Constraining past environmental changes of cold-water coral mounds with geochemical proxies in corals and foraminifera

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Abstract
Cold-water coral (CWC) reefs and mounds are and have been biodiversity hotspots of the deep sea. As their occurrence depends on specific environmental parameters, gaining hindsight on changing ocean conditions under on-going climate change is the key to a better understanding of CWC mound development through time. A convenient technique for reconstructing the palaeoenvironment during periods of CWC mound growth is by extracting geochemical proxies from biologically mediated carbonates. Here, the focus is on probably the two most abundant calcareous archives, that are, cold-water Scleractinia and Foraminifera, with an overview of the geochemical proxies (selection) used in these aragonitic and calcitic skeletons from CWC mounds. A particular emphasis is set on constraining proxies for temperature, salinity, seawater density, seawater carbonate systems parameters (pH, CO₃²⁻), nutrients, oxygen and water mass tracers.

KEYWORDS
Carbonate mounds, cold-water corals, deep-sea, Desmophyllum pertusum, intermediate water masses, palaeoceanography

1 | ENVIRONMENTAL CONTROL OVER COLD-WATER CORAL MOUNDS

Cold-water coral (CWC) reefs and carbonate build-ups are abundant along the continental margins of the North and South Atlantic Ocean (Roberts et al., 2009), the Mediterranean (Taviani et al., 2005; Freiwald et al., 2009) and Caribbean Seas (Reyes et al., 2009), and in the Gulf of Mexico (Newton et al., 1987; Davies et al., 2010; Hebbeln et al., 2014). Some rare occurrences have been also reported from the Indian Ocean (e.g. Maldives, Reolid et al., 2017) and in the NE Pacific (Hyland et al., 2005). The main constructors of these structures and the most abundant species at shallow and intermediate water depths (200–1200 m) are the framework-building CWCs Desmophyllum pertusum (formerly known as Lophelia pertusa, Addamo et al., 2016) and Madrepora oculata, which provide a habitat for many other organisms living within and around these reefs.

Several studies have already reviewed and summarized important biotic and abiotic parameters that control the distribution and development of CWC reefs and mounds (Freiwald et al., 2004; Wheeler et al., 2007; Roberts et al., 2009; Wienberg and Tischack, 2017; Hebbeln et al., 2019). Essential environmental controlling parameters are temperature, salinity, the deduced seawater density and dissolved oxygen (O₂) which characterize water masses and their circulation (Freiwald, 2002; Dorschel et al., 2005; Roberts et al., 2006; Rüggeberg et al., 2007; Wienberg et al., 2010; Fink et al., 2012). Important for the early (larval) settlement...
and further growth of CWCs are nutrition and the seawater carbonate parameters, that is, pH, alkalinity, carbonate ion concentration, or dissolved inorganic carbon (DIC; Maier et al., 2011; Form and Riebesell, 2012; McCulloch et al., 2012; Flögel et al., 2014). Over the past two decades, the typical values of environmental parameters in which CWC reefs and mounds thrive have been continuously updated. Table 1 summarizes the total ranges as well as the typical values for *D. pertusum* (and *M. oculata*) dominated reefs in the (North) Atlantic Ocean.

Several provinces have been discovered along the Atlantic margins, where CWC reefs are able to build carbonate mound structures as high as 350 m (Figure 1). In addition to parameters described above, sustained CWC reef development is (a) strongly dependent on the sediment supply and the sediment baffling capacity of the framework-building coral reefs (Wienberg and Titschack, 2017), (b) related to climate cycles (Frank et al., 2011) influencing intermediate water mass circulation and nutrient supply (White, 2007) and (c) influenced by hydrodynamics (Mienis et al., 2012; Cyr et al., 2016; Mohn et al., 2014; van Haren et al., 2014).

In this review the focus is on those environmental parameters which play a significant role in the build-up of large CWC mounds, and which can be reconstructed back in time applying well-established and newly developed geochemical proxies. A complex issue of geochemical proxy calibration and application is the effect of physiological adaption of the individual species to changing environmental conditions. This may affect elemental incorporation and isotope fractionation during biomineralization. This topic is not the principal focus of this review but will be emphasized in future studies. Furthermore, only a selection of geochemical proxies will be discussed:

| Parameter | Atlantic range | NE Atlantic range |
|-----------|----------------|------------------|
| Temperature (°C) | 4–14 | 5.9–10.6 |
| Salinity (PSU) | 35–38.8 | 35–36 |
| Seawater density (kg/m³) | 27.2–27.7 | 27.35–27.65 |
| Dissolved oxygen (ml/L) | (0.5) 2.6–7.2 | 3–6.7 |
| pH | 7.86–8.3 | 7.92–8.3 |
| Ωaragonite (mmol/kg) | 1.35–4.06 | 1.35–3.03 |
| TA (µmol/kg) | 2259–2742 | 2287–2377 |
| DIC (µmol/kg) | 2088–2349 | 2088–2186 |

These proxies have been developed for both calcitic and aragonitic biogenic carbonates where there may be different designated uses (i.e. Sr/Ca used for temperature reconstructions in aragonitic coral skeletons, for growth and calcification rates in calcitic coccolithophorids, or for temperature, salinity, seawater pH and carbonate ion concentration in calcitic foraminifera; see below). The calibration of proxies to *in situ* measured environmental parameters was tested on specimens collected live, mostly *D. pertusum, M. oculata* and *Desmophyllum dianthus*, a solitary CWC with a larger depth range (see also review by Robinson et al., 2014). Furthermore, benthic foraminifera are widely and more commonly used in the fossil record to reconstruct past environmental conditions. Both archives are discussed in more detail below, together with how they are used to decipher the history of CWC mounds.

### 2 Environmental Parameters Recorded by Geochemical Proxies in Calcareous Organisms from CWC Mounds

#### 2.1 Temperature

**2.1.1 Stable oxygen isotopes**

The δ¹⁸O signature of marine carbonates has long been used to reconstruct past ocean temperatures (Epstein et al., 1953),...
to infer changes in continental glaciation (Shackleton, 1974) and, coupled with another temperature proxy such as Mg/Ca ratios in foraminifera, to track relative changes in seawater salinity (Pahkle et al., 2003; Nürnberg and Groeneveld, 2006; Bahr et al., 2011). However, many carbonates exhibit consistent offsets from isotopic equilibrium due to the so-called vital effects. For example, the calcitic benthic foraminifera genus Cibicidoides (epibenthic) has a consistent offset between 0.64–0.83‰ (Shackleton and Opdyke, 1973). Such offsets appear to be more complex in scleractinian CWCs as they are highly variable (Smith et al., 2000; Adkins et al., 2003) and vary strongly with the coral microstructure (Rollion-Bard et al., 2010; Marali et al., 2013). Different models have been proposed to explain these large biologically induced spreads in stable oxygen and carbon isotopes, such as sulphate polysaccharides in the centre lines of calcification (COC; Cuif and Dauphin, 1998), the presence of organic matrix (Johnston, 1980), hydration and hydroxylation (McConnaughey, 1989) or the existence of a biologically induced pH gradient (Adkins et al., 2003). Yet, studies have found a strategy to use coral δ18O and δ13C to extract the temperature signal—the so-called ‘lines technique’ (Smith et al., 2000). In particular, Smith et al. observed that the heavy end of the strong δ18O–δ13C relationship points towards isotopic equilibrium (Figure 2). The actual approach is that, if the δ13C at equilibrium is known (either from seawater or coeval

