



# Decarbonation of subducting slabs: Insight from petrological–thermomechanical modeling



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## ARTICLE INFO

### Article history:

Received 3 February 2015

Received in revised form 7 July 2015

Accepted 13 July 2015

Available online 28 August 2015

Handling Editor: A.R.A. Aitken

### Keywords:

Decarbonation

Numerical modeling

Subduction

Melting

H<sub>2</sub>O–CO<sub>2</sub> fluids

## ABSTRACT

Subduction of heterogeneous lithologies (sediments and altered basalts) carries a mixture of volatile components (H<sub>2</sub>O ± CO<sub>2</sub>) into the mantle, which are later mobilized during episodes of devolatilization and flux melting. Several petrologic and thermodynamic studies investigated CO<sub>2</sub> decarbonation to better understand carbon cycling at convergent margins. A paradox arose when investigations showed little to no decarbonation along present day subduction geotherms at subarc depths despite field based observations. Sediment diapirism is invoked as one of several methods for carbon transfer from the subducting slab. We employ high-resolution 2D petrological–thermomechanical modeling to elucidate the role subduction dynamics has with respect to slab decarbonation and the sediment diapirism hypothesis. Our thermodynamic database is modified to account for H<sub>2</sub>O–CO<sub>2</sub> binary fluids via the following lithologies: GLOSS average sediments (H<sub>2</sub>O: 7.29 wt.% & CO<sub>2</sub>: 3.01 wt.%), carbonated altered basalts (H<sub>2</sub>O: 2.63 wt.% & CO<sub>2</sub>: 2.90 wt.%), and carbonated peridotites (H<sub>2</sub>O: 1.98 wt.% & CO<sub>2</sub>: 1.50 wt.%). We include a CO<sub>2</sub> solubility P–x[H<sub>2</sub>O wt.%] parameterization for sediment melts. We parameterize our model by varying two components: slab age (20, 40, 60, 80 Ma) and convergence velocity (1, 2, 3, 4, 5, 6 cm year<sup>−1</sup>). 59 numerical models were run and show excellent agreement with the original code base. Three geodynamic regimes showed significant decarbonation. 1) Sedimentary diapirism acts as an efficient physical mechanism for CO<sub>2</sub> removal from the slab as it advects into the hotter mantle wedge. 2) If subduction rates are slow, frictional coupling between the subducting and overriding plate occurs. Mafic crust is mechanically incorporated into a section of the lower crust and undergoes decarbonation. 3) During extension and slab rollback, interaction between hot asthenosphere and sediments at shallow depths result in a small window (~12.5 Ma) of high integrated CO<sub>2</sub> fluxes (205 kg m<sup>−3</sup> Ma<sup>−1</sup>).

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## 1. Introduction

Subduction zones are geologically complex systems where rocks are subducted and recycled, partial melting and metamorphic devolatilization of the slab and mantle wedge take place, and mantle heterogeneities arise as they interact with these fluids (Schmidt and Poli, 1998; Poli and Schmidt, 2002; Stern, 2002; Grove et al., 2012; Kawamoto et al., 2012). Slab derived volatiles, primarily H<sub>2</sub>O ± CO<sub>2</sub> fluids, are a critical component to subduction processes and evolution. Fluid fluxes and resulting interactions within the wedge are shown to rheologically weaken depleted peridotites (Hirth and Kohlstedt, 1996), decouple the slab from an overriding plate and reduce friction (Gerya and Meilick, 2011), decrease solidus temperatures resulting in flux melting (Iwamori, 1998; Schmidt and Poli, 1998), and decrease density and viscosity of the mantle wedge (Gerya et al., 2002; Honda and Saito, 2003; Arcay et al., 2005; Gorczyk et al., 2007a; Iwamori, 2007). H<sub>2</sub>O in the subduction environment has been well

studied experimentally (e.g., Grove et al., 2012 and references therein; Tatsumi et al., 1986; Schmidt and Poli, 1998) and numerically (e.g., Peacock, 1990; Davies and Stevenson, 1992; Gerya and Yuen, 2003a; Arcay et al., 2005; Gorczyk et al., 2007a; Faccenda, 2014) owing to its significant effect on physicochemical properties, while CO<sub>2</sub>'s role in the subduction environment is open for much scientific study (Tumiati et al., 2012; Dasgupta et al., 2013; Manning et al., 2013).

