



## Highly CO<sub>2</sub>-supersaturated melts in the Pannonian lithospheric mantle – A transient carbon reservoir?



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### ABSTRACT

Subduction of carbonated crust is widely believed to generate a flux of carbon into the base of the continental lithospheric mantle, which in turn is the likely source of widespread volcanic and non-volcanic CO<sub>2</sub> degassing in active tectonic intracontinental settings such as rifts, continental margin arcs and back-arc domains. However, the magnitude of the carbon flux through the lithosphere and the budget of stored carbon held within the lithospheric reservoir are both poorly known. We provide new constraints on the CO<sub>2</sub> budget of the lithospheric mantle below the Pannonian Basin (Central Europe) through the study of a suite of xenoliths from the Bakony-Balaton Highland Volcanic Field. Trails of secondary fluid inclusions, silicate melt inclusions, networks of melt veins, and melt pockets with large and abundant vesicles provide numerous lines of evidence that mantle metasomatism affected the lithosphere beneath this region. We obtain a quantitative estimate of the CO<sub>2</sub> budget of the mantle below the Pannonian Basin using a combination of innovative analytical and modeling approaches: (1) synchrotron X-ray microtomography, (2) NanoSIMS, Raman spectroscopy and microthermometry, and (3) thermodynamic models (Rhyolite-MELTS). The three-dimensional volumes reconstructed from synchrotron X-ray microtomography allow us to quantify the proportions of all petrographic phases in the samples and to visualize their textural relationships. The concentration of CO<sub>2</sub> in glass veins and pockets ranges from 0.27 to 0.96 wt.%, higher than in typical arc magmas (0–0.25 wt.% CO<sub>2</sub>), whereas the H<sub>2</sub>O concentration ranges from 0.54 to 4.25 wt.%, on the low end for estimated primitive arc magmas (1.9–6.3 wt.% H<sub>2</sub>O). Trapping pressures for vesicles were determined by comparing CO<sub>2</sub> concentrations in glass to CO<sub>2</sub> saturation as a function of pressure in silicate melts, suggesting pressures between 0.69 to 1.78 GPa. These values are generally higher than trapping pressures for fluid inclusions determined by Raman spectroscopy and microthermometry (0.1–1.1 GPa). The CO<sub>2</sub>/silicate melt mass ratios in the metasomatic agent that percolated through the lithospheric mantle below the Pannonian Basin are estimated to be between 9.0 and 25.4 wt.%, values consistent with metasomatism either by (1) silicate melts already supersaturated in CO<sub>2</sub> before reaching lithospheric depths or (2) carbonatite melts that interacted with mantle peridotite to generate carbonated silicic melts. Taking the geodynamical context of the Pannonian Basin and our calculations of the CO<sub>2</sub>/silicate melt mass ratios in the metasomatic agent into account, we suggest that slab-derived melts initially containing up to 25 wt.% of CO<sub>2</sub> migrated into the lithospheric mantle and exsolved CO<sub>2</sub>-rich fluid that became trapped in secondary fluid inclusions upon fracturing of the peridotite mineral matrix. We propose a first-order estimate of 2000 ppm as the minimal bulk CO<sub>2</sub> concentration in the lithospheric mantle below the Pannonian Basin. This transient carbon reservoir is believed to be degassed through the Pannonian Basin due to volcanism and tectonic events, mostly focused along the lithospheric-scale regional Mid-Hungarian shear Zone.

