ss		Tr	Tr	4		4		2			
	-6B	L, T				3		6		1			
7	CC-7A	L, Ss		1	Tr	5		2	2	Tr			
	-7B	L, T				Tr			9		1		

Locality No. (fig. 3)	Sample No.	Sample location	Glass	Mica	Smectite	Clinop- tilolite	Potassium feldspar	Quartz	Opal- CT	Plagio- clase	Calcite	Gypsum	Other
8	CC-8A	M, lower part			Tr	10							
	8B	M, lower part			Tr	2					8		
	-8C	M, upper part			Tr	4		5			1		
9	CC-9A	M, lower part				4		2	3	1			
	-9B	M, middle part	8		2								
	–9C	M, middle part	10										
	–9D	M, upper part	2		2	3		2		1			
10	CC-10-1	–0.01, Ms		Tr	2	5	1	Tr	2	Tr			
	-10A	+0.01, M			1	2							7
	-10-2B	+0.03, M			${ m Tr}$	9						1	
	-10-2C	+0.06, M			Tr	10							
	-10B	+1.00, M			Tr	10							
	-10C	+2.75, M			Tr	10		Tr					
	-10D	+6.65, M			2	6			2				
	-10-3A	+14.95, T		Tr	1	4	1		4				
	-10-3B	+21.72, T		Tr	1	4	1		4				
	-10-3C	+23.36, T			Tr	4	1		5	Tr			
	-10-3D	+27.00, T			7	2				1			Tr
	-10-3E	+33.02, T			5	3				2		Tr	
	-10-4	+38.07, St		Tr	1	5	2		1	1			
	-10-5	+46.23, Ss		1	1	3		2	Tr	3		Tr	
	-10-6A	+58.27, St		1	Tr	6		2		1			
	-10-6B	+61.12, T			Tr	10							
11	CC-11A	L, Ss		Tr	Tr	6		2		2			
	-11B	L, T				Tr			9	1			
	-11C	L, T			Tr	9				1			
12	CC-12A	-0.30, St	2		6			1		1			
	-12B	+0.02, M	5		5								
	-12C	+5.00, M	7		2					1			
	-12D	+9.40, Ss	6	Tr	3					1			
	-12F	+16.50, Ss	6		3			Tr		1			
	-12G	+25.00, T	7		2				1				
	-12H	+26.00, T	7		2					1			
	-12I	+26.30, T	7	1	Tr				1	1			
	-12J	+27.00, T	6	1	2					1			
	-12K	+27.00, T	6		3					1			
	-12L	+28.00, T	5		2			2		1			

Mineralogic composition of tuffaceous rocks of the Chalk Hills Formation, Owyhee County, Idaho, as estimated from X–ray diffractometer patterns of bulk samples—Continued

Locality No. (fig. 3)	Sample No.	Sample location	Glass	Mica	Smectite	Clinop- tilolite	Potas- sium feldspar	Quartz	Opal- CT	Plagio- clase	Calcite	Gypsum	Other
13	CC-13A	U, T, lower part			4	5				1			Tr
	-13B	U, T, upper part			1	7				1		1	Tr
14	CC-14	L, T		Tr	Tr	4		1	5	Tr			
15	CC-15	L, Ss		Tr	Tr	6		2		2		Tr	
16	CC-16	L, T		Tr	Tr	7		2		1		Tr	
17	CC-17-2	U, T	6		2			Tr	1	1			
	-17-3A	U, T	7		2					Tr		1	
	-17-3B	U, Ms	2	1	4			1		1		1	
	-17-3C	U, T, lower part	1		1					2		6	Tr
	-17-3D	U, T, middle part	8							2	Tr		Tr
	-17-3E	U,T, upper part	8		Tr				1	1			
	-17-3F	U, T	10		Tr					Tr			
	-17-3G	U, Ms		1	4			1	2	2		Tr	
	-17-4A	U, Ms		1	4			2	1	2			
	-17-4B	U, T	9		1								
18	CC-18A	–0.15,Ms		Tr	2	3			4	1		Tr	
	-18B	+0.02, M			1	8						1	
	-18C	+0.30, M		Tr	Tr	8			2			Tr	
	-18D	+0.70, M			Tr	8			2				
19	CC-19A	L, T							10		Tr		
	-19B	L, T				Tr			10				
20	CC-20D	–0.50,Ms		Tr	1	5	2	1	Tr	1			
	-20A	+0.02, M		Tr	3	7							
	-20B	+1.00, M			Tr	10							
	-20C	+1.70, M			Tr	7			3				
21	CC-21-1	-1.27, Ss		Tr	Tr	2		4		4			
	-21-2	-0.74, T			1	5		2		2			
	-21-3	-0.36,Ms		Tr	1	6	2	1					
	-21-4	-0.10, Ss				2		3		3	2		
	-21-5A	+0.02, M				6					4		
	-21-5B	+0.30, M			Tr	10							
	-21-5C	+1.00, M			Tr	10							
	-21-5D	+2.00, M			Tr	9			1				
	-21-5E	+3.53, M		Tr	1	4	Tr		5				
	-21-7	+7.15, T		Tr	1	4		1	4	Tr			
	-21-1	T7.13, 1		11	1	7		1	7	11			