**FIGURE 1** Global distribution of cold-water coral mound provinces (stars, after Henriet et al., 2014; Hebbeln and Samankassou, 2015, and references therein, with additional findings in the Pacific off the Olympic Coast: Hyland et al., 2005 and new discoveries at the Maldives: Reolid et al., 2017, Namibia: Hebbeln et al., 2016, and Ghana: Buhl-Mortensen et al., 2017) on the background map of CWC occurrences of reef-forming Scleractinia, Antipatharia, Zoanthidae, and Pennatulacea (from Freiwald et al., 2017)

**FIGURE 2** Schematic example of the lines technique by Smith et al. (2000). δ18O versus δ13C relationship trendline (black) of any scleractinian cold-water coral. Two different scenarios are depicted. Scenario 1: δ13CDIC value is known: the corresponding intercept value results in a δ18O value which can be calculated into seawater temperatures. Scenario 2: δ13CDIC value is not known and a global ocean mean value of 0‰ is assumed: the corresponding intercept value leads to a different δ18O value. This difference was shown by the study of Marali et al. (2013) to be on average 1.4°C warmer. This offset however does not include the actual offset to the expected isotopic equilibrium. (For further details see text)
benthic foraminifera), seawater temperature can be calculated. However, Smith et al. (2000) also showed that even if δ13C at equilibrium is unknown (assuming δ13C = 0‰), the intercept of δ18O–δ13C revealed a similar temperature relationship as those experimentally derived by Grossman and Ku (1986). Nevertheless, as a palaeotemperature proxy CWC δ18O exhibits several disadvantages: (a) the metabolic effect of each individual coral might have changed through time, (b) the δ13C of the ambient seawater may also have been different in the past and (c) contrasting results yielding errors in temperature reconstructions between ±1.5 to ±2.95°C have been published (Lutringer et al., 2005; Marali et al., 2013). Such a large error may not be limiting at shallower depth, such as upper intermediate water masses, but certainly at greater depth and more stable environments. It is therefore suggested that future studies should aim at understanding oxygen and carbon isotope fractionation in more detail in order to constrain oxygen isotopic equilibrium in scleractinian CWCs. One possibility to tackle this research question might be to use coupled clumped (see Section 2.1.2) and triple oxygen isotopes. The three-isotope method including clumped isotopes (Δ47 and Δ48) may be a promising tool in order to calibrate equilibrium fractionation factors of the seawater-carbonate system (Bao et al., 2016 and references therein; Fiebig et al., 2019).

2.1.2 Clumped isotopes

Clumped isotopes are the study of not only the isotopic ratio of 13C/12C and 18O/16O, but also the clumping of the atoms that are joined together in the same carbonate ion group (e.g. 13C18O16O2−). Carbonates may contain up to 20 different isotopologues (Eiler et al., 2007, 2011; Ghosh et al., 2007), of which 13C18O16O2− is the most abundant doubly substituted.

Clumped isotopes measure the isotopologue with a mass of 47 and contain the two heavy rare isotopes 13C and 18O. The amount of clumping is controlled by the law of thermodynamics independently of the δ18O of seawater. This means that as temperatures increase, the clumping decreases and will exhibit a purely stochastic distribution at temperatures >1,000°C. In 2006, the study by Ghosh et al. (2007) was the first to demonstrate that this theoretical assumption holds true for a suite of different marine carbonates implying that clumped isotopes were free of any kinetic or so called ‘vital effects’. However, even though the study of clumped isotopes is a relatively new field, the scientific community is growing and so is our knowledge. More and more studies reveal that Δ47 values are in disequilibrium (Bajnai et al., 2018), as for example for CWCs (Thiagarajan et al., 2011; Kimball et al., 2016; Spooner et al., 2016). To date, there only exists one study with reconstructed seawater temperatures using Δ47 values from CWC (Thiagarajan et al., 2014). As suggested by Spooner et al. (2016), clumped isotopes also need species-specific calibrations similar to elemental/Ca ratios in foraminifera (Regenberg et al., 2009) and validation with, for example, Li/Mg ratios (see below, Montagna et al., 2014) in order to establish this tool in CWCs. In benthic foraminifera a combination of Mg/Ca ratios and Δ17 may be used for bottom water temperature (BWT) reconstructions (Breitenbach et al., 2018).

2.1.3 Sr/Ca in Scleractinia

Besides stable oxygen isotopes, Sr/Ca ratios belong to the most widely used proxy to reconstruct past oceanic temperatures in tropical coral skeletons (Weber, 1973; McCulloch et al., 1994; Felis et al., 2012). Strontium incorporation into coral skeletons decreases with increasing seawater temperatures with a sensitivity of ~0.1 mmol/mol per °C (Corrêa, 2006). As the distribution coefficient of Sr into coral aragonite is close to 1, a fundamental assumption is that the past Sr2+/Ca2+ seawater value has not changed through time. Similar to Li, Mg and Ca it has been shown that seawater Sr/Ca ratios changed over long time periods (Coggon et al., 2010), on glacial/interglacial timescales by 5%–10% (Stoll et al., 1999) and may even vary by location by up to 0.1 mmol/mol (Shen et al., 1996; De Villiers, 1999). This phenomenon is intriguing as Sr has a residence time of 2–4 Myr (Hodell et al., 1990) and therefore its concentration and isotopic composition should be well mixed in the oceans. This should be further investigated in detailed studies.

Besides the many Sr/Ca to sea surface temperature calibrations, only two studies tried to calibrate Sr/Ca to BWTs in CWC skeletons (Cohen et al., 2006; Raddatz et al., 2013). Both studies reveal significant limitations for the use of this proxy in CWC mound settings. In particular, the study by Cohen et al. (2006) investigated the Sr/Ca variability within one single specimen of D. pertusum using secondary ion mass spectrometry and found a temperature sensitivity twice as strong as that of tropical corals, with 0.18 mmol/mol per °C. However, Raddatz et al. (2013) examined a suite of D. pertusum specimens from seawater temperatures between 6 and 14°C along the European continental margin and found a similar temperature sensitivity to that exhibited by tropical corals (0.1 mmol/mol°C). Yet, the scatter (>0.3 mmol/mol) in the calibration slope limits the use of CWC Sr/Ca ratios as a palaeotemperature proxy. Interestingly, heterogeneity profiles within different CWCs did not reveal such variability in, for example, the Mg/Ca or Li/Ca ratios (Sinclair et al., 2006; Gagnon et al., 2007; Raddatz et al., 2013). This suggests that Sr/Ca ratios in CWCs are controlled by environmental parameters other than seawater temperature. Future studies should therefore aim at investigating Sr/Ca ratios at
high resolution within a suite of CWC samples from different oceanographic settings with contrasting seasonal temperature variations and, if possible, with different Sr/Ca seawater values. Furthermore, an improvement of this proxy could be made by performing Sr/Ca analyses on temperature-controlled CWCs.

