Nephrite is a near-monomineralic rock composed of tremolite-actinolite, Ca$_2$(Mg,Fe)$_5$Si$_8$O$_{22}$(OH)$_2$. It occurs worldwide (figure 1) and is classified as dolomite-related or serpentine-related according to the different parent rocks and ore-hosting rocks, and both types form by metasomatism (Yui et al., 1988; Tang et al., 1994; Yang and Abduriyim, 1994; Harlow and Sorensen, 2005; Burtseva et al., 2015). The large and well-known dolomite-related nephrite deposits are distributed in the Xinjiang Uyghur Autonomous Region [hereafter abbreviated as Xinjiang] of China, Qinghai Province of China, Siberia in Russia, and Chuncheon in South Korea (figure 1). Data from smaller-scale deposits such as Val Malenco in Italy and Złoty Stok in Poland are also used in this study (figure 1). The rest of the data were collected from nephrites produced at multiple small-scale sources in China: Xiuyan, Tanghe, Dahua, and Luodian (figure 2).

With nephrite jade, a premium is placed on geographic origin since the gem’s cultural significance differs by location. It is possible to have an opinion on the origin of a small amount of nephrite by simple visual examination, since some varieties with unique features can be identified by experienced gemologists. However, the unique hydrogen and oxygen stable isotope ratios of nephrites from different origins can provide a more precise and objective method for determining geographic origin. These isotope ratios are highly effective for discriminating dolomite-related nephrites from the four most important origins worldwide: Xinjiang, Qinghai, Chuncheon, and Vitim.

In Brief

- Geographic origin can have a significant impact on the value of nephrite.
- Hydrogen and oxygen stable isotope ratios, particularly the latter, provide a robust tool for origin determination.
- Dolomite-related nephrites from Vitim, Chuncheon, Xinjiang, and Qinghai differ from one another by distinct hydrogen and oxygen stable isotope ratios.
- Differences in hydrogen and oxygen stable isotope ratios for nephrite are related to ore-forming fluids.
gemological characteristics such as color, luster, and transparency have mainly occurred in specific deposits. In Xinjiang, for example, high-quality white primary nephrite occurs in Qiemo County. There it is commonly associated with brown nephrite (figure 3A, rough with white core and very thick brown rind). The brown is a color seldom found in nephrite from other deposits in Xinjiang. The highest-quality white primary nephrite (figure 3B, white plate) mostly comes from the Hetian region and Qiemo County. Placer nephrite (figures 3C and 3D, pendants with figures carved out of brownish red skin) occurs in the Yulongkashi River and Kalakashi River basins. A considerable quantity of primary nephrite from Ruoqiang County features a yellow color component (figure 3E, greenish yellow fish) that is absent from other samples. Black nephrite (figure 3F, bangle bracelet) colored by graphite, on the other hand, mainly occurs in the Hetian region and has not been found in Qiemo County or Ruoqiang County. However, the origin determination of a tremendous amount of dolomite-related nephrite cannot be solved by this simple observation. Previous researchers used trace elements combined with appearance to identify geographic origin and obtained some informative results [Zhong et al., 2013; Luo et al., 2015]. Unfortunately, rigorous and scientific determination of geographic origin is still not available.

Hydrogen and oxygen isotope ratio values (see box A), which might vary for the same gemstone from different regions due to diverse ore-forming environments and models, can be used for geographic origin determination.

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**TABLE 1. Location names expressed in Chinese pinyin and their English equivalents.**

| Chinese pinyin | English                  |
|---------------|-------------------------|
| A'erjinshan   | Altyng Tagh             |
| Alamasi       | Almas                   |
| Bayinguoleng region | Bayingholin region   |
| Hetian        | Hoten/Hotan             |
| Kalakashi, Hetian | Qaraqash, Hoten        |
| Kashi region  | Kashkar region          |
| Keliya River  | Keriye/Keriya River     |
| Qiemo County, Bayinguoleng | Cherchen County, Bayingholin |
| Ruoqiang County, Bayinguoleng | Chaqiliq County, Bayingholin |
| Takelamagan Desert | Taklimakan Desert     |
| Tashiku'ergan County | Taxkorgan/Tashkurgan County |
| Yecheng County, Kashi | Qaghilig County, Kashkar |
| Yulongkashi, Hetian | Yurungqash, Hoten       |
| Yutian County | Keriye County, Hoten    |

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1This paper uses Chinese pinyin to express all Chinese location names involved, and the corresponding commonly used English names are listed in table 1.
nation [Giuliani et al., 1998, 2000, 2005, 2007]. A mass
spectrometer is needed to determine the isotope ratio
values (see box B). The spot produced by secondary
ion mass spectrometry (SIMS), laser ablation–inductively
coupled plasma–mass spectrometry (LA-ICP-MS), and
laser ablation inductively coupled plasma time-of-
flight mass spectrometry (LA-ICP-TOF-MS) for stable
isotope analysis of gemstones can be restricted to
craters of 10–100 μm in diameter and a few angstroms
to microns deep [Giuliani et al., 2000, 2005; Abduriyim
and Kitawaki, 2006; Wang et al., 2016, 2018]. The
craters produced are very small, to the point of not

Figure 2. The distribution of main dolomite-related nephrite deposits of China. Primary and placer nephrite occur
at Xiuyan, but the latter is not plotted in the map.
being noticeable without magnification. This permits the method to be applied to gemstones and historical antiques (Giuliani et al., 2000, 2005).

Geographic origin discrimination of nephrite by isotopes is seldom reported, even though many hydrogen and oxygen isotope ratio studies on this material have been carried out (table 2). By summarizing and analyzing all available hydrogen and oxygen isotopic data of dolomite-related nephrites worldwide from published references, this study discusses the geographic origin discrimination based on the relationship between the characteristics of nephrite and its formation environment.