H<sub>2</sub>O enters the subduction zone primarily hosted within clays, zeolites, pore fluids, and structurally bound minerals and is released at deeper depths (Plank and Langmuir, 1998; Schmidt and Poli, 1998). In contrast, CO<sub>2</sub> is primarily found within carbonates, organic material (as carbon), and cements (Staudigel et al., 1989; Alt and Teagle, 1999; Alt et al., 2013). Both H<sub>2</sub>O and CO<sub>2</sub> enter the trench within lithologies of metasediments, altered basalts, and extend as far as upper parts of the serpentinized mantle lithosphere (Staudigel et al., 1989; Rudnick and Fountain, 1995; Plank and Langmuir, 1998; Alt and Teagle, 1999; Poli and Schmidt, 2002; Jarrard, 2003). Rates of fluid release and retention in mineral bound phases (nominally anhydrous or carbonates) to deeper depths are highly dependent on the thermal regime, composition, extent of alteration, and dynamics of the subducting slab itself

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(van Keken et al., 2002; van Keken et al., 2011; Faccenda, 2014). H<sub>2</sub>O is considered to be the most mobile fluid and reaches peak devolatilization rates at ca. 2.4–2.8 GPa (~70–80 km) in depth (Poli and Schmidt, 2002; van Keken et al., 2011). Additionally, when carbonate is included to subducting lithologies, it affects the stability of hydrous phases and aids in the generation of new ones. An example occurs in the hydrous phase lawsonite, where its thermal stability field is expanded by 30 °C at pressures >1.5–2.0 GPa as a consequence of Ca–Mg partitioning between garnets and carbonate (Molina, 2000; Poli et al., 2009). Paragonite, another hydrous phase, forms due to plagioclase breakdown which results in delayed omphacite formation to ~2.0 GPa (Schmidt and Poli, 2014). Furthermore, van Keken et al. (2011) and Faccenda et al. (2012) have demonstrated that nearly one third of the water content potentially remains in the slab beyond subarc depths as a function of thermal structure, seismic in-pumping, and OH<sup>−</sup> stored in nominally anhydrous minerals. However, prograde metamorphism of subducting oceanic crust yields a series of fluids or melts through mineralogical reactions that inevitably result in eclogites that may or may not contain hydrous phases and/or carbonates. Alternating continuous and discontinuous reactions causes devolatilization, yielding a mobile phase. This mobile phase may be a low-density fluid, a high density solute-rich fluid, a silicate melt, or a carbonatite melt.

Volcanic gas monitoring and melt inclusions attest to the presence and transfer of carbon during subduction into the overlying mantle wedge. CO<sub>2</sub> is the second most abundant volcanic gas with concentrations averaging between 2500 and 7600 ppm (Fischer and Marty, 2005; Wallace, 2005) in arcs and reaching maximum concentrations up to 0.4–1.7 wt.% (based on Mount St. Helens melt inclusions Blundy et al., 2010). Furthermore, Sano and Marty (1995) show that sediments, carbonate limestones, and organic matter contribute the majority of carbon output in arc volcanism with the remaining 20% of carbon derived from a MORB-type source. In addition, field observations in HP–UHP terranes demonstrate the presence of carbonic fluid flow across the slab–mantle wedge interface, where carbonated peridotites and eclogites as well as microdiamond and graphite are detected (van Roermund et al., 2002; Crespo et al., 2006; Zhang et al., 2007; Scambelluri et al., 2008; Sapienza et al., 2009; Mposkos et al., 2010; Scambelluri et al., 2010; Frezzotti et al., 2011; Luque et al., 2013).

In contrast, studies of decarbonation reactions show that the amount of fluid released as a function of *P–T* tends to decrease CO<sub>2</sub> in the system as it is slowly partitioned into carbonate minerals (i.e., dolomite and magnesite). Carbonate partitioning occurs at pressures >2 GPa with minor decarbonation at forearc depths derived primarily from subducting oceanic sediments (Molina, 2000; Kerrick and Connolly, 2001a; Connolly and Kerrick, 2002; Dasgupta et al., 2004; Gorman et al., 2006; Poli et al., 2009). Therefore, according to both thermodynamic (Kerrick and Connolly, 1998, 2001a; Connolly and Kerrick, 2002; Connolly, 2005; Gorman et al., 2006) and experimental investigations (Molina, 2000; Thomsen and Schmidt, 2008; Poli et al., 2009; Tsuno and Dasgupta, 2010; Grassi and Schmidt, 2011) decarbonation at subarc depths should be limited. Subsequently, a calculated carbon deficit exists between the amount of carbon recycled and the amount of carbon returned to the surface via degassing. Four primary mechanisms are hypothesized to reconcile the imbalance of carbonic fluid transfer into the wedge and are detailed in a review by Dasgupta (2013). A summary of mechanisms is as follows: (1) Rayleigh–Taylor instabilities, where a hydrated subducting mélange may detach from the slab and rise into the convecting wedge (Gerya and Yuen, 2003b; Gerya et al., 2006; Tumiaty et al., 2012), (2) sediment diapirs driven solely on density contrast without the aid of hydration (Currie et al., 2007; Behn et al., 2011), (3) carbonate dissolution at high pressures (Caciagli and Manning, 2003; Dolejš and Manning, 2010; Frezzotti et al., 2011; Kawamoto et al., 2013; Ague and Nicolescu, 2014) and (4) infiltration driven decarbonation (Kerrick and Connolly, 2001a; Connolly, 2005; Gorman et al., 2006).