2.1.4 Mg/Ca ratios in calcitic foraminifera

In the 1970s, studies showed that tropical marine carbonates appear to be enriched in Mg compared to high latitude marine calcite (Katz, 1973; Savin and Douglas, 1973). The first important group of organisms studied for their Mg contents were marine protozoa (foraminifera). These studies resulted in a well-defined, species-specific T-calibration utilizing the Mg/Ca ratios in the tests of cold-water planktonic foraminifera Neogloboquadrina pachyderma from the North and South Atlantic (Nürnberg, 1995) and the tropical planktonic species Globigerinoides sacculifer from cultivation studies (Nürnberg et al., 1996). The Mg$^{2+}$ uptake involves probably both (a) a changing distribution coefficient $D = \frac{Mg/Ca_{carbonate}}{Mg/Ca_{seawater}}$ and (b) a physiological effect. Subsequently, many laboratories further established this tool as a palaeothermometer (Rosenthal et al., 1997; Lear et al., 2002; Anand et al., 2003; Marchitto et al., 2007; Bryan and Marchitto, 2008; Regenberg et al., 2009; Raddatz et al., 2017; among others). Benthic organisms that exhibit a Mg/Ca ratio that is temperature dependent are calcitic foraminifera (Rosenthal et al., 1997; Lear et al., 2002; Bryan and Marchitto, 2008), brachiopods (Brand et al., 2013) and ostracods (Dwyer et al., 1995). In contrast to planktonic foraminifera (Anand et al., 2003), the temperature dependency of benthic organisms is relatively small. Most of the calibration studies suggest an exponential relationship between temperature and Mg/Ca ratios (Elderfield and Ganssen, 2000; Anand et al., 2003; Regenberg et al., 2009), whereas other studies exhibit simple linear relationships in benthic foraminifera (Rosenthal et al., 1997; Quillmann et al., 2012).

On average, the Mg/Ca ratio of benthic foraminifera at 2°C is about 1 mmol/mol with a temperature sensitivity of 10%–15% per °C (Elderfield et al., 2006; Sadekov et al., 2014). Furthermore, the Mg incorporation into benthic foraminifera is also controlled by the carbonate ion saturation resulting in 1.5°C per 10 μmol/kg, which requires additional proxies for the carbonate system (e.g. B/Ca or Sr/Ca, see Section 2.4). Overall, all calibrations provide an accuracy of −1 to 1.5°C and thus allow for reconstruction of temperature dynamics at a carbonate mound at intermediate ocean depths.

Many palaeoceanographic reconstructions suffer from calcite dissolution leading to a preferential dissolution of Mg-enriched calcite. It has been shown that removal of Mg occurs below the critical threshold of ΔCO$_3^{2−}$ between 18 and 26 μmol/kg (Regenberg et al., 2009; Regenberg et al., 2014). Although there are some approaches to correct for this effect, it is unclear to what extent ΔCO$_3^{2−}$ has changed through time. In particular, if both carbonate polymorphs are in the same mound, the saturation state should always be defined. Although many studies applied the Mg/Ca ratio as a temperature proxy, only one has used this proxy in CWC mound settings to reconstruct BWTs (Matos et al., 2015). According to this study, there is not a single core-top study available (planktonic or benthic) on foraminiferal Mg/Ca ratios from CWC carbonate mounds. Therefore, absolute temperature reconstructions based on foraminiferal Mg/Ca ratios should proceed with caution.

The only dataset currently published is an example from the eastern North American continental margin which does not originate from the CWC mound sediments directly, but from an off mound sediment core (Matos et al., 2015). Here, the authors showed that Mg/Ca ratios measured in the test of the benthic foraminifera Cibicidoides pachyderma from Holocene Cape Lookout CWC mounds resulted in a relatively narrow range of BWTs between 6.5 and 7.5°C. Nevertheless, in situ BWTs show highly variable temperature conditions (Stefánsson et al., 1971; Mienis et al., 2014), which are only partially reflected in the reconstructed BWT (Matos et al., 2015).

2.1.5 Li/Mg ratios in Scleractinia

Mg/Ca ratios have not only been studied in calcite builders but also, for example, in aragonitic foraminifera (Hoeglundina elegans, Bryan and Marchitto, 2008) as well as Scleractinia (Mitsugushi et al., 1996; Cohen et al., 2006). However, recent findings could not reproduce a single temperature relationship in Scleractinia (Case et al., 2010; Hathorne et al., 2013; Raddatz et al., 2013; Montagna et al., 2014), but made another interesting observation: based on the pioneering study of Marriot et al. (2004a, b), Li/Ca ratios in biogenic (corals, foraminifera) and inorganic carbonate decrease with increasing temperature by 4% per °C, making this a very powerful tool.

However, different calibration offsets between brachiopods, scleractinian corals and inorganic calcite suggest a strong biological control on Li incorporation. A first hypothesis to overcome this was made by Bryan and Marchitto (2008) by combining Li/Ca with Mg/Ca ratios to Mg/Li ratios in *H. elegans*, which resulted in a strong low-scattered linear correlation between temperature and the Mg/Li ratio. Currently there are only a few different linear and exponential calibration lines for different Scleractinia (Case et al., 2010; Hathorne et al., 2013; Raddatz et al., 2013; Montagna et al., 2014). The latter study introduced the Li/Mg ratio (the lower concentrated element in the first place). A fundamental issue with calibrating Mg/Ca or Li/Ca ratios in scleractinian CWCs
is the sample heterogeneity revealing a two to threefold change within the COC similar to that observed for stable oxygen isotopes (Rollion-Bard et al., 2010). By combining Mg/Ca with Li/Ca ratios this variability is reduced, suggesting that both elements may have been controlled by a similar vital effect (Montagna et al., 2014; Marchitto et al., 2018). In particular, the calibration by Montagna et al. (2014) involves scleractinian CWCs, temperate corals as well as tropical corals, and thus appears to be the most suitable to reconstruct temperatures from high to very low temperatures. However, a few points should be considered when using Li/Mg ratios as a palaeotemperature proxy:

1. Although the calibration by Montagna et al. (2014) is the most suitable for CWC carbonate mound settings, future studies should aim at extending this with more samples around 4°C, particularly because the Li/Ca—temperature relationship is exponential and thereby exhibits a larger uncertainty at lower temperatures.

2. The CWC samples should be screened for the amount of COC-like structures as these could alter the resulting Li/Mg-based BWTs.

3. In order to identify possible diagenetic overprints, which may be not resolvable with XRD scanning, the Mg/Ca and Li/Ca ratios should always exhibit a positive linear relationship (Case et al., 2010; Raddatz et al., 2013, 2014a, 2016; Montagna et al., 2014).

4. Magnesium, Li and Ca have relatively long residence times in the ocean of 14 Myr, 1.2 Myr and 1 Myr, respectively, which is considerably longer than the ocean mixing time of ~1,000 yr, guaranteeing vertically and horizontally well-mixed concentrations (relative to salt) and isotopic compositions (Li, 1982). However, there is evidence that the Mg/Ca (today 5.1 mol/mol) and Li/Ca (0.4 mmol/mol) ratio of seawater has changed by a factor of 2–3 over the last 5 Myr (Fantele and DePaolo, 2005, 2006; Hathorne and James, 2006), implying severe consequences on directly reconstructed Mg/Ca-based seawater temperatures (Evans et al., 2015, 2016). Calculating Li/Mg ratios not only reduces the vital effect on each element within the coral skeleton, it also holds promise to level out any secular seawater Mg/Ca and Li/Ca changes as these appear to be controlled by similar processes in seawater. However, applying these proxies to considerably older settings could lead to incorrect reconstructions since secular element seawater changes may significantly alter the Mg/Ca and Li/Ca ratios in the coral. This needs to be further refined in cultivation studies similar to those of Evans et al. (2016), as the distribution of Li into calcite or aragonite with changing seawater Li/Ca ratios is not well-constrained.

5. Cleaning procedures should guarantee organic matter loss, as this can significantly alter the element concentration of the measured sample (Hathorne et al., 2013).