**WHY DO ISOTOPE RATIOS MATTER TO GEMOLOGISTS?**

The application of isotopes has gradually attracted the attention of gemologists (Wang et al., 2016). In addition to hydrogen and oxygen isotope ratios, which can help determine the geographical origins of corundum and emerald (Giuliani et al., 1998, 2000, 2005, 2007; Wang...
et al., 2019), carbon isotopes are considered useful in identifying natural and synthetic diamonds (Wang et al., 2014), and radioactive isotopes have also been used to determine the ages of gemstones (Link, 2015).

Traditional methods using the parameters of inclusions, optical characteristics, and trace elements are often not enough to solve the problems of geographic origin determination of nephrite. Isotopic analysis has provided geochemical and chronological information for all sorts of geological samples: Stable isotopes can be used to study gemstone origin (source materials, formation process, and geographical locations), whereas radioactive isotopes can be utilized to determine the formation ages. The stable isotope study of dolomite-related nephrite in our works, together with previous studies on corundum and emerald (Giuliani et al., 1998, 2000, 2005, 2007; Wang et al., 2019), show that the geographic origin characteristics of isotopes in gemstones can be explained from their formation environment and formation process. Thus, relative isotopic abundances are reliable parameters for determining geographic origin and offer a sound complement to traditional methods.

### DATA AND CALCULATION

In all, 120 sets of hydrogen and oxygen isotope data (some lacking hydrogen data) for dolomite-related nephrites were collected from all known related published studies, from a variety of researchers (table 2 and figure 4), to illustrate geographic origin discrimination with stable isotopic ratios.

Hydrogen and oxygen isotope delta values of nephrite can be used to calculate the corresponding values of its formation fluids. Hydrogen isotope fractionation of tremolite relative to water is not affected by temperature in the approximate range of 350° to 650°C (Graham et al., 1984), and thus

$$
ln \alpha_{H_{2}O} = -21.7 \pm 2
$$

### Box A: Introduction to Oxygen and Hydrogen Stable Isotope Ratios

Atoms with an equivalent atomic number (i.e., atoms of the same element) can differ from one another in their number of neutrons. For example, $^{16}$O has 8 protons and 8 neutrons, and $^{18}$O has 8 protons and 10 neutrons. Such atoms with the same number of protons but different numbers of neutrons are defined as isotopes.

The mass difference inherent from divergent neutrons causes isotopic fractionation, which occurs as the isotopes of an element are distributed between two substances or phases in differing ratios in a given system. This process can be affected by temperature, equilibrium or kinetic processes, and other physiochemical processes. The isotope fractionation will reach and maintain equilibrium unless conditions change. Therefore, isotope abundance can be used as a tracer to reveal certain geochemical processes in geological bodies.

Isotope ratio, defined as the measured relative abundance of a heavy isotope to its lighter counterpart (e.g., $^{18}$O/$^{16}$O and H/H), is typically used rather than the isotope abundance itself. The isotopic fractionation factor ($\alpha$) is introduced to represent the extent of fractionation of isotopes between two phases. It is defined as the ratio of isotope ratios in one phase to the other coexisting phase. For example, in a system consisting of phase A and phase B, the oxygen isotope fractionation factor can be defined as

$$
\alpha_{A-B} = \frac{[^{18}\text{O}/^{16}\text{O}]_A}{[^{18}\text{O}/^{16}\text{O}]_B}
$$

The isotopic fractionation factor is always a function of temperature, which can be obtained by theoretical calculation or experiment (Graham et al., 1984; Zheng, 1993, 1995).

Both oxygen and hydrogen isotope ratios are also reported in so-called delta notation given in terms of per mil [%]. In other words, the delta value

$$
\delta^{18}\text{O} = \frac{[^{18}\text{O}/^{16}\text{O}]_{\text{sample}}-[^{18}\text{O}/^{16}\text{O}]_{\text{standard}}}{[^{18}\text{O}/^{16}\text{O}]_{\text{standard}}} \times 1000\%
$$

and

$$
\delta^2\text{H} = \frac{[^{2}\text{H}/^{1}\text{H}]_{\text{sample}}-[^{2}\text{H}/^{1}\text{H}]_{\text{standard}}}{[^{2}\text{H}/^{1}\text{H}]_{\text{standard}}} \times 1000\%
$$

in which $^{18}$O/$^{16}$O and H/H are the isotope ratios defined above. Values of delta > 0 indicate that relative to the standard samples, the tested sample has a higher heavy isotope abundance, and a negative delta value indicates a higher light isotope abundance.

International general isotope standards are issued by the International Atomic Energy Agency (IAEA) and the U.S. National Institute of Standards and Technology (NIST). The delta values of hydrogen and oxygen isotopes are calculated using the value for Standard Mean Ocean Water (SMOW), which has $^{2}$H/H of (155.76 ± 0.10) × 10–6, $^{18}$O/$^{16}$O of (2005.20 ± 0.43) × 10–6, and $^{17}$O/$^{16}$O of (373 ± 15) × 10–6. Other hydrogen isotope standard samples include SLAP, GISP, NBS-22, and NBS-30.
while oxygen isotope fractionation (Zheng, 1993, 1995) can be expressed as

\[
10^3 \ln a_{\text{Tr}-\text{H}_2\text{O}} = (3.95 \times 10^6/T^2) - (8.28 \times 10^3/T) + 2.38
\]

In both equations, \(a_{\text{Tr}-\text{H}_2\text{O}}\) is the isotopic fractionation factor [see box A] between the nephrite and its formation fluid, and \(T\) is the absolute temperature (K) of the nephrite-forming system. The nephrite formation temperature is confined to approximately 223°–425°C, especially near 350°C (Tang et al., 1994; Yui and Kwon, 2002; Chen et al., 2014; Liu et al., 2016), by methods using the homogenization temperatures of tremolite fluid inclusions [Liu et al., 2011a; Chen et al., 2014], the combination of the pyrite decrepitation temperature and calcite homogenization temperature [Wang et al., 2007; Xu and Wang, 2016], the mineral assemblage [Yang, 2013], and isotopes [Yui et al., 1988]. Thus, the value of 350°C was used to calculate the isotopes of fluids from which nephrite forms.