Locality No. (fig. 3)	Sample No.	Sample location	Glass	Mica	Smectite	Clinop- tilolite	Potas- sium feldspar	Quartz	Opal- CT	Plagio- clase	Calcite	Gypsum	Other
	-21-8	+14.44, Ms		Tr	5			2	1	2		***	
	-21-9	+15.74, T	7	Tr	1					2			
	-21-10A	+19.90, T	6	Tr	2			1		1			
	-21-10B	+38.20, T	5		3			1		1			
	−21−10 C	+43.10, T	6	Tr	2			1	1	Tr			
22	CC-22	U, T		Tr	1	4		Tr	5	Tr			
23	CC-23-1	-14.37, Ss		Tr	1	3		2	2	2			
	-23-2	-13.85, T		1	Tr	4			5				
	-23-3	-9.57, Ms		Tr	4	3		1	1	1		Tr	
	-23-4A	–5.17, T		Tr		3	2		5	Tr			
	-23-4B	-4.61, Ss		Tr		3		4		3			
	-23-5	-3.37, Ss			Tr	1		5		4			
	-23-6A	-1.72, T			1	8				1			
	-23-6B	-0.20, Ms		Tr	1	4	2	1	1	1		Tr	
25	CC-25A	Uk, T			Tr	1			9				
	-25B	Uk, T				1		1	6	Tr	2		
26	CC-26A	-0.04, Ms			2	2		1	4	1			
	-26B	+0.02, M		Tr	5	5							
	-26C	+0.17, M		Tr	2	8							
	-26D	+1.12, M		Tr	2	4			4			Tr	
	-26E	+2.50, M		Tr	1	6			3			Tr	
27	CC-27	Uk, T		Tr	2	5	1	1		1			
28	CC-28	U, T	6		3					1			
29	CC-29	Uk, T	6	Tr	2			1		1			
30	CC-30	Uk, T	8	Tr	1			Tr		1			
31	CC-31A	L, T	5	Tr	5			Tr		Tr			
	-31B	L, T	6	Tr	2			1		1			
32	CC-32	U, T	10										
33	CC-33A	U, Ms	4	Tr	4			1		1		Tr	
-	-33B	U, Ms	4	Tr	5			Tr		1			
	-33C	U, T	7	Tr	1			Tr		1		1	

Mineralogic composition of tuffaceous rocks of the Chalk Hills Formation, Owyhee County, Idaho, as estimated from X-ray diffractometer patterns of bulk samples—Continued

