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Modeling the sensitivity of the marine $CaCO_3$ cycle to the formulation of the degree of undersaturation of seawater with respect to $CaCO_3$

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The uptake of anthropogenic CO_2 by the ocean leads to an increase in dissolved inorganic carbon and a redistribution of the marine carbonate system, i.e. decreasing pH and carbonate ion concentrations ($[CO_3^{2-}]$) in accordance with changing the calcium carbonate ($CaCO_3$) saturation state. This process is called ocean acidification, and its impact on marine biogeochemical cycles as well as potential climate feedbacks is presently under scientific debate. Generally, there are two definitions for the description of the saturation state of seawater with respect to $CaCO_3$:

(1) $\Delta[CO_3^{2-}]$, which reflects the difference between the in-situ carbonate ion concentration $[CO_3^{2-}]$ and the saturation concentration $[CO_3^{2-}]_{sat}$, and

(2) Ω, which can be approximated by the ratio of in-situ $[CO_3^{2-}]$ over $[CO_3^{2-}]_{sat}$,

where $[CO_3^{2-}]_{sat}$, the saturation concentration, is a function of temperature, salinity and pressure.

In contrast to $\Delta[CO_3^{2-}]$, Ω exhibits a vertical gradient in the sensitivity to changes in the $[CO_3^{2-}]$ concentration. This becomes clear when considering that a unity change in $[CO_3^{2-}]$ always results in the same amount of change in $\Delta[CO_3^{2-}]$ ($d\Delta[CO_3^{2-}]$ / $d[CO_3^{2-}]$ = 1), whereas for Ω the change depends on the background $[CO_3^{2-}]_{sat}$ level ($d\Omega$ / $d[CO_3^{2-}]$ = 1 / $[CO_3^{2-}]_{sat}$). As a result of the increasing solubility of $CaCO_3$ ($[CO_3^{2-}]_{sat}$) with pressure the sensitivity of Ω to $[CO_3^{2-}]$ decreases over depth in the water column.

When calculating a $CaCO_3$ dissolution rate (R) via application of a dissolution rate constant k, which transforms the degree of undersaturation of seawater with respect to $CaCO_3$ (S) into a time-dependent rate following the dissolution kinetics $R = k * S^n$ as expressed by Keir (1980), this has the counterintuitive effect that although solubility ($[CO_3^{2-}]_{sat}$) is vertically increasing the dissolution reaction in response to a unity $[CO_3^{2-}]$ change would be slower at greater depth. For a reaction rate order of $n = 1$ dissolution at depth is delayed by a factor of three (Archer et al., 1989), however, for $n = 4.5$ as reported by Keir (1980), this factor may rise up to 100.

Consequently, the two commonly used formulations to express the degree of undersaturation of seawater with respect to $CaCO_3$ are not equally applicable for the calculation of $CaCO_3$ dissolution in the ocean across pressure gradients. However, to assess the vulnerability of marine organisms, sediment buffering, and the time scales of climate carbon cycle feedbacks the appropriate formulation needs to be known. In the present study we apply a marine biogeochemical model to test the sensitivity of the marine carbon cycle, in particular the vertical flux of particulate $CaCO_3$, to the formulation of the saturation state of seawater with respect to $CaCO_3$.