Both the isotopic fractionation factor \(a_{\text{Tr}-\text{H}_2\text{O}}\) and delta values \(\delta^{18}O, \delta^{2}H\) are defined after isotope ratios \((^{18}O/^{16}O, ^2H/^{1}H)\) of nephrite and its formation fluids [see box A]. Thus, the delta values of the nephrite-forming fluids \(\delta^{18}O_{\text{Tr}-\text{H}_2\text{O}}, \delta^{2}H_{\text{Tr}-\text{H}_2\text{O}}\) table 2) can be calculated from the delta values of corresponding nephrite, which is acquired by isotope determination [see box B].

GEOGRAPHIC ORIGIN CHARACTERISTICS

Vitim in Russia, Chuncheon in South Korea, and Xinjiang and Qinghai in China are the four most important dolomite-related nephrite source areas. The relative abundances of the hydrogen and oxygen isotopes of nephrites from these regions differ significantly (figure 4). In particular, oxygen isotope \(\delta^{18}O\) values (see figure 4 and table 2) range from −20.0‰ to −14.6‰, −9.9‰ to −8.2‰, 0.5‰ to 7.9‰, and 11.4‰ to 12.6‰, respectively, without any overlap. Cowell in Australia is considered another large dolomite-related nephrite deposit but is seldom studied. The only \(\delta^{2}H–\delta^{18}O\) data (see figure 4 and table 2) fall within the range of Xinjiang placer nephrite; nevertheless, the \(\delta^{2}H\) values are significantly higher than those of Xinjiang primary nephrite.

The nephrites from Xinjiang, distributed in a belt longer than 1300 km, show convergent hydrogen and oxygen isotopic characteristics. The isotope delta values of their primary dolomite-related nephrites are covered by placer ones (figure 4).
BOX B: ANALYTICAL METHODS FOR DETERMINING OXYGEN AND HYDROGEN ISOTOPE CONCENTRATIONS

Isotope concentrations are commonly measured with a mass spectrometer operating on the principle that the degree of deflection of charged particles in a magnetic field is inversely proportional to the mass-to-charge ratio \([m/z]\) [figure B-1]. Generally, mass spectrometers can be divided into four parts: the sampling system, the ion source, the mass analyzer, and the detector.

Stable isotope analysis has advanced from macroanalysis to microanalysis and now includes methods of static mass spectrometry, laser ablation [multi-collector]–inductively coupled plasma–mass spectrometry (LA-(MC)-ICP-MS), and secondary ion mass spectrometry (SIMS). High accuracy and low sample loss make these technologies suitable for isotopic analysis of gemstones.

Samples from some relatively small deposits such as Xiuyan in Liaoning Province, Złoty Stok in Poland, and Dahua in Guangxi Province (figure 4) show slightly higher \(\delta^2\text{H}\) values than those of Qinghai nephrite and Xinjiang primary nephrite. Their ranges of \(\delta^{18}\text{O}\) values cover that of Sanchakou samples but do not overlap with Xinjiang primary nephrite. Fortunately, nephrites from these three regions typically have their own distinct appearances. Luodian nephrite from Guizhou has notably higher \(\delta^{18}\text{O}\) values than the others [no \(\delta^2\text{H}\) value data have been collected]. In recent years, secondary nephrite has been found in the Tanghe River in Hebei Province. It is speculated to be a dolomite-related nephrite according to the regional geology, field observation, and petrographic analysis [Chen et al., 2014]. Its hydrogen and oxygen isotope ratios are completely isolated from others in the plot of \(\delta^2\text{H}–\delta^{18}\text{O}\) [figure 4] by low \(\delta^2\text{H}\) and high \(\delta^{18}\text{O}\) values.

NEPHRITE-FORMING FLUIDS FROM MAGMATIC WATER AND METEORIC WATER

Fluids containing gases, liquids, and silicate compositions always occur as the most active parts of geological processes. They are composed mainly of \(\text{H}_2\text{O}, \text{CO}_2, \text{NaCl}, \text{metal components}, \text{silicate compositions}, \) and organic matter. The fluids that correspond to nephrite formation are hydrothermal fluids, which refer to gas-liquid two-phase systems having their own temperatures and pressures. Hydrothermal fluids are released from magma [magmatic fluids] or metamorphism [metamorphic fluids] due to changes in temperature and pressure. They also can be meteoric waters [including rainwater, lake water, seawater, river water, glacial water, and shallow groundwater] heated by geological processes.

The original characteristics of the hydrogen and oxygen isotopes of nephrite mainly result from the ore-forming fluids. The calculated \(\delta^2\text{H}_{\text{T-H}_2\text{O}}\) and \(\delta^{18}\text{O}_{\text{H}_2\text{O}}\) values of hydrothermal fluids forming the Vitim and Chuncheon nephrites plot near the Craig line\(^2\) [figure 5], indicating that their predominant ore-forming fluids were meteoric waters in an environment with a high fluid/rock ratio [Yui and Kwon, 2002; Burtseva et al., 2015].

