Solubility controls carbonate chemistry with local modifications in North American ocean margins

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Increase of anthropogenic CO_2 in the atmosphere has acidified the chemical environments of the ocean and thus affected the health of organisms and ecosystems therein on a global scale. While recent studies have demonstrated the severity and complexity of ocean acidification (OA) in many dynamic coastal sites, control mechanisms on large spatial distribution patterns are still lacking. Based on recent measurements of multiple carbonate parameters in North American ocean margins, we show that total dissolved inorganic carbon (DIC) distributions and carbonate saturation state (Ω) in the East Coast and Gulf of Mexico waters demonstrate a remarkable consistency with predictions from a solubility control mechanism which dictates that as more CO_2 uptake occurs the carbonate (CO_3^{2-}) concentration and Ω show a long-term decrease, despite large local variabilities in sea surface distributions of carbon dioxide partial pressure (pCO_2) and pH. The solubility driven mechanism still exerts a dominant role in the offshore waters but is greatly modified in the near coastal surface waters by strong upwelling and subsequent biological production of the West Coast with the former leading to low Ω and pH and high pCO_2 and the latter to opposite distributions. We further show that the contrasting features between the East-Gulf and West coasts and between Ω and pH are determined by the contrasting time scales of process and the nature of acid-base equilibrium of the systems. Our findings emphasize the needs for understanding contrasting organismal responses to Ω and pH and have ramifications for predicting future OA changes in ocean margins.