There are existing studies that apply the Li/Mg ratio to reconstruct BWTs in CWC settings (Montero-Serrano et al., 2013; Raddatz et al., 2014b; Raddatz et al., 2016; Bonneau et al., 2018). The Li/Mg-based BWT reconstruction from the Challenger Mound IODP Site U1317 on D. pertusum suggest that the Challenger Mound might have been growing in a relatively narrow temperature envelope between 8 and 10°C throughout the last 3 Myr (Raddatz et al., 2014b). However, the relatively poor sample resolution of this record may not capture its entire variability. Furthermore, on other time scales Li/Mg-based BWT reconstructions in CWC reef/mound settings may also capture other processes. For example, the very detailed study by Montero-Serrano et al. (2013) shows, by the use of a suite of geochemical proxies and Li/Mg ratios, that intermediate water masses in the Bay of Biscay appear to be controlled by the North Atlantic Oscillation (NAO). Interestingly, the BWT range is no larger than 2°C, implying again that CWC mound growth at the mid-latitudes is restricted to relatively narrow environmental boundary conditions. At higher latitudes (71°N), Li/Mg-based BWT reconstruction reveals a slightly larger range of up to 6°C, with temperatures between 4 and 10°C throughout the Holocene (Raddatz et al., 2016). Here, the larger range of BWT may have been recorded due to the resilience of CWC subject to ocean acidification and to a potentially higher sea-surface productivity (Titschack et al., 2015; Raddatz et al., 2016). Considering BWT reconstructions from CWC mounds so far, they exhibit a systematic picture with greater ΔBWT (overall BWT variability throughout the record) at shallower depth (Figure 3). This pattern appears to be logical as the BWT variability at shallower depth/closer to the surface/seasonal thermocline is expected to be larger. Although this observation raises concerns about the temperature control on CWCs, it also highlights the potential for using CWC skeletons from various regions to reconstruct changes in thermocline variability through time. A step forward for future studies would be to combine different archives (foraminifera, corals) with different or same temperature proxies to fully capture the entire temperature variability.

2.2 Salinity

2.2.1 Coupled Mg/Ca and δ18O

A frequently used method to reconstruct relative changes in seawater salinity is the coupling of Mg/Ca ratios and δ18O (Elderfield and Ganssen, 2000; Nürnberg, 2000). Here, Mg/Ca ratios (for T) are measured on the same biotic carriers used for δ18O, which avoids any differences in seasonality and/or habitat introduced by using different faunal groups or archives. Furthermore, the magnitude and timing of determined seawater temperature and δ18O changes not only allows us to understand temperature changes in relation to continental ice sheet dynamics, but also constrains seawater
FIGURE 3 Range of reconstructed (and observed) bottom-water-temperatures (BWT) in CWC mounds plotted against water depth. (1) Oslofjord based on coral Li/Mg (Raddatz et al., 2016) and modern long-term oceanographic observation at Tisler Reef (Guhen et al., 2012), (2) Lopphavet, based on coral Li/Mg (Raddatz et al., 2016), (3) Cape Lookout, based on foraminiferal Mg/Ca (Matos et al., 2015), (4) Bay of Biscay, based on coral Li/Mg (Montero-Serrano et al., 2015, 2012), (5) Lopphavet, based on coral Li/Mg (Raddatz et al., 2013), and (5) long-term (3 Myr, low resolution) Challenger Mound, based on coral Li/Mg (Raddatz et al., 2014b).

salinity changes (Lea et al., 2000). The temperature effect can be removed by using the Mg/Ca-based temperatures of the initial foraminiferal $\delta^{18}$O and a temperature versus $\delta^{18}$O$_{\text{calcite}}$ equation (Shackleton, 1974) to resolve it towards $\delta^{18}$O$_{\text{seawater}}$. Then, the regional ice volume free $\delta^{18}$O$_{\text{sw}}$ record has to be computed ($\delta^{18}$O$_{\text{ivf-sw}}$) by accounting for changes in global $\delta^{18}$O$_{\text{sw}}$ due to continental ice volume variability using a relative sea-level curve (Austermann et al., 2013). The error in $\delta^{18}$O$_{\text{ivf-sw}}$ is assumed to be close to $\pm 0.4\%$, based on the approach in error propagation analysis by Bahr et al. (2013). However, as the relationship between $\delta^{18}$O$_{\text{sw}}$ and salinity possibly varied through time, a conversion of $\delta^{18}$O$_{\text{ivf-sw}}$ values into absolute salinities may cause unrealistic values and variations. Therefore, there is an urgent need to develop better and more robust salinity proxies.

2.2.2 Na/Ca

Recently, it was shown that the amount of Na incorporated into the tests of benthic foraminifera is related to seawater salinity (Wit et al., 2013). By the 1960s and 1970s it was known that the incorporation of Na depends on seawater salinity for the Atlantic oyster *Crassostrea virginica* (Rucker and Valentine, 1961), barnacle shells (Gordon et al., 1970) and inorganic calcite (Kitano et al., 1975). The more recent study by Wit et al. (2013) reproduced these findings in the benthic foraminifera *Ammonia tepida* and found strong relationships between Na/Ca ratios and seawater salinity which can be described by the following equations: Na/Ca = 0.22 × salinity−0.75, with $r^2$ of 0.96. Subsequent studies again reproduced these findings and found similar relationships in various planktonic foraminifera (Allen et al., 2016; Mezger et al., 2016). However, *A. tepida* lives at shallow water depths and has rarely been used in palaeoceanographic reconstructions. Therefore, future studies should focus on calibrations using more common species like *Cibicidoides* sp. or *Uvigerina* sp. To date there are only a few studies that systematically evaluate the impact of seawater salinity or temperature on Na incorporation into cold-water Scleractinia. For example, the study by Rollion-Bard and Blamart (2015) suggests that changes in growth rate due to varying seawater temperature might control the incorporation of Na (Li and Mg) and the studies of Rollion-Bard et al. (2017) and Schleinkofer et al. (2019) imply that a temperature control on Na/Ca ratios might also exist in aragonitic marine species.

2.3 Seawater density

2.3.1 Stable oxygen isotopes

The reconstruction of seawater densities (here: sigma theta or $\sigma_\Theta$) has been developed by Lynch-Stieglitz et al. (1999a, b) on the basis of stable oxygen isotopes ($\delta^{18}$O$_c$) from the calcite tests of epibenthic foraminifera. Salinity (evaporation and precipitation) and temperature (ice volume) control both, seawater density and the $\delta^{18}$O$_c$, and its dependency is assumed to be constant throughout the oceans and geological time (Lynch-Stieglitz et al., 1999a). Different seawater density–$\delta^{18}$O$_c$ relationships have been proposed for different oceanographic settings (salinity, $\delta^{18}$O$_{\text{sw}}$) and temperature regimes (Lynch-Stieglitz et al., 1999b). These show good results for reconstructed values and related geostrophic transport estimates (Lynch-Stieglitz et al., 1999a).