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## 1. Introduction

The mantle is thought to be the largest terrestrial carbon reservoir (Dasgupta, 2013; Dasgupta and Hirschmann, 2010; Shcheka et al., 2006; Sleep et al., 2001), containing more than 90% of the Earth's carbon budget (Poinar, 2012). Many geological processes depend upon and attest to the existence of deep carbon and its exchange with other major terrestrial reservoirs, such as CO<sub>2</sub> degassing at mid-ocean ridges, CO<sub>2</sub>-rich magmatism rooted in the mantle (kimberlites, carbonatites, etc.), and diamond or carbonate-bearing xenoliths originating from the deep mantle (Hammouda, 2003 and references therein). In order to balance the continuous CO<sub>2</sub> degassing at volcanic centers, carbon must have been recycled into the mantle over geological time scales, most likely at convergent margins where carbonate-altered, sediment-bearing oceanic crust returns into the mantle (Zhang and Zindler, 1993). Carbon dioxide is known to be a dominant volatile species in magmatic fluids and gases in subduction zone environments (Anderson, 1975; Fyfe et al., 1978) where its sources include subducted carbonate sediments and carbonated oceanic crust ( $\delta^{13}\text{C} \sim 0\%$ ), organic material in marine sediments ( $\delta^{13}\text{C} < -20\%$ ) and, indigenous mantle CO<sub>2</sub> ( $\delta^{13}\text{C} = -6.5 \pm 2.5\%$ ; Sano and Marty, 1995). Several recent reviews of the carbon cycle in subduction environments (Burton et al., 2013; Dasgupta, 2013; Dasgupta and Hirschmann, 2010; Gorman et al., 2006; Jarrard, 2003; Johnston et al., 2011; Wallace, 2005) have each estimated that about half of the subducted carbon is removed from the subducting plate beneath fore-arcs or arcs and subsequently returned to the Earth's surface [40% in Gorman et al. (2006); 20–80% in Dasgupta and Hirschmann (2010); and 18–70% in Johnston et al. (2011)]. A large part of the subducted CO<sub>2</sub> therefore transits the upper mantle and, depending on residence times, this implies the possible temporary storage of a large mass of subducted carbon in the upper mantle (Kelemen and Manning, 2015). Indeed, the upper-bound estimates of Kelemen and Manning (2015) suggest that most subducted carbon is returned to the lithospheric mantle and crust, whereas previous reviews suggested that about half of it ends up being recycled into the convecting mantle. This recent study suggests that diffuse escape of CO<sub>2</sub> to the surface and, more importantly, transient storage of CO<sub>2</sub> in the back-arc lithospheric mantle may, when added to the more commonly described fluxes of arc magma and mid-ocean ridge basalt (MORB) degassing, fully balance the subducted carbon (Kelemen and Manning, 2015). The present study aims to constrain the magmatic CO<sub>2</sub> budget of the lithospheric mantle below the Pannonian Basin (PB, Central Europe) through the study of a suite of petrographically and geochemically well-studied peridotite xenoliths from the Balaton-Bakony Highland Volcanic Field (BBHVF, Szabó et al., 2010). The PB is known for its substantial mantle-derived CO<sub>2</sub> accumulations in aquifers

and petroleum reservoirs (Ballentine et al., 1991; Sherwood Lollar et al., 1997) and a complex geodynamic history including active subduction of oceanic and continental plates, formation of backarc basins (Horváth et al., 2015) and volcanic activity (Harangi and Lenkey, 2007) that have all developed within a relatively short time scale of 16 My (Kovács and Szabó, 2008). The PB represents an ideal setting for studying the fluxing of mantle CO<sub>2</sub> through the lithosphere. This study develops an innovative multi-technique approach in order to investigate the abundance and distribution of CO<sub>2</sub> fluid inclusions and vesicles within mantle xenoliths by coupling: (1) three-dimensional synchrotron X-ray microtomography, (2) nanoSIMS, Raman spectroscopic and microthermometric characterization and (3) thermodynamic modeling of composition-dependent melt CO<sub>2</sub> saturation.

## 2. Geological background

The Pannonian Basin (PB) is located in the Carpathian–Pannonian region of Central Europe. The PB is a typical inter-arc basin (Embey-Isztin et al., 2001), yet it shows a number of features that are characteristic of rift zones: high heat flow, recent alkaline volcanism, a thin crust and lithosphere, and upwelling asthenosphere (Embey-Isztin et al., 1990; Falus et al., 2007; Praus et al., 1990; Spakman, 1990). The Pannonian Basin experienced a complex geodynamic evolution from Miocene to Pleistocene time, characterized by a succession of extension-compression-extension events (Horváth et al., 2015). The major driving forces that led to the formation of the Pannonian Basin (~20 Ma) were continuous subduction and slab roll-back on its eastern boundary accompanied by synchronous eastward extrusion of the ALCAPA (Alpian-Carpathian Pannonian) blocks (Kazmer and Kovacs, 1985) from the Alpine compressional belt (Fodor et al., 1999; Horvath, 1993).

