

Data Repository Item

1. SAMPLING LOCATIONS AND OXYGEN ISOTOPE DATA

Table DR1. Sample Locations, Estimated Ages (Packer, 1979; Smith, 1988; Lindsey, 1996), $\delta^{18}\text{O}$ values of Authigenic Smectites, and Calculated $\delta^{18}\text{O}$ Values of Ancient Meteoric Water (Sheppard and Gilg, 1996).

Number	Name	Latitude	Longitude	Estimated Age (Ma)	$\delta^{18}\text{O}_{\text{VSMOW}} \pm \sigma (\text{‰})$ n = 3	Calculated $\delta^{18}\text{O}_{\text{VSMOW}}$ of ancient meteoric water (‰)
①	Naches-Wenas Grade	N46° 44.240'	W120° 40.844'	9.4 ± 0.1	11.0 ± 0.08	-16.1
				10.0 ± 0.1	11.7 ± 0.13	-15.4
②	Wenas Reservoir	N46° 48.349'	W120° 39.127'	< 0.1	9.3 ± 0.12	-18.1
				~ 13 ± 0.3	12.5 ± 0.11	-14.6
				~ 14 ± 0.3	12.4 ± 0.1	-14.7
③	East of the Ellensburg	N46° 00.006'	W120° 31.816'	< 0.1	9.6 ± 0.08	-17.7
				~ 1.0	10.4 ± 0.12	-16.9
④	East of Kittitas	N46° 59.001'	W120° 15.895'	15.6	13.0 ± 0.1	-14.1
⑤	Vantage Highway	N46° 58.734'	W120° 15.538'	15.6	13.4 ± 0.11	-13.7
⑥	Frenchman Hill	N46° 57.174'	W119° 39.792'	15.0	12.6 ± 0.14	-14.5
⑦	East of the Saddle Mt.	N46° 48.231'	W119° 23.653'	11 ± 0.1	12.4 ± 0.18	-14.7
				11 ± 0.2	11.8 ± 0.10	-15.3
⑧	Potholes Reservoir	N46° 58.932'	W119° 13.791'	15.0	13.1 ± 0.12	-14.0
⑨	Lind Coulee	N47° 00.530'	W119° 08.365'	< 0.1	10.0 ± 0.18	-17.3
				~ 3.5 ± 0.1	9.6 ± 0.12	-18.0
⑩	White Bluff	N46° 37.501'	W119° 23.715'	~ 4.6 ± 0.2	10.6 ± 0.14	-16.7
				~ 4.8 ± 0.2	10.4 ± 0.12	-17.0

VSMOW—Vienna standard mean ocean water.

2. ANALYTICAL DETAILS**2.1 ISOLATION OF AUTHIGENIC SMECTITE**

About 200 g of each bulk sample was lightly ground using a porcelain mortar and pestle, and was disaggregated in de-ionized water using an ultrasonic bath for about 25 minutes. Then, each disaggregated sample was separated into < 0.2 μm size particles by timed centrifugation.

Not only coarser particles but also finer mineral particles that have higher specific gravity than clay minerals (i.e. feldspar, amphibole, and pyroxene) were removed then.

Each $< 0.2 \mu\text{m}$ -sized sample was chemically treated to remove non-silicate and amorphous phases such as organic matter, carbonates, and oxides. We followed the suggested procedures of Jackson (1979). First, 30% of hydrogen peroxide was used to remove organic matter and manganese oxides. Second, diluted glacial acetic acid was used to remove carbonate minerals. Finally, the sodium citrate-bicarbonate-dithionite (Na-CBD) method was conducted to remove ferric and aluminum oxides. It was recognized that these chemical treatments had no effect on the isotopic compositions of clay minerals (Yeh, 1980). Clay mineralogy for each purified $< 0.2 \mu\text{m}$ -sized particle was identified by the X-ray diffraction (XRD). The rationality of the obtained XRD pattern for each smectite sample was tested by measuring the 00 l position carefully and shape of the 001 reflection (Moore and Reynolds, 1997).

2.2 OXYGEN ISOTOPE ANALYSIS OF AUTHIGENIC SMECTITE

Oxygen isotope analyses of our smectite samples were performed with a laser-fluorination line at the Washington State University GeoAnalytical Lab. Each analysis required about 3 to 5 mg of the purified smectite sample, and each sample was physically mixed with a LiF binder using a porcelain mortar and pestle (Kirschner and Sharp, 1997). Then, each mixture was pressed into a pellet to prevent dispersion.

Because interlayer water in smectites isotopically exchanges almost completely with atmospheric water vapor at room temperature in a few hours (Savin and Epstein, 1970), it was necessary to remove the interlayer water by heating under vacuum at $\sim 200^\circ\text{C}$ for about two hours. After removing the water, a sample holder was placed into a sample chamber while the sample holder was still hot. Before laser heating method described by Sharp (1990), two or three

brief (~1.5 minutes duration) pre-fluorinations were conducted in order to liberate any excess water molecules on the mineral surfaces, and on the internal walls of the sample chamber and the vacuum system. Since smectites are one of the labile minerals, the pre-fluorinations worried us about removal of some stoichiometric oxygen. However, no measurable amount of oxygen was extracted after each short pre-fluorination. Also, reproducibility of each smectite sample was higher with the short pre-fluorinations.

Each sample was heated with a 20W CO₂ laser, and oxygen was liberated as reacted with BrF₅ (Clayton and Mayeda, 1963; Sharp, 1990). Valley et al. (1995) noted that effect of reaction with BrF₅ in the sample chamber on other samples at room temperature was negligible. The released oxygen was passed successively over cold traps and cleaned with KBr. The $\delta^{18}\text{O}$ values were measured with the FinniganTM Delta S Isotope Ratio Mass Spectrometer operated by the ISODAT NT software operating system. The isotopic compositions are expressed in the δ -notation as the relative difference in the isotope values between sample and the Vienna Standard Mean Ocean Water (VSMOW) standard in parts per thousand (‰). The $\delta^{18}\text{O}$ values of the samples were corrected by repeated analyses of the UWG-2 garnet ($\delta^{18}\text{O} = \sim 5.8\text{‰}$) (Valley et al., 1995), with the range from 0.05‰ to 0.22‰ (1 σ) of analytical precision.

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