The density of seawater has been identified as one important parameter affecting CWC growth and mound development in the NE Atlantic with recent thriving CWC reefs at densities of $27.5 \pm 0.15$ kg/m$^3$ (Dullo et al., 2008; De Mol et al., 2011; Rüggeberg et al., 2011; Flögel et al., 2014). Other studies reported on CWC reefs outside this range, for example, in the Mediterranean Sea with $\sigma_\Theta$ values of 29.1 kg/m$^3$ (Dullo et al., 2009) or the Gulf of Mexico with values of 27.1–27.3 kg/m$^3$ (Davies et al., 2010; Hebbeln et al., 2014). This indicates that CWC reefs and mounds grow preferentially at pycnoclines where bottom or intermediate nepheloid layers develop carrying nutrients to the reefs, but resulting density values may vary depending on the ocean basin as different intermediate water masses are involved (e.g. Norway: Norwegian Coastal Water/Atlantic Water: Dullo et al., 2008; NE Atlantic: Eastern North Atlantic Water/Mediterranean Outflow Water: Dullo et al., 2008; De Mol et al., 2011;
Flögel et al., 2014; Somoza et al., 2014; Sánchez et al., 2014; low latitude Atlantic [North Atlantic Central Water/Antarctic Intermediate Water, Vanderpoe et al., 2016]). However, most of the NE Atlantic reefs and mounds are in the documented density range of 27.35–27.65 kg/m³. Applicability to the palaeo-record was tested on sediment records of CWC mounds for the past 300 kyr (Rüggeberg et al., 2016) using a regional salinity-δ¹⁸Osw relationship to setup a seawater density–δ¹⁸O relationship following the method of Lynch-Stieglitz et al. (1999b). These reconstructions show comparable ranges for both recent environments and the Holocene period and at times of CWC growth during the past glacial and interglacial cycles (Rüggeberg et al., 2016). Here, additional seawater density data were compiled using the regional calibration for the Porcupine Seabight (Rüggeberg et al., 2016) and benthic δ¹⁸O data published from different carbonate mounds (Rüggeberg et al., 2016). Here, additional seawater density data were compiled using the regional calibration for the Porcupine Seabight (Rüggeberg et al., 2016) and benthic δ¹⁸O data published from different carbonate mounds (Rüggeberg et al., 2016) and benthic δ¹⁸O data published from different carbonate mounds (Dorschel et al., 2005; Eisele et al., 2008; Raddatz et al., 2011; Bergmans, 2013; see Figure 4). The data indicate a distribution within the Atlantic range of CWC occurrences (27.2–27.7 kg/m³) and mainly within the narrower range of the NE Atlantic (27.35–27.65 kg/m³) with a mean value of 27.41 kg/m³ for the past 3 Myr.

To date not many studies have tried to reconstruct past seawater densities. A combined study based on oxygen isotopes and an independent temperature proxy (e.g. Mg/Ca, Li/Mg, etc.) as well as an additional salinity proxy (e.g. Na/Ca) may help to improve both the method and the accuracy.

2.4 Carbonate system (seawater pH, CO₃²⁻ concentration and saturation)

2.4.1 Sr/Ca

Besides the above mentioned temperature effect on Sr/Ca ratios in aragonitic CWC and tropical corals, the incorporation of Sr into calcite is influenced by many other factors such as growth and calcification rates in coccolithophorids (Stoll and Schrag, 2000) and foraminifera (Lea et al., 1999). Strontium incorporation into foraminiferal calcite appears to vary also with temperature, salinity and seawater pH (Kisakürek et al., 2008) as well as water depth, which could hint to a possible effect of the carbonate ion concentration on the Sr/Ca ratio of calcitic foraminifera (Russel et al., 2004; Dissard et al., 2010; Raitzsch et al., 2010). Recent cultivation studies and palaeoceanographic reconstructions, as well as core top calibrations, underline the strong impact of the seawater carbonate system parameters on Sr/Ca ratios in foraminifera (Yu et al., 2014; Keul et al., 2017). Which of the carbonate system parameters have the strongest effect on foraminiferal Sr/Ca is still a matter of debate, but cultivation studies on the benthic foraminifera Ammonia sp. suggest that CO₃²⁻, pCO₂ as well as pH do not primarily influence Sr incorporation, but rather the DIC/bicarbonate ion concentration (Keul et al., 2017). This observation makes the Sr/Ca ratio in (benthic) foraminiferal calcite a powerful tool to constrain the carbonate system, especially by combining the Sr/Ca ratio with other carbonate system proxies such as U/Ca, B/Ca and δ¹¹B. Nevertheless, the benthic foraminifera used in this study is a shallow water species and hardly used in any palaeoceanographic reconstruction. Therefore, future studies should aim at investigating the Sr incorporation in frequently used benthic foraminifera testing the possible application of this proxy to CWC mound settings.

2.4.2 U/Ca

Uranium is a conservative element in seawater under oxygenated conditions and exhibits a residence time of about 300–600 kyr, which is considerably longer than the ocean mixing time of 1500 years (Ku et al., 1977; Barnes and Cochran, 1990). Moreover, U exists in seawater in the form of multiple uranyl (UO₂(²⁺)) carbonate complexes, which change their abundances with varying pH and CO₃²⁻ (Djogić et al., 1986; Krestou et al., 2003). Marine carbonates appear to preferentially incorporate UO₂(CO₃)²⁻ and UO₂CO₃ resulting in increasing U/Ca ratios with decreasing seawater pH or carbonate ion concentrations (Russel et al., 1994; Reeder et al., 2000; Russel et al., 2004). Accordingly it has been shown that U/Ca ratios in planktonic (Russel et al., 2004; Allen et al., 2016) and benthic foraminifera (Raitzsch et al., 2011) are primarily controlled by the seawater carbonate ion concentration. Besides seawater temperature (Min et al., 1995), the U/Ca ratios in tropical corals appear to be also controlled by seawater pH (Inoue et al., 2011), whereas in scleractinian CWCs both the carbonate ion concentration and pH have been shown to be controlling parameters, depending on the species. In particular, the solitary slow growing CWC D. dianthus appears to largely incorporate U due to varying CO₃²⁻ concentrations, whereas the fast growing CWC D. pertusum incorporates U depending on the seawater pH (Anagnostou et al., 2011; Raddatz et al., 2014a). This observation is systematically consistent as seawater pH generally decreases with decreasing CO₃²⁻ concentrations. However, the obviously different growth behaviour of these two different CWC species may imply a growth effect on U incorporation, with lower U/Ca ratios a result of faster growth.

Another concern with CWC U/Ca ratios is the large internal variation with low values in the COC and higher values in the theca wall (Robinson et al., 2006; Sinclair et al., 2006; Raddatz et al., 2014a). The latter study suggested that this variability can be explained by the pH up-regulation of the CWC against seawater saturation (McCulloch et al., 2012), which would, in turn, be related to changes in growth rate. Based on this, the U/Ca ratio has been applied to reconstruct
seawater pH in Holocene CWC *D. pertusum* samples from northern Norway (71°N, Lopphavet; Raddatz et al., 2016). The reconstruction revealed a major change in seawater pH to values as low as 7.6 or even (unrealistically) 7.0. This severe shift is probably associated with advances of CO₂ enriched Arctic waters due to a southward shift of the polar front during the mid-Holocene (Raddatz et al., 2016). However, culturing studies are needed to fully understand U incorporation into CWC skeletons. These should be carried out using a similar approach as Keul et al. (2017, see Section 3.2) to identify (a) which main carbonate system parameter controls U incorporation into the CWC skeleton, and (b) if the internal pH up-regulation changes with a varying carbonate system and if this impacts the internal variability of U/Ca ratios.

### 2.4.3 Boron isotopes

Boron isotopes have been used in many studies to reconstruct changing seawater pH in the marine carbonate record (Hönisch et al., 2012) and, based on this, to constrain past atmospheric CO₂ concentrations (Foster et al., 2012).