The Bakony–Balaton Highland Volcanic Field (BBHVF), on the northern shore of Lake Balaton, is situated near the center of the Carpathian-Pannonian region, in the western Pannonian Basin (western Hungary). The volcanic centers of the BBHVF were active between  $7.96 \pm 0.03$  Ma and  $2.61 \pm 0.03$  Ma (Balogh and Németh, 2005; Balogh and Pécskay, 2001; Hidas et al., 2010; Wijbrans et al., 2007) and produced mostly alkali basaltic volcanic rocks (Embey-isztin et al., 1993; Szabó et al., 1992).

Ultramafic xenoliths can be found in basanitic lava flows and pyroclastic volcanic products at six locations (Tihany, Bondoró Hill, Füzes-tó, Szentbékállá, Mindszentkállá, Szigliget). Most of the xenoliths are spinel lherzolites, but harzburgite, clinopyroxenite, orthopyroxenite, wehrlite, websterite and occasional composite xenoliths also occur (Table 1).

**Table 1**  
Petrographic description of BBHVF mantle xenoliths. H- heterogranular, E- equigranular, Lz- Lherzolite, Hzb- Harzburgite, 2ndary- trails of secondary fluid inclusions, Ol- Olivine, Opx- Orthopyroxene, Cpx- Clinopyroxene, Sp- Spinel, Amp- Amphibole, MP- Melt pocket.

Sample	Whole size	Grain size	Texture	Rock name	Fluid inclusions	Melt	Petrographic observations
SZB16	9 cm	<1 to >3 mm	H Protogranular	Lz	2 <sup>ndary</sup>	Vein	Minerals in contact with melt are destabilized
SZB44	7 cm	<1 mm	E Mosaic + Poikilitic features	Lz	2 <sup>ndary</sup>	MP	Spongy Sp at melt contact
SZB50	7 cm	~1 mm	E Mosaic	Lz	2 <sup>ndary</sup>	MP + Vein	Spongy Sp
SZB51	11 cm	1 to 5 mm	H Protogranular	Lz	2 <sup>ndary</sup>	MP	Spongy Sp
SZB52	9 cm	<1 to 5 mm	H Protogranular	Lz	2 <sup>ndary</sup>	Vein	Large vein. No mineral destabilization
SZB66	18 cm	4 to 6 mm	E Protogranular + Poikilitic features	Lz	2 <sup>ndary</sup>	Vein	Poikilitic Ol (containing Opx). Little reaction between melt and minerals
SZG14	7 cm	~1 mm	E Protogranular	Lz	2 <sup>ndary</sup>	Vein + SMI	Poikilitic Opx (containing Ol). Spongy Opx
SZG23	7 cm	<1 to 4 mm	H Protogranular	Lz	2 <sup>ndary</sup>	MP + Vein	Lot of melt and fluid percolation
SZG30	5 cm	~1 mm	E Mosaic	Lz	2 <sup>ndary</sup>	Vein	Lot of carbonate melts + fluid. Amp are destabilized
SZG44	7 cm	~1–2 mm	E Protogranular with Mosaic tendency	Lz	2 <sup>ndary</sup>	MP + Vein	Ol and Cpx are destabilized at melt contact
FT12	9 cm	~1 mm	E Mosaic	Lz	2 <sup>ndary</sup>	MP + Vein	Destabilized Amp at melt contact
FT01P	12 cm	<1 to 5 mm	H Protogranular	Hzb	2 <sup>ndary</sup>	Vein	Cryptic metasomatism. Neoblasts crystallization at grain boundaries
FT08P	9 cm	2 to 2 mm	E Tabular	Hzb	2 <sup>ndary</sup>	MP + Vein	Destabilized Amp with Cpx re-crystallization. Beginning of Opx destabilization

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