For the Xinjiang nephrite, magmatic fluid, meteoric water, and metamorphic water are all possible candidates for the ore-forming fluids [figure 5], and a low fluid/rock ratio is indicated [Yui and Kwon, 2002; Liu et al., 2011a, 2011b, 2016]. The \(\delta^{18}\text{O}_{\text{H}_2\text{O}}\) values of the nephrite-forming fluids for Alamasi nephrite, which occurs in granite-dolomite contact zones [Liu et al., 2010, 2011a], decrease in the con-

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\(^2\)The Craig line, also referred to as the meteoric water line, represents the relationship between \(\delta^2\text{H}\) and \(\delta^{18}\text{O}\) of meteoric water—i.e., \(\delta^2\text{H} = 8\delta^{18}\text{O} + 10\) (Craig, 1961). The kaolinite line (Zheng and Chen, 2000) shown in figure 5 represents the relationship between \(\delta^2\text{H}\) and \(\delta^{18}\text{O}\) of kaolinite in weathering profile (i.e., \(\delta^2\text{H} = 7.5\delta^{18}\text{O} - 220\)). Most of the soil samples in nature fall on or near the kaolinite line.
tact zone in the order of granite → nephrite → wall rock. The δ¹⁸O_H₂O values of magmatic fluids, seldom influenced by crustal rocks during intrusion, should equal the high values of the Xinjiang nephrite-forming fluids (figure 5). The δ¹⁸O_dol values of wall rock are far lower than those of common carbonates of sedimentary origin, at only 6.1‰ (Wan et al., 2002). Then, the δ¹⁸O_H₂O value for the water in equilibrium with wall rock is 1.6‰ (1000 lnα_{dol,H₂O} = 3.06 × 10⁶/T²−3.24 after Zheng and Chen (2000), assuming that the temperature for the wall rock during nephrite formation was between 252°C and 295°C). This value is lower than those of the fluids in equilibrium with most of the Xinjiang nephrite (figure 5). In addition, considering the characteristics of the chemical zoning (Liu et al., 2010), the higher δ¹⁸O_H₂O values for green nephrite fluids than for white ones in the Alamasí deposit (Wan et al., 2002) provide another indicator that oxygen isotopes decrease from granite to wall rock. However, the δ¹⁸O_H₂O value should have increased gradually if water unilaterally diffused from the granite to the wall rock, since water in equilibrium with nephrite is enriched or slightly depleted in ¹⁸O (depending on the temperature, calculated according to Equation 2 with T around 350°C). Considering that the δ¹⁸O value of the Alamasí nephrite is negatively related to the δ¹⁸O value (figure 4), the conflict can be explained by dualistic fluid sources. One is post-magmatic hydrothermal fluids provided by the granite forming the nephrite, while the other must be the meteoric water from the dolomite marble.

Figure 5. δ²H_{H₂O} and δ¹⁸O_{H₂O} data of dolomite-related nephrite-forming fluids at 350°C. Nephrites from Alamasí, Yulongkashi, and Kalakashi show a trend marked by a set of arrows from the bottom right to the top left, which can be explained as dualistic fluid sources. Compared with the primary nephrite from Alamasí, the hydrogen isotope of placer nephrite changes greatly (indicated by the vertical arrow) without distinct δ¹⁸O_{H₂O} variations, which are caused by fluid-rock reaction possibly. The ellipses represent hydrogen and oxygen isotopes of water from (1) the Qiemo River basin (Wang et al., 2013), covering the A’erjinshan region, (2) the Hetian River basin and Keliya River basin, covering the Alamasí, Agejugai, and Hetian regions, and (3) a pond in the Taklimakan Desert hinterland (Li et al., 2006).
TABLE 2. Hydrogen and oxygen isotope delta values of dolomite-related nephrites.