Locality No. (fig. 3)	Sample No.	Sample location	Glass	Mica	Smectite	Clinop- tilolite	Potas- sium feldspar	Quartz	Opal- CT	Plagio- clase	Calcite	Gypsum	Other
	-33D	U, T	10							Tr			
	-33E	U, T	8		1			Tr		1		Tr	Tr
	-33F	U, T	10										
	-33G	U, T	8		1			Tr		1			
	-33H	U, Ms	2	1	5			1		1			
	-33I	U, T, lower part	2	Tr	5			1		2			
	-33J	U, T, upper part			7			1		1			1
	-33K	U, Ss	Tr	i	2			4		3			Tr
34	CC-60A	U, Ms		Tr	6			3		Tr	1		
	-60B	U, Ms		Tr	5								5
35	CC-35A	U, Ms	1	Tr	6			2		1			
	-35B	U, T	9	Tr	Tr					1			
36	CC-36A	L, T			1	9				Tr			
	-36B	L, Ss		Tr	Tr	4		3		3			
	-36C	L, Ss		1	Tr	5		2		2			
37	CC-37A	+0.20, M			1	9					Tr		
	-37B	+3.25, M			Tr	6			4				
	-37C	+5.90, M			Tr	6			4				
38	CC-38	M, middle part			1	9					Tr		
40	CC-40A	–0.20, Ms		Tr	2	1	1	1	5	Tr			
	-40B	+0.15, M			4	2			4				
	-40C	+0.80, M	5	Tr	5								
	-40D	+1.68, M	5		5					Tr			
	-40E	+7.50, St		Tr	Tr	4			5	1			
	-40F	+10.00, Ms		Tr	1	1		1	7	Tr			
41	CC-41A	M, lower part		Tr	1	4			5				
	41B	M, upper part			4	2			4	Tr			
42	CC-42A	+0.02, M		Tr	3	3		2		2			
	-42B	+0.80, M		Tr	2	2		3		2		1	
	-42C	+1.12, M		Tr		10				Tr		Tr	
	-42D	+1.68, M		Tr	Tr	8				2			
43	CC-43A	U, T	9							1			Tr
	-43B	U, D	3	1	2			2		2			
	-43C	U, Ms	2	1	3			2		2			

Mineralogic composition of tuffaceous rocks of the Chalk Hills Formation, Owyhee County, Idaho, as estimated from X–ray diffractometer patterns of bulk samples—Continued

Locality No. (fig. 3)	Sample No.	Sample location	Glass	Mica	Smectite	Clinop- tilolite	Potas- sium feldspar	Quartz	Opal- CT	Plagio- clase	Calcite	Gypsum	Other
44	CC-44A	–0.20, Ms			5	2		1	2	Tr			
	-44B	+0.20, M			2	3			4	1			
	-44C	+0.90, M			1	6			3				
	44D	+4.43, M	5		5			Tr				Tr	
	-44E	+8.10, M	5		5			Tr		Tr			
	-44F	+8.30, M	5		5					Tr			
45	CC-45-1	–0.20, Ms		Tr	3	2			3	2		Tr	
	-45-2	+0.30, M	6		2				2	Tr			
	-45-4	+40.41, D	3	1	4			1		1		Tr	
46	CC-46A	+0.05, M	10							Tr			
	-46D	+0.10, M	10										
	-46B	+0.30, M	5		5					Tr	Tr		
	-46C	+1.99, M	7	Tr	3					Tr			
47	CC-47	M, lower part	8		2					Tr			
48	CC-48	Uk, Ss		1	1			4		4			
49	CC-49A	-3.41, T	5		5					Tr			
	-49C	-1.68, Ms		Tr	4	2		2		2			
	-49D	–0.02, Ms		1	6	1	***	1	1	Tr			
	-49E	+0.63, M	10	Tr				Tr					
	–49F	+3.36, M	9	Tr	1			Tr					
50	CC-50A	L, Ss		Tr	3			3		4			
	-50B	L, Ss		Tr	2	1		3		4			
	-50C	L, T				Tr		Tr	5	Tr	5		
51	CC-51A	L, St		Tr	2	2		2	3	1			
	-51B	L, Ss		Tr	Tr	4		2	1	3			
52	CC-52A	M, lower part	1		Tr	9							
	-52B	M, middle part	9		1	Tr				Tr			
	-52C	M, upper part			1	1		Tr	8				
53	CC-53A	–0.50, Ms		1	2	3		2	1	1			
	-53B	+0.05, M			Tr	10							
	-53D	+1.75, M			1	9					Tr		
	-53E	+3.50, M			1	8		Tr		1			

