## **Ion content and carbon speciation set ocean carbon and alkalinity inventories for balanced budgets**

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The inventories of alkalinity and carbon in Earth's global surface environment are dynamically balanced by two restorative climate-carbon cycle feedbacks:  $CaCO<sub>3</sub>$  compensation with a relaxation timescale of  $10^3 - 10^4$  years, and silicate weathering with a timescale of  $10^5 - 10^6$  years. These feedbacks operate to regulate climate through CO<sub>2</sub> greenhouse gas forcing and deep sea  $CaCO<sub>3</sub>$  burial through the bulk ocean calcite saturation state, whereby feedback strengths are moderated by the chemical speciation of carbon and other elements dissolved in seawater. Pitzer-type seawater speciation models suggest that secular fluctuations in bulk ocean ion content (e.g.,  $Ca^{2+}$  and  $Mg^{2+}$ ) should significantly affect atmospheric  $CO<sub>2</sub>$  and ocean  $CaCO<sub>3</sub>$ saturation; however, Earth system carbon cycle models do not accurately account for these effects. To address this, we couple the newly developed MarChemSpec speciation model with the cGENIE Earth System Model to evaluate the impact of seawater chemistry and carbon speciation on ocean carbon and alkalinity storage under early Eocene conditions. We find that, in isolation, ion content and subsequent speciation effects raise the Eocene ocean carbon inventory by ~400 PgC and lower ocean alkalinity by  $\sim$ 10  $\mu$ mol/kg when compared with the older correction factors previously used in cGENIE. Our findings highlight the sensitivity of reconstructed carbon and alkalinity inventories and steady-state budgets to the choice of modeled carbon system corrections and bulk ocean chemistry. Ultimately, this research demonstrates the equal significance of carbon and alkalinity budgets in setting atmospheric  $CO<sub>2</sub>$ , thereby supporting alkalinity enhancement as a strategy for future  $CO<sub>2</sub>$  removal and climate change mitigation efforts.