Boron exists in seawater as boric acid in trigonal (threefold) coordination \([\text{B(OH)}_3^+]\) and as borate in tetrahedral (fourfold) coordination \([\text{B(OH)}_4^-]\). The speciation of these in seawater is pH dependent, in which the borate ion increases with increasing pH and, logically, the boric acid increases with decreasing pH. The borate ion has a preference for the lighter \(^{10}\text{B}\) isotope and the boric acid for the heavier isotope \(^{11}\text{B}\). This pronounced fractionation factor can reflect the boron species present in seawater (Vengosh et al., 1991; Zeebe, 2005; Klochko et al., 2006). One fundamental assumption made in the study of boron isotope systematics in marine carbonates is that only the borate is incorporated into the carbonate shells. This was initially proposed by Hemming and Hanson (1992), who observed a large offset between the seawater boron isotopic composition (39.6‰, Foster et al., 2010) and marine carbonates, with the latter much closer to the borate ion isotopic composition. Recent studies utilizing nuclear magnetic resonance and electron-loss spectroscopy have cast doubt on this assumption (Klochko et al., 2009; Rollion-Bard et al., 2011). However, a possible explanation was added to the ongoing discussion with crystallographic control modifying the boron isotopic composition in calcite and aragonite instead of the assimilation of boric acid (Rasbury and Hemming, 2017).

Irrespective of this, many studies have revealed a strong pH control on the boron isotopic composition of inorganic synthesized carbonates (Uchikawa et al., 2015; Kaczmarek et al., 2016). Contrasting results have been published for biologically mediated marine carbonates, in which three different groups of marine carbonates have been identified (for a detailed discussion see Foster and Rae, 2016) as follows.

1. No offset to the theoretical \(\delta^{11}\text{B} \ [\text{B(OH)}_4^-]\) (e.g. coralline red algae, benthic foraminifera) (Rae et al., 2011; Fietzke et al., 2015).
2. Above the theoretical \(\delta^{11}\text{B} \ [\text{B(OH)}_4^-]\) (e.g. symbiont bearing planktonic foraminifera, cold and warm-water Scleractinia) (Hönisch et al., 2003; McCulloch et al., 2012; Henehan et al., 2013).
3. Below the theoretical \(\delta^{11}\text{B} \ [\text{B(OH)}_4^-]\) (non-symbiont bearing planktonic foraminifera, aragonitic benthic foraminifera; Rae et al., 2011; Yu et al., 2014; Martinez-Boti et al., 2015).

While the apparent fit to the theoretical \(\delta^{11}\text{B} \ [\text{B(OH)}_4^-]\) values in various calcitic benthic foraminifera makes this archive very robust to reconstruct the seawater carbonate system through time, the remaining offsets to the theoretical \(\delta^{11}\text{B}\) values...
have been explained by vital effects. The $\delta^{11}$B values of various scleractinian CWCs are significantly above the inorganic seawater borate equilibrium curve (Anagnostou et al., 2012; McCulloch et al., 2012). In line with biominalization models (Al-Horani et al., 2003; Marubini et al., 2008), this may indicate a strong pH up-regulation of the internal calcifying fluid ($pH_{cf}$) against ambient seawater (Anagnostou et al., 2012; McCulloch et al., 2012; Jurikova et al., 2019). With 0.6–0.8 pH units above seawater this offset of internal coral pH compared to seawater pH (ΔpH) appears to be even higher than in tropical corals (0.2–0.4, Hönsich et al., 2003) and even increases with decreasing seawater pH (McCulloch et al., 2012). Such a process of internal pH$_{cf}$ up-regulation can only be sustained by an energetic cost of about 10% per 0.1 decrease in seawater pH (McCulloch et al., 2012), which in turn can only be achieved with sufficient food supply. This appears to be consistent with a recently published cultivation study on D. pertusum (Büscher et al., 2017). Here, the authors showed that coral calcification has the largest benefit by food availability even with a seawater pH as low as 7.7 (Büscher et al., 2017). Moreover, boron isotopes in scleractinian CWCs are also highly systematic as they strongly correlate to seawater aragonite saturation (McCulloch et al., 2012), holding promise that the $\delta^{11}$B ratio in CWCs can be used to either reconstruct seawater pH or seawater aragonite saturation as well as the internal coral pH (= coral viability). Combining independent seawater pH proxies such as coral U/Ca ratios and $\delta^{11}$B values in benthic foraminifera and corals may be a powerful tool to reconstruct not only seawater pH, but also the physiological responses of CWCs to a changing environment.

One study that followed this approach applying $\delta^{11}$B values and U/Ca ratios in CWCs focused on the Mid-Holocene CWC reef decline off Norway (Raddatz et al., 2016). Here, the authors found a severe seawater pH decline accompanied by a drop in temperature (based on Li/Mg ratios) which they explained as the result of the southward migration of the Polar Front associated with advances of Arctic water masses enriched in CO$_2$ (low seawater pH). The resulting ΔpH ($pH_{cf}$—seawater pH) record tends to show an increase during times of seawater pH and temperature decrease, which possibly caused the Holocene CWC reef decline off Norway. Probably sufficient food supply at these high-latitude Norwegian reefs helped the coral to balance their additional energetic costs. However, these results are intriguing as it was shown that scleractinian corals do ‘feel the acid’ recorded in lower $\delta^{11}$B values (Venn et al., 2013), recently also shown for CWCs (D. dianthus) by Rae et al. (2018). However, the study by Raddatz et al. (2016) corroborates the findings of McCulloch et al. (2012), in that CWCs are to some extent resilient to ocean acidification by up-regulating their internal pH. This is especially true if sufficient food and nutrients are available. In line with these observations, the recent cultivation experiment by Büscher et al. (2017) showed that increased food supply results in higher growth even with decreased seawater pH down to 7.7, but not for higher T and lower seawater pH (Büscher et al., 2017). However, to fully understand boron isotope systematics, we need species-specific calibrations in various CWCs as well as multivariate cultivation experiments to disentangle the above-mentioned effects.

### 2.5 Nutrients

#### 2.5.1 Ba/Ca, P/Ca and Cd/Ca

As discussed above, food supply and nutrient availability is probably the most crucial factor controlling CWC reef and mound growth. Major nutrients (macronutrients) such as C, nitrate, Si and phosphate (PO$_4^{3-}$) are taken up by marine phytoplankton and exported down to the seafloor. In particular, over 86% of the macronutrients are remineralized in the first 1000 m of water depth resulting in the characteristic distribution pattern in the water column (Martin et al., 1987; Hendry et al., 2008). Besides Fe other micronutrients have been the focus of research on biogeochemical cycling in seawater. Cadmium for example may also be biologically important since it catalyzes the uptake of bicarbonate HCO$_3$− (Morel and Price, 2003) and correlates with primary productivity in the Atlantic Ocean (Pinedo-Gonzales et al., 2015).

Micronutrients such as Cd (Bryan and Marchitto, 2010) and Ba (Chow and Goldberg, 1960) have a similar vertical distribution pattern as the major nutrients like Si. In contrast to other elements, where biological activity extracts these elements from the surface (Bernstein et al., 1998), the draw-down of Ba in surface waters has also been attributed to the precipitation of barite (BaSO$_4$) (Monnin et al., 1999).

Nevertheless, due to its nutrient-type distribution, dissolved Ba also co-varies with alkalinity (Chan et al., 1977). This is the foundation of several studies attempting to reconstruct past ocean alkalinity and circulation utilising benthic foraminifera (Lea and Boyle, 1990; Lea, 1993; Hall and Chan, 2004). The fundamental assumption with this is that Ba incorporation into the foraminiferal calcite depends solely on the Ba/Ca ratio of seawater. This could be shown for foraminiferal calcite (Hönisch et al., 2011) as well as for scleractinian CWC (Anagnostou et al., 2011; Hemsing et al., 2018; Spooner et al., 2018). However, Ba/Ca ratios in foraminifera sometimes reveal a strong sample heterogeneity (Hathorne et al., 2009) and exhibit different sensitivities in scleractinian CWCs, which may indicate that other parameters also exert a control on Ba incorporation (Anagnostou et al., 2011; Hemsing et al., 2018) or it may be a location effect (Spooner et al., 2018).