In this paper we introduce a new binary fluid component (H<sub>2</sub>O–CO<sub>2</sub>) into a geodynamic modeling code, I2VIS (Gerya and Yuen, 2003a), to explore how dynamic processes aid in decarbonation. We accomplish this by incorporating thermodynamic modeling (Connolly, 2005) that accounts for carbonated rock compositions entering the trench coupled to a *P–x*[H<sub>2</sub>O wt.%] parameterization for CO<sub>2</sub> solubility (Duncan and Dasgupta, 2014) in partially molten sediments. We apply this new method to a series of numerical tests investigating geodynamic influences a binary fluid has on the dynamics of the subduction system in addition to a H<sub>2</sub>O fluid. We quantify decarbonation of subducting lithologies, assess the sediment plume (molten and density driven) hypothesis as a viable mechanism for CO<sub>2</sub> transfer, and parameterize subduction conditions where decarbonation processes can be physically attainable.

## 2. Methods

### 2.1. Petrological model

This work's petrologic model follows that of previously coupled petrological–thermomechanical modeling paradigms (e.g., Petrini et al., 2001; Gerya and Yuen, 2003a; Gerya et al., 2006; Gorczyk et al., 2007a). This work differs in the following: additional solid-solution models (Table 2) and bulk oxide compositions (Table 1) are included to account for carbonated lithologies entering the subduction trench. Bulk oxide compositions representative of their altered rocks of basalts, gabbros, peridotites, and sediments are taken as averages (Hart and Zindler, 1986; Staudigel et al., 1989; Plank and Langmuir, 1998; Alt and Teagle, 1999; Bach et al., 2001; Poli and Schmidt, 2002; Jarrard, 2003) since they may be vertically and horizontally heterogeneous due to extraneous factors (e.g., age dependence, plate spreading rates, and oceanic environmental conditions).

Sediments and altered basalts are the most well constrained compositions entering the trench. Sediments are modeled after GLOSS average composition (Plank and Langmuir, 1998) and altered basalt compositions are averaged after ODP legs (Staudigel et al., 1989; Alt and Teagle, 1999). Lower oceanic crust with more H<sub>2</sub>O does contain some carbonate material, typically at slow spreading centers (Bach et al., 2001). Despite this evidence, we consider the CO<sub>2</sub> wt.% to be negligible in the lower oceanic crust as it appears to be an exception. The oceanic mantle lithosphere is also considered to be anhydrous and carbonate free. However, alteration of the upper layer of the oceanic mantle lithosphere may occur, giving rise to bulk compositional changes and increases in volatile concentrations. Processes that can participate in alteration of the uppermost layer are the development of normal faulting in the slab bending region that pump water further down (Faccenda et al., 2012; Faccenda,

**Table 1**

New and old (H<sub>2</sub>O only) bulk oxide wt.% compositions for representative subducting lithologies. Sediments are GLOSS average sediments (Plank and Langmuir, 1998), metabasalts taken from Staudigel et al. (1989) and Gerya et al. (2006), and mantle values taken as LOSIMAG after Hart and Zindler (1986). LOSIMAG composition altered to account for CO<sub>2</sub>.

Oxides	Sediments		Oceanic Crust		Mantle	
	New	Old	New	Old	New	Old
SiO <sub>2</sub>	58.57	61.10	46.6	47.62	44.05	45.55
Al <sub>2</sub> O <sub>3</sub>	11.91	12.43	15.26	14.48	4.03	4.03
FeO	5.21	5.43	9.84	10.41	7.47	7.47
MgO	2.48	2.59	6.54	6.92	37.42	37.42
CaO	5.95	6.21	12.66	13.39	3.18	3.18
Na <sub>2</sub> O	2.43	2.54	2.03	2.15	0.33	0.33
K <sub>2</sub> O	2.04	2.13	0.55	0.58	0.03	0.03
H <sub>2</sub> O	7.29	7.60	2.63	2.78	1.98 (max)	1.98 (max)
CO <sub>2</sub>	3.01	–	2.9	–	1.5 (max)	–

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