INTRODUCTION

Mid-ocean ridge basalts (MORB), derived from partial melting of the upper mantle, are central to our understanding of the geochemistry of the mantle and the formation of the oceanic crust (e.g., Langmuir et al. 1992; Asimow et al. 2004; O’Neill and Jenner 2012; Gale et al. 2013). MORB glasses, formed by rapid cooling at the rim of pillow lavas, enable the composition of the melts to be determined (e.g., Jenner and O’Neill 2012), including volatile contents (Kendrick et al. 2013) and oxidation states (Christie et al. 1986; Bézos and Humler 2005; Cottrell and Kelley 2011, 2013; Berry et al. 2018; Zhang et al. 2018; O’Neill et al. 2018). This, in turn, allows the intensive and extensive thermodynamic conditions of the mantle source to be calculated (e.g., Asimow et al. 2004). In particular, the oxidation state of Fe in MORB, expressed as Fe\(^{3+}/\)Fe\(^{TOT}\) with Fe\(^{TOT}\) = Fe\(^{2+}\) + Fe\(^{3+}\), is important because it affects estimates of temperature, mineral assemblages, and the speciation of volatile elements, both during the production of the parental magmas in the mantle and their subsequent low-pressure evolution.

The latest average Fe\(^{3+}/\)Fe\(^{TOT}\) values of natural MORB glasses from global sources have been determined to be 0.10 ± 0.02 (Berry et al. 2018) and 0.14 ± 0.01 (Zhang et al. 2018) by Fe K-edge XANES spectroscopy, and 0.11 ± 0.02 by wet-chemistry (corrected for plagioclase phenocrysts, Bézos and Humler 2005). Wet chemistry is a destructive method that lacks spatial resolution and is unsuitable for inhomogeneous glasses or small samples such as melt inclusions (e.g., see Bézos and Humler 2005). Fe K-edge XANES spectroscopy has excellent precision and micrometer spatial resolution in two dimensions but requires glass standards that are compositionally matched and for which Fe\(^{3+}/\)Fe\(^{TOT}\) has been determined by another technique, such as \(^{57}\)Fe Mössbauer spectroscopy (e.g., Berry et al. 2008). There are also questions concerning possible photo-oxidation during the analysis of hydrous glasses (e.g., Cottrell et al. 2018). A considerable drawback for routine analysis is that it requires access to a synchrotron light source.

Raman spectroscopy is an alternative technique for determining Fe\(^{3+}/\)Fe\(^{TOT}\) that is often readily accessible, is non-destructive, has micrometer-scale spatial resolution, requires minimal sample preparation, is easy to perform, and spectra can be acquired within minutes. Previous studies have shown how Raman spectroscopy can be used for quantifying the concentration of \(\text{H}_2\text{O}\) (Thomas 2000; Zajacz et al. 2005; Behrens et al. 2006; Thomas et al. 2008; Mercier et al. 2009, 2010; Le Losq et al. 2012) and \(\text{CO}_2\) (Amalberti et al. 2011; Morizet et al. 2013) in glasses, with applications to pumices and melt inclusions (Shea et al. 2014; Métrich et al. 2016). The potential of Raman spectroscopy to determine Fe\(^{3+}/\)Fe\(^{TOT}\) in glasses has been demonstrated previ-