Mineralogic composition of tuffaceous rocks of the Chalk Hills Formation, Owyhee County, Idaho, as estimated from X–ray diffractometer patterns of bulk samples—Continued

Locality No. (fig. 3)	Sample No.	Sample location	Glass	Mica	Smectite	Clinop- tilolite	Potas- sium feldspar	Quartz	Opal- CT	Plagio- clase	Calcite	Gypsum	Other
54	CC-54	L, Ss		Tr	1	4		3		2		Tr	
57	CC-57A -57B	M, lower part M, upper part		Tr 	2 2	6 6			2 2				
58	CC-58A -58B	U, T U, Ms	6	 Tr	2 7			1 2	1	Tr 1	 Tr	 Tr	Tr
	-58C	U, Ss			1			9	Tr				Tr
59	CC-59	U, Ms		Tr	2	2		1	5	Tr		Tr	
61	CC-61A	L, T	9		1			Tr		Tr			
	-61B	L, T			Tr				10				
	-61C	L, T		Tr	1	5		2		2			
63	CC-63A	U, T	8		2								
	–63B	U, Ms		1	5			2	1	1			
	-63C	U, T	5		2			2	Tr	1			
64	CC-64A	M, lower part	4		2			2	1	1		Tr	
	-64B	M, middle part	6		3				Tr	1		Tr	
65	CC-65A	M, lower part			3	4			3				
	–65B	M, middle part			2	5			3				
	-65C	M, upper part			3	5			2				
66	CC-66A	–0.05, Ms		1	6			2		1			
	-66B	+0.30, M	8		2								
67	CC-67A	–0.20, Ms			5			3	Tr	2			
	67B	+0.04, M	9	Tr	1								
	-67C	+0.20, M	10										
68	CC-68A	–0.20, Ms	2	Tr	2	4		1		1			
	–68B	+0.10, M	7		Tr	1		Tr	2	Tr			
	–68C	+0.40, M	9		1	Tr							
	-68D	+2.33, M	2		Tr	7			1				
	-68E	+3.10, M	8		1	1				Tr			
69	CC-69	L, T			Tr	10							
70	CC-70A	M, lower part				8			2				
	-70B	M, upper part			2	8							

Mineralogic composition of tuffaceous rocks of the Chalk Hills Formation, Owyhee County, Idaho, as estimated from X-ray diffractometer patterns of bulk samples—Continued

Locality No. (fig. 3)	Sample No.	Sample location	Glass	Mica	Smectite	Clinop- tilolite	Potas- sium feldspar	Quartz	Opal- CT	Plagio- clase	Calcite	Gypsum	Other
71	CC-71-1	–25.56, T			1	5		2		2			
	-71-2	–22.35, T			1	2	2	5		Tr			
	-71-3	−19.00, Ss		Tr	1	3		4		2			
	-71-5A	+0.04, M			Tr	10							
	-71-5B	+3.75, M			1	9							
	-71-5C	+6.50, M			Tr	10							
	-71-6	+8.27, St			2	6				2			
	-71-7	+9.22, St		Tr	2	5		1		2			
72	CC-72A	+0.20, M			Tr	10							
	-72B	+2.00, M			Tr	10							
	-72C	+5.60, M		Tr	1	8		Tr		1			
73	CC-73A	+0.20, M			1	5			4				
	-73B	+1.00, M			1	6			3				
	-73C	+2.00, M			1	9							
	-73D	+4.50, M			2	8							
	-73E	+6.25, M			1	7			2				
74	CC-74	M, lower part			1	7		2					
75	CC-75	L, T	6		4								
76	CC-76A	Uk, T, lower part				6			4				
	-76B	Uk, T, middle part				7			3				
	-76C	Uk, T, upper part		Tr	2	6		2				Tr	
	-76D	Uk, Ss		Tr		2		4	1	3			
77	CC-77	Uk, T	8	Tr	2								
78	CC-78A	M, lower part	9	Tr	1								
	-78B	M, upper part	9	Tr	1			Tr					

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