| No. | δ¹⁸O (‰) | δ²H (‰) | δ¹⁸O_H₂O (350°C) (‰) | δ²H_H₂O (350°C) (‰) | Locality | Description | Mass spectrometer | Reference |
|-----|----------|----------|-----------------------|-----------------------|-----------|-------------|------------------|-----------|
| 1   | 3.8      | −86.7    | 4.5                   | −65                   | White     | White       | MAT-252          | Liu et al. (2011a) |
| 2   | 3.2      | −83      | 3.9                   | −61.3                 | White     | White       | MAT-252          | Liu et al. (2011a) |
| 3   | 6.1      | −93.1    | 6.8                   | −71.4                 | White-green| White-green | MAT-252          | Liu et al. (2011a) |
| 4   | 4.6      | −89      | 5.3                   | −67.3                 | White-green| White-green | MAT-252          | Liu et al. (2011a) |
| 5   | 3.5      | −85.1    | 4.2                   | −63.4                 | White-green| White-green | MAT-252          | Liu et al. (2011a) |
| 6   | 3.6      | −85.9    | 4.3                   | −62.2                 | Alamosi, Yutian County, Hetian (Xinjiang, China)| White-green | MAT-252          | Liu et al. (2011a) |
| 7   | 6.2      | −94.7    | 6.9                   | −73                   | Green     | Green       | MAT-252          | Liu et al. (2011a) |
| 8   | 4.1      | −90.2    | 4.8                   | −68.5                 | Green     | Green       | MAT-252          | Liu et al. (2011a) |
| 9   | 3.6      | −85      | 4.3                   | −63.3                 | Green     | Green       | MAT-252          | Liu et al. (2011a) |
| 10  | 4.9      | −91.6    | 5.6                   | −69.9                 | Green     | Green       | MAT-252          | Liu et al. (2011a) |
| 11  | 4.8      | −90.4    | 5.5                   | −68.7                 | Green     | Green       | MAT-252          | Liu et al. (2011a) |
| 12  | 3.8      | −86.2    | 4.5                   | −64.5                 | Green     | Green       | MAT-252          | Liu et al. (2011a) |
| 13  | 3.8      | 4.5      |                       |                       | White     | White       | MAT-252          | Liu et al. (2011a) |
| 14  | 3.7      | 4.4      |                       |                       | White-green| White-green | MAT-252          | Liu et al. (2011a) |
| 15  | 3.6      | 4.3      |                       |                       | Green     | Green       | MAT-252          | Liu et al. (2011a) |
| 16  | 2.3      | 3.0      |                       |                       | Mutton-fat| Mutton-fat  | MAT-252          | Liu et al. (2011a) |
| 17  | 5.8      | 6.5      |                       |                       | Agejygai, Hetian County, Hetian (Xinjiang, China)| White     | MAT-252          | Liu et al. (2011a) |
| 18  | 5.6      | 6.3      |                       |                       | White-green| White-green | MAT-252          | Liu et al. (2011a) |
| 19  | 6.5      | 7.2      |                       |                       | Green     | Green       | MAT-252          | Liu et al. (2011a) |
| 20  | 5.3      | 6.0      |                       |                       | Yecheng County, Kashi (Xinjiang, China)| Green     | MAT-252          | Liu et al. (2011a) |
| 21  | 4.6      | 5.3      |                       |                       | Datong, Tashiku’ergan County, Kashi (Xinjiang, China)| Green     | MAT-252          | Liu et al. (2011a) |
| 22  | 4.4      | 5.1      |                       |                       | Ruoqiang County, Bayinguoleng (Xinjiang, China)| White-green| MAT-252          | Liu et al. (2011a) |
| 23  | 3.4      | 4.1      |                       |                       | Qiemo County, Bayinguoleng (Xinjiang, China)| White-green| MAT-252          | Liu et al. (2011a) |
| 24  | 3.9      | 4.6      |                       |                       | Light brown| Light brown| MAT-252          | Liu et al. (2011a) |
| 25  | 4.8      | 5.5      |                       |                       | Brown     | Brown       | MAT-252          | Liu et al. (2011a) |
| 26  | 4.7      | 5.4      |                       |                       | Brown     | Brown       | MAT-252          | Liu et al. (2011a) |
| 27  | 5.6      | 6.3      |                       |                       | White-green| White-green | MAT-252          | Liu et al. (2011a) |
| 28  | 4.6      | 5.3      |                       |                       | Green     | Green       | MAT-252          | Liu et al. (2011a) |
| 29  | 3.3      | 3.7      |                       |                       | Green     | Green       | MAT-252          | Liu et al. (2011a) |
| 30  | 3.6      | 4.3      |                       |                       | Tashisayi Qiemo County, Bayinguoleng (Xinjiang, China)| Green     | MAT-252          | Liu et al. (2011a) |
| 31  | 3.7      | 4.4      |                       |                       | Green     | Green       | MAT-252          | Liu et al. (2011a) |
| 32  | 3.1      | 3.8      |                       |                       | Green     | Green       | MAT-252          | Liu et al. (2011a) |
| 33  | 3.9      | 4.6      |                       |                       | Green     | Green       | MAT-252          | Liu et al. (2011a) |
| 34  | 4.4      | 4.7      |                       |                       | Light brown| Light brown| MAT-252          | Liu et al. (2011a) |
| 35  | 4.6      | 5.3      |                       |                       | Green     | Green       | MAT-252          | Liu et al. (2011a) |
| 36  | 4.8      | 5.5      |                       |                       | Green     | Green       | MAT-252          | Liu et al. (2011a) |
| 37  | 5.2      | −71.8    | 5.9                   | −50.1                 | Yulongkashi, Hetian (Xinjiang, China)| White, placer| MAT-252          | Liu et al. (2011b) |
| 38  | 3.7      | −67.3    | 4.4                   | −45.6                 | Yulongkashi, Hetian (Xinjiang, China)| White, placer| MAT-252          | Liu et al. (2011b) |
| 39  | 5.6      | −72.4    | 6.3                   | −50.7                 | Kalakashi, Hetian (Xinjiang, China)| White-green, placer| MAT-252          | Liu et al. (2011b) |
| 40  | 1.1      | −55.7    | 1.8                   | −34                   | Kalakashi, Hetian (Xinjiang, China)| White-green, placer| MAT-252          | Liu et al. (2011b) |
| 41  | 5        | −71.4    | 5.7                   | −49.7                 | Kalakashi, Hetian (Xinjiang, China)| White-green, placer| MAT-252          | Liu et al. (2011b) |
| 42  | 2.9      | −65.7    | 3.6                   | −44                   | Kalakashi, Hetian (Xinjiang, China)| White-green, placer| MAT-252          | Liu et al. (2011b) |
| No. | \( \delta^{18}O \) (‰) | \( \delta^2H \) (‰) | \( \delta^{18}O_{H_2O} \) (350°C) (‰) | \( \delta^2H_{H_2O} \) (350°C) (‰) | Locality | Description | Mass spectrometer | Reference |
|-----|----------------|----------------|-----------------|----------------|-------------|-------------|-----------------|-----------|
| 43  | 3.2            | −68.7          | 3.9             | −47            | Cowell, South Australia (Australia) | Black, placer | MAT-252         | Liu et al. (2011b) |
| 44  | 2.4            | −63.3          | 3.1             | −41.6          | Vitim area, Buryatia (Russia) | Black, placer |               |           |
| 45  | 4.5            | −69.3          | 5.2             | −47.6          | Burtseva et al. (2015) | Black, placer |               |           |
| 46  | 3.1            | −67.1          | 3.8             | −45.4          | Burtseva et al. (2015) | Black, placer |               |           |
| 47  | 0.8            | −97            | 1.5             | −75.3          | Black, placer | Green, placer |               |           |
| 48  | 7.3            | −67            | 8.0             | −45.3          | Black, placer | Green, placer | MAT-252 | Liu et al. (2016) |
| 49  | 2.7            | −80            | 3.4             | −58.3          | Black, placer | Green, placer |               |           |
| 50  | 6.6            | −77            | 7.3             | −55.3          | Black, placer | Black, placer |               |           |
| 51  | 3.6            | −87            | 4.3             | −65.3          | Green, placer | Kalakashi, Hetian (Xinjiang, China) | Black, placer | MAT-252 | Liu et al. (2016) |
| 52  | 6.7            | −58            | 7.4             | −36.3          | Black, placer | Black, placer |               |           |
| 53  | 3              | −86            | 3.7             | −64.3          | Black, placer | Black, placer |               |           |
| 54  | 4.9            | −85            | 5.6             | −63.3          | Green, placer | Green, placer | MAT-252 | Liu et al. (2016) |
| 55  | 2.2            | −100           | 2.9             | −78.3          | Black, placer | Black, placer |               |           |
| 56  | 7.9            | −79            | 8.6             | −57.3          | Black, placer | Black, placer |               |           |
| 57  | 2.5            | −88            | 3.2             | −66.3          | Green, placer | Black, placer |               |           |
| 58  | 4.3            | −109           | 5.0             | −87.3          | Green, placer | Green, placer |               |           |
| 59  | 1.6            | −93            | 2.3             | −71.3          | Green, placer | Green, placer |               |           |
| 60  | 0.5 to 2.3     | −108 to −124   | 1.24 to 3.04    | −86.3 to −102.3 | Kunlun (Xinjiang, China) | Kunlun (Xinjiang, China) | Primary or placer unknown | Yu and Kwon (2002) |
| 61  | 12.3           | −76.9          | 13.0            | −55.2          | Dahua (Guangxi, China) | Dahua (Guangxi, China) |               | Xu and Wang (2016) |
| 62  | 10.5           | −79.8          | 11.2            | −58.1          | Dahua (Guangxi, China) | Dahua (Guangxi, China) |               |           |
| 63  | −8.7           | −108           | −8.0            | −86.3          | Black, placer | Black, placer |               |           |
| 64  | −8.4           | −114           | −7.7            | −92.3          | Black, placer | Black, placer |               |           |
| 65  | −9.9           | −105           | −9.2            | −83.3          | Black, placer | Black, placer |               |           |
| 66  | −9             | −107           | −8.3            | −85.3          | Black, placer | Black, placer |               |           |
| 67  | −8.2           | −108           | −7.5            | −86.3          | Black, placer | Black, placer |               |           |
| 68  | −8.6           | −112           | −7.9            | −90.3          | Black, placer | Black, placer |               |           |
| 69  | −8.9           | −109           | −8.2            | −87.3          | Black, placer | Black, placer |               |           |
| 70  | −9.3           | −110           | −8.6            | −88.3          | Black, placer | Black, placer |               |           |
| 71  | −9.2           | −109           | −8.5            | −87.3          | Black, placer | Black, placer |               |           |
| 72  | 3.4            | −57            | 4.1             | −35.3          | Cowell, South Australia (Australia) | Cowell, South Australia (Australia) |               | Yu and Kwon (2002) |
| 73  | −15.52         | −119.3         | −14.8           | −97.6          | Chuncheon (South Korea) | Chuncheon (South Korea) |               |           |
| 74  | −16.8          | −16.1          | −16.1           | −156.8         | Chuncheon (South Korea) | Chuncheon (South Korea) |               |           |
| 75  | −17.24         | −178.5         | −16.5           | −156.8         | Chuncheon (South Korea) | Chuncheon (South Korea) |               |           |
| 76  | −15.51         | −14.8          | −14.2           | −111.5         | Chuncheon (South Korea) | Chuncheon (South Korea) |               |           |
| 77  | −14.95         | −14.2          | −14.2           | −111.5         | Chuncheon (South Korea) | Chuncheon (South Korea) |               |           |
| 78  | −14.93         | −133.2         | −14.2           | −111.5         | Chuncheon (South Korea) | Chuncheon (South Korea) |               |           |
| 79  | −15.1          | −14.4          | −14.4           | −111.5         | Chuncheon (South Korea) | Chuncheon (South Korea) |               |           |
| 80  | −14.58         | −13.8          | −13.8           | −111.5         | Chuncheon (South Korea) | Chuncheon (South Korea) |               |           |
| 81  | −18.63         | −17.9          | −17.9           | −111.5         | Chuncheon (South Korea) | Chuncheon (South Korea) |               |           |
| 82  | −17.33         | −16.6          | −16.6           | −111.5         | Chuncheon (South Korea) | Chuncheon (South Korea) |               |           |
| 83  | −20.02         | −19.3          | −19.3           | −111.5         | Chuncheon (South Korea) | Chuncheon (South Korea) |               |           |
| 84  | −17.24         | −16.5          | −16.5           | −111.5         | Chuncheon (South Korea) | Chuncheon (South Korea) |               |           |
TABLE 2 (continued). Hydrogen and oxygen isotope delta values of dolomite-related nephrites.

