

I come to review this manuscript from a paleo perspective. This study by Cheng et al. has provided fresh and deeper understandings on simulating the marine iodine cycle in the cGENIE model. I find the model ensembles are well designed from both modern and paleo angles. I agree with the authors that the simulations have overall good matches with modern observations and paleo data. The authors have also offered their detailed evaluations on model performance from three perspectives. Their explanations of model-data mismatches are reasonable and have pointed out some future research directions. I think the manuscript is well written and the main points are very clear.

I do have two questions regarding the paleo simulations. It has been speculated that the total iodine concentration in seawater in the geologic past may be different than modern oceans (Zhou et al., 2016 Paleo; Lu et al., 2018 Fig. S12). But the Cretaceous simulations seem to use modern total iodine value? If you used a higher total iodine in the pre-OAE simulations, I assume it will bring all the model-simulated I/Ca to higher values, thus presumably closer to observations?

Thank you for addressing this point. Yes, the total iodine concentration in the Cretaceous cGENIE simulations is 500nM, the same as modern values. Importantly, total iodine can be changed in the model and the impacts of total iodine changes on surface iodate distribution were previously tested in Lu et al., (2018), figure S12 (see below). One interesting outcome is that lower total iodine led to more limited iodate reduction and a relative increase in the proportion of iodate in the surface.

Further, as discussed by Zhou et al., (2015), the total marine iodine budget could have varied due to increased continental weathering or organic carbon burial. However, no such evidence has been reported to date for iodine inventory changes across OAE2.

To clarify this, a brief discussion will be added to L636:

“It is possible that the total iodine inventory has varied through Earth history relative to the modern-day value (~500 nM), which was adopted for our Cretaceous model. Indeed, the overall underestimated I/Ca by cGENIE might be the result of overall higher Cretaceous total iodine inventory (Zhou et al., 2015; Lu et al., 2018). However, such difference is easily masked by local-regional redox variation and is thus difficult to track (Zhou et al., 2015). Due to the lack of evidence otherwise, we assume the average total iodine during the Cretaceous is close to the modern, and the consistent I/Ca

underestimation is caused by uncertainty in model simulation.”

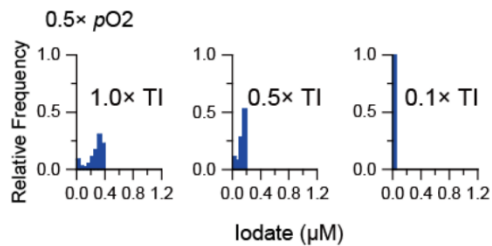


Fig. S12. Surface-ocean iodate IO_3^- distributions with decreasing ocean iodine inventory. These frequency distributions do not reproduce the lognormal type of distribution as observed in Paleozoic I/Ca data. Modern fluxes of iodine input and output from seawaters may be orders of magnitude lower than that of iodine recycling in the water column (7). IO_3^- (instead of I^-) sorption on organic matter at the sediment-water interface is a major sink of iodine, which appears to be stabilized by a negative feedback between the amount of organic matter as a substrate on the seafloor and bottom-water oxygenation preventing IO_3^- reduction (52). No evidence supports either global organic matter burial rate or bottom water $[\text{O}_2]$ level mimicking the I/Ca record. Therefore, secular changes in total iodine concentration are also unlikely to dominate the trends observed in our I/Ca record.

L593-596: The conversion from seawater IO_3^- and Ca^{2+} to I/Ca may be more complex than the authors have suggested. For example, the substitution of IO_3^- into calcite may involve Na^+ , CO_3^{2-} ions (Podder et al., 2017 GCA); the seawater Ca^{2+} concentration in Cretaceous may be different than modern day, so whether Cretaceous Ca^{2+} is well-simulated needs to be considered. I understand this may be beyond the scope of this model-focused study, but I recommend the authors should at least acknowledge such complications.

Thanks for the very helpful suggestion. An additional comment will be added between L593-596 to address these uncertainties:

“Beyond temperature, we acknowledge that IO_3^- incorporation into carbonate lattice through substitution $\text{IO}_3^- + \text{Na}^+ \leftrightarrow \text{CO}_3^{2-} + \text{Ca}^{2+}$ is controlled by $[\text{Na}^+]$, $[\text{CO}_3^{2-}]$, and $[\text{Ca}^{2+}]$ (Podder et al., 2017). However, either quantifying these ions during the Cretaceous seawater or quantitative calculation of ion substitution dynamics requires further constraints. Although uncertainties are inevitable, we assume our temperature- controlled $[\text{IO}_3^-]$ -I/Ca conversion based on current quantitative knowledge meets the requirement for Cretaceous model-data comparison.”

Minor comments:

L65: strictly speaking, it should be “regional rather than in-situ redox conditions”

Will adopt reviewer’s suggestion for more accurate wording.

L78: I- re-oxidation

Will correct the typo.

Fig. 6 caption: may add a short note to refer readers to see transect locations shown in Fig. 1

We will either add a note to Fig. 6 caption or change to a new Figure 1 and add a map view to Fig. 6.

L373: strictly speaking, these papers studied both planktic and benthic forams

Will change “benthic foraminiferal” to “planktic and benthic foraminiferal” according to the reviewer’s suggestion.

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