For comparison the apparent different relationships between Ba/Ca$_{carbonate}$ against Ba/Ca$_{seawater}$ are listed below and shown in Figure 5.
These apparently different sensitivities observed for the Ba/Ca ratio may not only reveal a biological effect, but also different incorporation mechanisms of Ba into the two different mineral phases. The sensitivity in aragonite appears to be slightly stronger when compared to calcite. However, it shows that the reconstruction of absolute Ba/Ca seawater values requires at least a mineral phase specific calibration. To further understand these different distribution coefficients $D$ in aragonitic and calcitic marine carbonates, an interesting study might be to test Ba/Ca systematics in aragonitic benthic foraminifera such as *H. elegans*.

Reconstructions on CWC mounds using coral Ba/Ca ratios have so far been used to either reconstruct meltwater/

![FIGURE 5](image-url) **Linear regression lines of Ba/Ca ratios in scleractinian cold-water corals (equations 1 and 2) and planktonic and benthonic foraminiferal (equations 3 and 4) shell against Ba/Ca of seawater.** Calibrations are taken from Hemsing *et al.* (2018) for equation 1 and Anagnostou *et al.* (2011) for equation 2. Calibrations for the foraminifera are taken from De Nooijer *et al.* (2017) for 3 and Honisch *et al.* (2011) for 4. For the detailed equation and further discussion please see text. Note to avoid confusion instead of 95% confidence interval, a constant 5% error for each of the calibration slopes are plotted (shaded areas). Note: Only calibration 1 is not forced to zero.

1. \[\text{Ba/Ca}_{\text{multispecies}} = 0.149 \pm 0.050 \times \text{Ba/Ca}_{\text{sw}}\] \[\text{Hönisch et al. (2011)}\]
2. \[\text{Ba/Ca}_{\text{Scleract. CWC (D. diannah)} = 1.400 \pm 0.300} \times \text{Ba/Ca}_{\text{sw}} + 0 \pm 2\] \[\text{Anagnostou et al. (2011)}\]
3. \[\text{Ba/Ca}_{\text{Bent. foram.}} = 0.777 \pm 0.007 \times \text{Ba/Ca}_{\text{sw}}\] \[\text{De Nooijer et al. (2017)}\]
4. \[\text{Ba/Ca}_{\text{Scleract. CWC (multispecies) = 1.800 \pm 0.400} \times \text{Ba/Ca}_{\text{sw}} + 0.7 \pm 2.6}\] \[\text{Hemsing et al. (2018)}\]

Cadmium in its dissolved form is distributed in the world’s oceans in a similar manner to the major nutrient phosphate (Boyle, 1992). Both are removed from the sea-surface by biological activity and regenerated at depth, characterized by concentration maxima in intermediate water masses (1 km, Boyle, 1992). Cadmium in foraminiferal calcite has been used to reconstruct deep (Marchitto and Broecker, 2006), upper (Came *et al.*, 2008; Poggemann *et al.*, 2017) as well as surface nutrients (Rickaby and Elderfield, 1999). Benthic foraminiferal Cd/Ca ratios reflect the Cd increase with water depth (Boyle, 1988, 1992; Boyle and Rosenthal, 1996). At depths >1150 m the distribution coefficient for Cd ($D_{Cd}$) may change, however, for our purpose these depth ranges appear to be irrelevant for probably all of the Quaternary CWC mounds shallower than 1150 m water depth. On a global scale the correlation of Cd and phosphate and their distribution can be attributed to the preferential uptake of Cd by particulate organic matter (Elderfield and Rickaby, 2000). However, Cd/Ca ratios in scleractinian CWC have been little studied (Adkins *et al.*, 1998; Mangini *et al.*, 2010). In particular, Adkins *et al.* (1998) showed that the Cd/Ca ratios of *D. dianthus* may reflect those of seawater, however, there is still no proper calibration determining the exact $D_{Cd}$ for scleractinian CWC. Mangini *et al.* (2010) showed that coral Cd/Ca in CWC mounds off Brazil tend to increase during the Heinrich 1 (H1) event indicating a higher nutrient concentration in Southern Component Waters. This is supported by the recent study of Poggeman *et al.* (2017) investigating Cd/
Ca ratios of benthic foraminifera at intermediate water mass depths of northern South America. Their coupled δ^{13}C and Cd/Ca approach could reveal abrupt deglacial nutrient enrichments in Antarctic Intermediate Water (AAIW) during the YD and H1 (Figure 6). Interestingly, this is partly accompanied by the CWC occurrences off Brazil at similar intermediate water depths (Mangini et al., 2010). The CWC mound growth generally benefits from sea-surface productivity resulting in enhanced organic matter supply. This was proven for different time intervals and for various locations initializing the theory of biogeographical movements of CWC ecosystems from glacial to interglacial periods (Wienberg et al., 2010; Eisele et al., 2011; Frank et al., 2011; Raddatz et al., 2014b). Periods of rapid climate cooling in the Northern Hemisphere were characterized by a shift of the Intertropical Convergence Zone (ITCZ), and the associated west winds in the South Atlantic which in turn may have led to a displacement of high sea-productivity cells off South America (Anderson et al., 2009; Hoffmann et al., 2014). It may therefore, suggest again that organic matter supply is a primary trigger for CWC growth. However, it needs to be proven if CWC mounds off South America are really restricted to those periods of rapid climate cooling such as YD and H1 events and if enhanced CWC growth off Brazil is facilitated by high sea-surface productivity. Overall, Ba/Ca, P/Ca as well as Cd/Ca ratios are promising tools to understand past nutrient cycling and to constraint nutrient boundary conditions for CWC mound growth. However, we still lack basic species-specific calibrations (for Ba possibly not needed) in order to quantitatively use these proxies accordingly. As a consequence, the use of both archives, foraminifera and corals, is recommended to fully understand changes in nutrient and element (Ba, P and Cd) cycling.

2.6 | Oxygen

2.6.1 | Mn/Ca

Manganese is an essential micronutrient and is strongly redox sensitive in seawater. In particular Mn is removed by oxidation from soluble Mn(II) to insoluble Mn(III, IV) oxides. Manganese has therefore the potential to act as a proxy for changes in oxygen concentrations. In benthic foraminifera, Mn/Ca ratios are routinely measured along with Mg/Ca to monitor the presence of diagenetic Mn oxyhydrates and Mn carbonates (Barker et al., 2003), but have also been identified as a redox proxy, in which higher Mn/Ca ratios correspond to low dissolved oxygen concentrations (Glock et al., 2012; Groeneveld and Filipsson, 2013). An important feature appears to be the microhabitat of the benthic foraminifera. Endobenthic foraminifera tend to be a better recorder of the oxygenation state than epibenthic foraminifera (Koho et al., 2015). The study by McKay et al. (2015) revealed that a combined approach of foraminiferal Mn/Ca and sedimentary Mn/Al ratios might be the approach to tackle changes in the redox system. Several working groups are studying past oxygenation dynamics with the use of Mn/Ca ratios in CWC mound settings; however, there is still no study published applying this proxy. It is suggested that future studies should focus on calibrating this proxy in CWC settings using benthic foraminifera from different microhabitats and compare this to sedimentary, for example, Mn/Al ratios.