| No. | δ¹⁸O (%) | δD (%) | δ¹⁸O_H₂O(350°C) (%) | δD_H₂O(350°C) (%) | Locality                   | Description                      | Mass spectrometer | Reference                  |
|-----|----------|--------|---------------------|-------------------|----------------|-----------------------------|------------------|-----------------------------|
| 85  | 10.2     | −76.4   | 10.9                | −54.7             | Złoty Stok, Lower Silesian (Poland) | MAT-253           | Gil et al. (2015a)          |
| 86  | 8.3      | −76.2   | 9.0                 | −54.5             | Luodian (Guizhou, China) |                |                 |                             |
| 87  | 10.4     | −77.2   | 11.1                | −55.5             | Chen et al. (2014)                               |                 |                             |
| 88  | 10.2     | −74.6   | 10.9                | −52.9             | Yang (2013)                                              |                 |                             |
| 89  |         | −113 ± 4.8 | −91.3                |                   | Val Malenco, Sondrio (Italy) |                |                 | Adamo and Bocchio (2013)    |
| 90  | 10       | −70     | 10.7                | −48.3             | Placer/secondary | White |                |                             |
| 91  | 9.3      | −74     | 10.0                | −52.3             | Placer/secondary | Yellow-white          |                 |                             |
| 92  | 8.5      | −74     | 9.2                 | −52.3             | Placer/secondary | Yellow            |                 |                             |
| 93  | 8.1      | −72     | 8.8                 | −50.3             | Placer/secondary | Yellow            |                 |                             |
| 94  | 13.3     | −70     | 14.0                | −48.3             | Green-white     | White             |                 |                             |
| 95  | 11.7     | −73     | 12.4                | −51.3             | Green           | White             |                 |                             |
| 96  | 10.4     | −76     | 11.1                | −54.3             | Xiuyan (Liaoning, China) |                |                 |                             |
| 97  | 10.3     | −72     | 11.0                | −50.3             | White           | White             |                 |                             |
| 98  | 9.1      | −74     | 9.8                 | −52.3             | Green-white     | White             |                 |                             |
| 99  | 9        | −70     | 9.7                 | −48.3             | White           | White             |                 |                             |
| 100 | 12.4     |         | 13.1                |                   | White           | White             |                 |                             |
| 101 | 9.3      |         | 10.0                |                   | Yellow-white    | Yellow            |                 |                             |
| 102 | 8.7      |         | 9.4                 |                   | Yellow-white    | Yellow            |                 |                             |
| 103 | 11.4     | −86     | 12.1                | −64.3             | Green           | White             |                 |                             |
| 104 | 12.3     | −87     | 13.0                | −65.3             | Green-white     | MAT-251EM         | Zhou (2006)     |                             |
| 105 | 12.2     | −78     | 12.9                | −56.3             | White           | White             |                 |                             |
| 106 | 12.6     | −84     | 13.3                | −62.3             | “Water line” in white nephrite |                |                 |                             |
| 107 | 7.8      | −144    | 8.5                 | −122.3            | Tanghe (Hebei, China) | Placer/secondary |                 | Chen et al. (2014)         |
| 108 | 15.4     | −135    | 16.1                | −113.3            | Tanghe (Hebei, China) | Placer/secondary |                 | Chen et al. (2014)         |
| 109 | 18.9     | −109    | 19.6                | −87.3             | Tanghe (Hebei, China) | Placer/secondary |                 | Chen et al. (2014)         |
| 110 | 11       | −143    | 11.7                | −121.3            | Tanghe (Hebei, China) | Placer/secondary |                 | Chen et al. (2014)         |
| 111 | 15.3     |         | 16.0                |                   | Green-white     | Green             |                 |                             |
| 112 | 14.3     |         | 15.0                |                   | Green           | Green             |                 |                             |
| 113 | 15.6     |         | 16.3                |                   | Green           | Green             |                 |                             |
| 114 | 16.5     |         | 17.2                |                   | Green           | Green             |                 |                             |
| 115 | 14.7     |         | 15.4                |                   | Green           | Green             |                 |                             |
| 116 | 14.5     |         | 15.2                |                   | Green           | Green             |                 |                             |
| 117 | 14.3     |         | 14.8                |                   | Green           | Green             |                 |                             |
| 118 | 14.6     |         | 15.3                |                   | Green           | Green             |                 |                             |
| 119 | 15.5     |         | 16.2                |                   | Green           | Green             |                 |                             |
| 120 | 16.3     |         | 17.0                |                   | Green           | Green             |                 |                             |

*The deposits in Hetian and Kashi are counted in the West Kunlun region, while those in Bayinguanlong are counted in the A'erjinshan rengion.

1 The sample is marked as “Bi yu” in Chinese in the original reference, which mostly equates with serpentine-related nephrite. However, we tend to believe the original authors meant a nephrite with dark green color.

2 Authors Kong Gao, Ting Fang, and Yuanyuan Wang once participated in the research project sponsoring the thesis. Therefore, we can supplement the content of the original literature, which is not detailed enough.

3 Only those tremolite contents higher than 99 wt. % are chosen from Liu et al. (2016). However, it is not ruled out that individual samples may be serpentine-related since their Fe contents can be high.

4 The figure in parentheses is the number of samples tested. The number before the parentheses is the average value.

5 A “water line” refers to the band in nephrite that is more transparent than the matrix. It is composed of prismatic coarse-grained tremolite crystals parallel to each other.
NEPHRITE-FORMING FLUIDS MODIFIED BY METAMORPHISM OR METASOMATISM

The hydrogen and oxygen isotopes of nephrites from Xiuyan (Duan and Wang, 2002; Wan et al., 2002; Wang et al., 2007) and Złoty Stok (Gil et al., 2015a) overlap with each other to some extent (figure 4). Their calculated fluid isotopes plot in the regional metamorphic water field (figure 5), which is in accordance with their geological environment. The Xiuyan nephrite occurs not far from the famed serpentinite jade deposit formed from metamorphic hydrothermal fluids [Wu et al., 2014]. Silicon isotope studies support the interpretation that the formation of the Xiuyan nephrite was related to metamorphic fluids [Duan and Wang, 2002; Wu et al., 2014]. At Złoty Stok, some geological bodies related to serpentinite occur not far from the dolomite-related nephrite deposit [Gil et al., 2015a,b].

Like the nephrite-forming fluids of Xiuyan and Złoty Stok, those of Dahua and Sanchakou plot in the metamorphic water field (figure 5). The δ18O values of the Dahua, Sanchakou, and Luodian nephrites are higher than others [with the exception of Tanghe], and these deposits are related to basic igneous rocks of diabase or gabbro [Zhou et al., 2006; Yang et al., 2012; Li et al., 2014; Zhang et al., 2015; Xu and Wang, 2016], which is distinct from other dolomite-related nephrites. The presence of siliceous components in the wall rocks is another common feature for these three deposits. The wall rock for Dahua nephrite is a suite of interbedded layers of calcirudite, calcarenite, and micrite mixed with laminar siliceous rocks and paramoudra (Xu and Wang, 2016). Yang et al. (2013) discussed the relationship between nephrite formation and siliceous veins in the Sanchakou deposit. The country rocks around the Luodian nephrite are siliceous clayey micrites and cherty limestones (Yang et al., 2012; Li et al., 2014). These silicalites compensate for the Si shortage during the formation of nephrite from basic rocks. For Luodian nephrite, this is supported by the δ18O equilibrium between quartz and nephrite. The δ18O Qz value of the quartz from the deposit is 22.4‰ [Yang, 2013]. Thus, the calculated δ18O Tr value for tremolite by the quartz-tremolite fractionation equation 10δ18O Tr = 2.25 × 106/T2 + 0.46 [Zheng, 1995] at 350°C equals 16.15‰, which is in the range of its nephrite δ18O value = 14.1‰–16.5‰ [Yang, 2013]. The speculation of compensation is also supported by the Si isotope fractionation between the nephrite and the siliceous veins, paramoudra, and silicalites [δ8Si = 1.1‰–1.7‰; Yang, 2013]. These values, in combination with field observations, indicate that the hydrothermal fluid forming Luodian nephrite derived from either diabase intrusion [Yang et al., 2012; Zhang et al., 2015] or seawater circulation driven by diabase intrusion [Li et al., 2014]. A comparable process occurred at Sanchakou: The water in the sediments convected with magmatic hydrothermal fluids [Zhou, 2006], or the acidic magmatic hydrothermal fluids that extracted Mg from gabbro [Yang et al., 2013] reacted with wall rocks and formed nephrite. Obviously, the hydrothermal fluids that formed these nephrites were no longer the original magmatic hydrothermal fluids, but rather the fluids that had been modified by metasomatism.