2.6.2 | Other oxygenation proxies

In the last decades several potential tools have been developed and evaluated in order to reconstruct past oceanic oxygen dynamics. Two very promising tools are uranium isotopes (235U/238U) (Lau et al., 2017; Clarksen et al., 2018) as well as Molybdenum-isotopes (δ^{98}Mo, Siebert et al., 2003; Scholz et al., 2017). These palaeo redox proxies may give new insights into the timing, duration and extent of marine anoxia associated with biological radiation and extinctions (Siebert et al., 2003; Voegelin et al., 2009; Lau et al., 2017). In the case of U it is commonly assumed that U incorporation into marine carbonates directly records the 238U/234U of seawater. However, recent findings by Chen et al. (2016 and 2017) revealed that U isotope fractionations may depend also on the aqueous speciation of U, which is controlled by pH, pCO2, ionic strength as well as Ca^{2+} and Mg^{2+} concentrations, having considerably changed throughout Earth history (Fante and DePaolo, 2006). Similar to U, Mo isotopes are also not a straightforward proxy. For example, the study by Scholz et al. (2017) carried out in the oxygen minimum zone off Peru showed that Fe and nitrate dependent Mo shuttling under nitrogenous conditions needs to be evaluated with respect to Mo enrichments during times of oceanic anoxia. Considering that mounds are usually growing near the pycnocline a thoughtful investigation of this isotope system is required. All these findings suggest that a multi-proxy approach appears to be the most promising way to reconstruct oceanic oxygen near CWC mounds.

2.7 | Water Masses

2.7.1 | Neodymium isotopes

Neodymium belongs to the group of rare earth elements (REE, Lanthanide series). As with all other REE (except for Ce, which can be oxidized to the +4 state), under oxygenated conditions Nd exists in seawater exclusively in the +3 state. The Nd isotopic composition of seawater depends on the source area, where water masses are imprinted with the Nd isotopic signature of the rocks of the adjacent landmass through weathering processes (Frank, 2002). In
FIGURE 6  Compilation of $^{230}$Th/U ages from the Brazilian margin (Mangini et al., 2010; Henry et al., 2014). Additionally, plotted are the Cd/Ca-based nutrient reconstructions on benthic foraminifera (green) from Poggemann et al. (2017). For comparison to the Northern Hemisphere, the abundance of the cold-water planktonic foraminifera *Neogloboquadrina pachyderma* sinistral (NPS) of Barker et al. (2015) is indicated (note flipped axis).

TABLE 2  Cross table highlighting geochemical proxies that can be extracted from calcareous organisms such as scleractinian cold-water corals (CWC) and benthic foraminifera (F) from cold-water coral mounds

| Environmental parameter | Temperature | Salinity | Seawater-density | Carbonate system | Nutrients | Dissolved Oxygen | Water Masses |
|-------------------------|-------------|---------|------------------|------------------|----------|------------------|--------------|
| Archive                 | CWC         | F       | CWC              | F                | CWC      | F                | CWC          | F            | CWC          | F            |
| Stable oxygen isotopes  | (x)         | x       | x                | x                | x        |                  |              |              |              |
| (δ¹⁸O)                  |             |         |                  |                  |          |                  |              |              |              |
| Clumped isotopes        |             | x       |                  |                  |          |                  |              |              |              |
| (Δ₄⁷)                   |             |         |                  |                  |          |                  |              |              |              |
| Sr/Ca ratios            | (x)         |         |                  |                  |          |                  | x            |              |
| Mg/Ca ratios            |             | x       |                  |                  |          |                  |              |              |              |
| Li/Mg ratios            |             | (x)     |                  |                  |          |                  |              |              |              |
| Na/Ca                   | (x)         |         |                  |                  |          |                  | x            |              |              |
| U/Ca                    |             |         |                  |                  |          |                  |              |              |              |
| Boron isotopes          |             |         |                  |                  |          |                  | x            | x            |              |
| (δ¹¹B) and B/Ca         |             |         |                  |                  |          |                  |              |              |              |
| Ba/Ca, P/Ca, Cd/ Ca     |             |         |                  |                  |          |                  | (x)          | (x)          |              |
| Mn/Ca                   |             |         |                  |                  |          |                  |              |              |              |
| Uranium isotopes        |             |         |                  |                  | (x)      | (x)              |              |              |              |
| Molybdenum isotopes     |             |         |                  |                  | (x)      | (x)              |              |              |              |
| Neodymium isotopes      |             |         |                  |                  |          |                  | x            |              |              |

*Note: x = possible, (x) = more studies dedicated to CWCs and their mounds are needed, ? = not systematically studied by now.*

*aIn combination with Mg/Ca or clumped isotopes.*

*bIn combination with δ¹⁸O.
particular, old continental rocks contribute to much lower Nd values than younger mantle-derived material (Goldstein and O’Nions, 1981). Due to the fact that Nd has an average oceanic residence time of >400 years (Rempp et al., 2011), Nd isotopes from various archives have been used as a quasi-conservative tracer to reconstruct changes of past ocean circulation (Frank, 2002; Roberts et al., 2010; Crocket et al., 2011). One issue when using Nd isotopes are processes involving boundary exchange between sediments and bottom water. Studies of the modern ocean revealed that boundary exchange leads to the addition of Nd from oceanic margin sediments, which in turn may be an important part of the oceanic Nd cycle and thus complicates the use of Nd isotope as a water mass tracer (Osbourne et al., 2014; Haley et al., 2017). However, CWCs thrive significantly above the sediment/seawater interface and may therefore be a promising archive for Nd isotopes. Accordingly, CWCs may record (if sufficiently cleaned) the Nd isotopic composition of seawater in a 1:1 relationship (Copard et al., 2010; Van de Flierdt et al., 2010; Struve et al., 2017) and have successfully been applied in water mass reconstructions (Colin et al., 2010; Montero-Serrano et al., 2011; Copard et al., 2012; Struve et al., 2017) and to track meltwater pulses in Norwegian fjords (López Correa et al., 2012). Water mass reconstructions in CWC mounds based on Nd isotopes exhibit severe changes of up to 3 units in εNd for periods with relatively stable climates or even on high-resolution scale recording the NAO (Copard et al., 2012; Montero-Serrano et al., 2013). In particular, the study by Copard et al. (2012) shows that CWC skeletons record within about 20 years a shift from −14.4 to −11.8 in εNd, which suggests strong variability of the subtropical gyre. These findings convincingly show that CWCs do indeed record intermediate water mass dynamics in their Nd isotopic compositions, but it also suggests that CWC mound growth is not bound to specific intermediate water masses.

3 | FUTURE DIRECTIONS

The unique opportunities to reconstruct various environmental parameters in CWC mounds have been highlighted here by the use of foraminifera and scleractinian CWCs. With our current knowledge it is possible to constrain almost all oceanographic parameters facilitating and restricting growth (Table 2).

Based on the studies that are available so far it becomes clear that the best scenarios follow a multi-proxy approach which can then be compared to aggradation rates as suggested by Wienberg and Titschack (2017) in order to define CWC mound boundaries for optimal growth.

One possibility to study past seasonality in CWCs is the use of deep-sea marine bivalves such as Acesta excavata. A multi archive approach will reveal the most realistic situation, hopefully capturing most of the possible environmental variations while also addressing times of no coral growth.

Since age control is a re-occurring issue when using multiple archives, future research should attempt to further enhance the age control of calcareous organisms in CWC mounds.

Another still prominent issue when calibrating and applying geochemical proxies is the effect of biomineralization on element incorporation and isotope fractionation, especially if these processes have changed through time. Therefore, one aim should be to identify proxies that are either unbiased by biomineralization kinetics and/or that are able to characterize such processes (e.g. Δ48, Fiebig et al., 2019).

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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