XINJIANG PLACER NEPHRITE ISOTOPES AND FLUID-ROCK REACTION

The Xinjiang placer nephrites, which are mainly dug out from paleo river beds flowing through the Take-lamagan Desert, differ from the primary ones by their wide ranges of hydrogen and oxygen isotope ratios, especially δ2H [figure 4]. There are four factors potentially influencing this difference:

- **Impurities**: Impurities may induce a conspicuously high δ2H value [Liu et al., 2016], as well as a wide range of variation.
- **Compositional effect**: Most of the placer nephrite tested featured high Fe [Liu et al., 2011b, 2016], which can result in a compositional effect on hydrogen isotope fractionations in a tremolite-H2O system [Vennemann and O’Neil, 1996].
- **Complicated derivations**: Since several primary deposits occur in the upper reaches of the Yulongkashi and Kalakashi Rivers, the placer nephrite might come from different primary deposits, even including serpentine-related nephrite [Liu et al., 2016].
- **Fluid-rock reaction**: The δ2H–δ18O trends of some of the Xinjiang placer nephrites are similar to those of the Alamasi nephrite [figure 4]. The δ18O value, which is mainly controlled by the nephrite itself [Yui et al., 1990], has remained nearly constant after nephrite formation due to its high closure temperature of 424°C [Brady, 1995].

The closure temperature can be understood as the lowest temperature of isotope diffusion or loss. That is, the δ18O value of the placer nephrite is almost equal to that of the primary nephrites. The δ2H value...
of the placer nephrite, however, can be enhanced by the reaction between meteoric water (desert water that has been fractionated by evaporation; figure 5) and rock (nephrite). The hydrogen in hydrous minerals diffuses rapidly and shows a closure temperature, below which it will no longer diffuse and change its composition, in cooling metamorphic rocks far below the formation temperature of the mineral assemblages (Graham, 1981). The closure temperature \( T_c \) for hydrogen isotope volume diffusion can be expressed as (Dodson, 1973):

\[
T_c = R/\left[ E_d T \left( \frac{\ln \left( \frac{\alpha D_0}{a^2} \right)}{\tau} \right) \right]
\]

(3)

where the time constant is

\[
\tau = \frac{-RT^2}{E_d T \frac{dT}{dt}}
\]

(4)

in which the activation energy for tremolite \( E = 71.5 \) kJ/mol (Graham et al., 1984; Farver, 2010); the gas constant \( R = 8.314 \) J/mol/K; the anisotropic factor for cylinder case \( A = 27 \) (Dodson, 1973); the pre-exponential factor in the Arrhenius relationship \( D_0 = 1.21 \times 10^{-8} \) m²/s, calculated from figure 5 of Graham et al. (1984). Thus, the closure temperature can be as low as 61°C (calculated by grain radius \( a = 0.5 \) μm, cooling rate \( dT/dt = -10^3 \) °C/d) to 123°C (calculated by \( a = 1 \) μm, \( dT/dt = -50 \) °C/d). Since the radius of nephrite tremolite can be smaller, the calculated closure temperature will decrease. Furthermore, an experiment showed that tremolite can dissolve at a pH of 7 (Diedrich et al., 2014). Grapes and Sun (2010) suggested that higher porosity created by actinolite dissolution results in an exponential increase in weathering. Tremolite fibers, with lower iron concentration than actinolite, have high chemical reactivity as well (Pacella et al., 2015). Thus, the hydrogen isotope ratio can re-equilibrate at low temperature between the placer nephrite and meteoric water, enhancing the \( \delta^2H \) value of the former.

CONCLUSIONS

On the basis of formation environment and formation process, hydrogen and oxygen isotope ratios of nephrites from around the world can be analyzed. These isotope ratios, even for oxygen alone, appear to be discrimination criteria for the geographic origin determination of dolomite-related nephrites, especially those from Vitim (Russia), Chunccheon (South Korea) and the Xinjiang Uyghur Autonomous Region and Qinghai Province of China. However, the nephrite \( \delta^{18O} \) values from Xiuyan, Dahua, and Złoty Stok overlap. The isotopic ratio differences are mainly derived from the ore-forming fluids. The isotopes of dolomite-related nephrites from Russia, South Korea, Xinjiang, and Qinghai Province increase in sequence, and the ore-forming fluids vary in the order of meteoric water → mixture of magmatic water and meteoric water → mixed water that experienced metamorphism to some extent or is even dominated by metamorphic fluid. Furthermore, the hydrogen isotope of the placer nephrite from the Hetian region of Xinjiang could have been modified by meteoric water when it was buried in paleo river beds flowing through the desert.

Based on this limited data set, we show that isotope ratio analysis is a new gem origin identification tool for gemologists studying nephrite (similar to what other researchers have shown for emerald and corundum). However, we point out with caution that more data is needed to optimize our findings.
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GEM & GEMOLOGY Summer 2020

Stable Isotope Ratios of Dolomite-Related Nephrite

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