Answer to Anonymous Referee #1:

The authors used numerical models and an optimization technique to explore how the spatially variable Zn:P uptake ratios by phytoplankton can be constrained by synthetic observations. The authors focused on how limited spatial data coverage, model circulation uncertainty, and the choice of objective function, which constitute three major sources of uncertainty in data-model assimilation studies, influence the optimization results. I found the results informative and potentially important to the marine bio-geoscience community. The manuscript is overall well-written except for some minor points listed below.

We thank Anonymous Referee #1 for taking the time to review our manuscript and for her/his positive assessment and valuable comments.
Below, we include our detailed answers to all comments and questions.

Main point:
(1) The only major concern is that this study might appear to be esoteric and technical to many readers if model-derived synthetic data are the only target. More specifically, the numerical experiments performed and the results discussed here seem to be an important preliminary step towards using the real (not synthetic) observations to constrain the model formulation of the variable Zn:P uptake ratios. Why don't the authors use the best observation data coverage, the best ocean circulation model, and the best objective function to suggest the best estimate for the relationship between Zn:P uptake ratios and Zn? Has this optimization been done already or is this beyond the scope of the current study? When I came to the section 3.7, I expected something along the line, but was disappointed by reading what has been already written and some discussions only. Perhaps, a previous study already found an optimal estimate for the parameter set, which was used as a reference parameter set in this study? Even if so, it would be worth being stated.
We appreciate the questions raised in this comment, and have re-focused Section 3.7 in response (the revised text is included below).
Model optimisations to real data have indeed been carried out previously, without analysis of the sensitivity to data distribution or misfit function. Our choice to focus on synthetic observations is motivated by the need to assess such sensitivities. In the revised manuscript, we (i) explicitly raise the awareness of the currently largely lacking discussion about impacts of the choice of misfit function and model imperfections, (ii) refer to a previous optimisation study using real Zn data, and (iii) highlight how implications can be drawn from our study with regard to model calibration towards real data *and* with regard to interpretation/discussion of optimisation results. We hope that this addresses the referee's concerns about Section 3.7. Elsewhere in the manuscript, we have extended our reasoning for discussing the results mainly in the light of uptake systematics and large-scale export fluxes when we introduce the Figure that shows the results of synObs experiments as their resulting uptake systematics (Fig. 6), and we clarified in the Methods section that the choice of our reference parameter set was based on Eq. (2) to best fit to *E. huxleyi BT6* culture data of Sunda and Huntsman (1992).
More broadly, we would like to strongly assert that our analysis of the influence of data distribution and subjective choices during optimisation is fundamental to the question of what scientific inferences can be drawn from objective model optimisation. Assessing this influence rigorously is only possible with full control on the observations, hence our study's focus on optimisation to synthetic observations. As such, we would contend that it is neither esoteric nor technical.

Revised Section 3.7 now reads:
"Although there are modelling studies of marine trace metal cycling that objectively calibrate a variety of their model parameters (e.g. Frants et al., 2016; Weber et al., 2018; Pasquier et al., 2022), the impact of data distribution, model

imperfections or choice of misfit function on optimisation results are often not discussed. In this study, we have separately assessed how optimisation results are impacted by these sources of uncertainty. In accordance with other work (e.g. Löptien and Dietze, 2019; Kriest et al., 2020), our ensemble of optimisations shows that biogeochemical parameters are often optimised to compensate for the inability of model formulations to reproduce the target field (Sect. 3.3). Reconstructed Zn uptake systematics were most different from the reference uptake systematics in experiments with systematic differences between the large-scale circulation of the model and that underlying the target field (synObs_circ; Fig. 6); misfits obtained in this experiment type were also about an order of magnitude higher than those for synObs_seas, which differ only in terms of the presence or absence of seasonality within the same physical model. While optimisation to real Zn data (e.g. Weber et al., 2018) is outside this study's focus, the results of our ensemble have direct implications for such optimisations and the inferences that may be drawn from them:

- Because biogeochemical parameters are often optimised to compensate for the inability of model formulations to reproduce the target field, any optimisation of a simple biogeochemical model such as ours towards real data must be seen as attempting to retrieve the systematics of biogeochemical behaviour, rather than physically meaningful parameter values. This is especially the case since – even though observations from wild phytoplankton (Twining and Baines, 2013) indicate geographical systematics that are similar to those that result from this model formulation (de Souza et al., 2018) – there is no reason to believe that the stoichiometry of Zn:P uptake in the real ocean should follow a single dependence on dissolved Zn concentration.
- Because increasing both spatial and temporal model resolution might be computationally unaffordable, even for a relatively efficient global optimisation algorithm like CMA-ES, it is important for studies focusing on optimisation towards global (micro)nutrient distributions with long whole-ocean residence times to prioritise the choice of circulation model, with special focus on accurate simulation of large-scale circulation timescales.
- It is important to recognise the subjectivity that the choice of misfit function introduces to objective parameter optimisation, and to carefully weigh the sensitivities implicit to the misfit function in making this choice for any particular application. A misfit function that appears suitable for optimisation in a simple TWIN experiment, in which the model can perfectly describe the target field, may not be the best choice for optimisation towards noisy, incomplete and/or irregularly distributed real-world data.

It should also be emphasised that our study has not considered the influence of model simplifications, such as the lack of external sources of Zn or simplifications in the underlying P cycling model. External inputs such as those from marginal sediments, atmospheric deposition, or hydrothermal vents (e.g. Conway and John, 2014; Roshan et al., 2016; Lemaitre et al., 2020; Liao et al., 2020) are not relevant to our optimisation ensemble to synthetic observations, but their potential significance should be taken into account during optimisation to real data. With regard to the underlying P cycling model, it directly affects Zn cycling in our model formulation, since Zn uptake is related to PO_4 uptake through $r_{Zn:P}$ (Sunda and Huntsman, 1992), and Zn remineralises with the same globally constant length-scale as P (Twining et al., 2014). […]".

Minor points:

(1) I can see the role of three parameters, i.e., a, b, and c in Equation (2), in determining the relationship between Zn:P uptake ratio and Zn. However, how the ligand concentration L is controlling the relationship is not clear. Is there a formulation relating L and the Zn:P uptake ratio or a formulation relating L and Zn2+?

Thanks for pointing this out (Referee 2 raises a similar point). We have introduced the derivation of Zn^{2+} from our tracer Zn and the ligand concentration L. The revised paragraph in Sect. 2.1.2 now reads:

"Following Ellwood and Van den Berg (2000), concentrations of Zn^{2+} are calculated from total dissolved Zn (the tracer carried in the model) in two steps: first, by assuming rapid equilibration of non-ligand-bound Zn (Zn') with an organic ligand with conditional stability constant K_L=10^{10} M^{-1} and spatially constant concentration, which allows calculation of Zn' by solving the quadratic equation:

$$K_L \cdot (Zn')^2 + (K_L \cdot L - K_L \cdot Zn + 1) \cdot Zn' - Zn = 0 \quad (3)$$

and second, by calculating Zn^{2+} from Zn' using the inorganic side-reaction coefficient α_{Zn} = 2.1:

$$Zn^{2+} = \frac{Zn'}{\alpha_{Zn}} \quad (4)"$$

(2) In Figure 2, the line color for the parameter "a" does not match between the legend and plots. In the legend, it looks like purple to me while it is black in the figures.

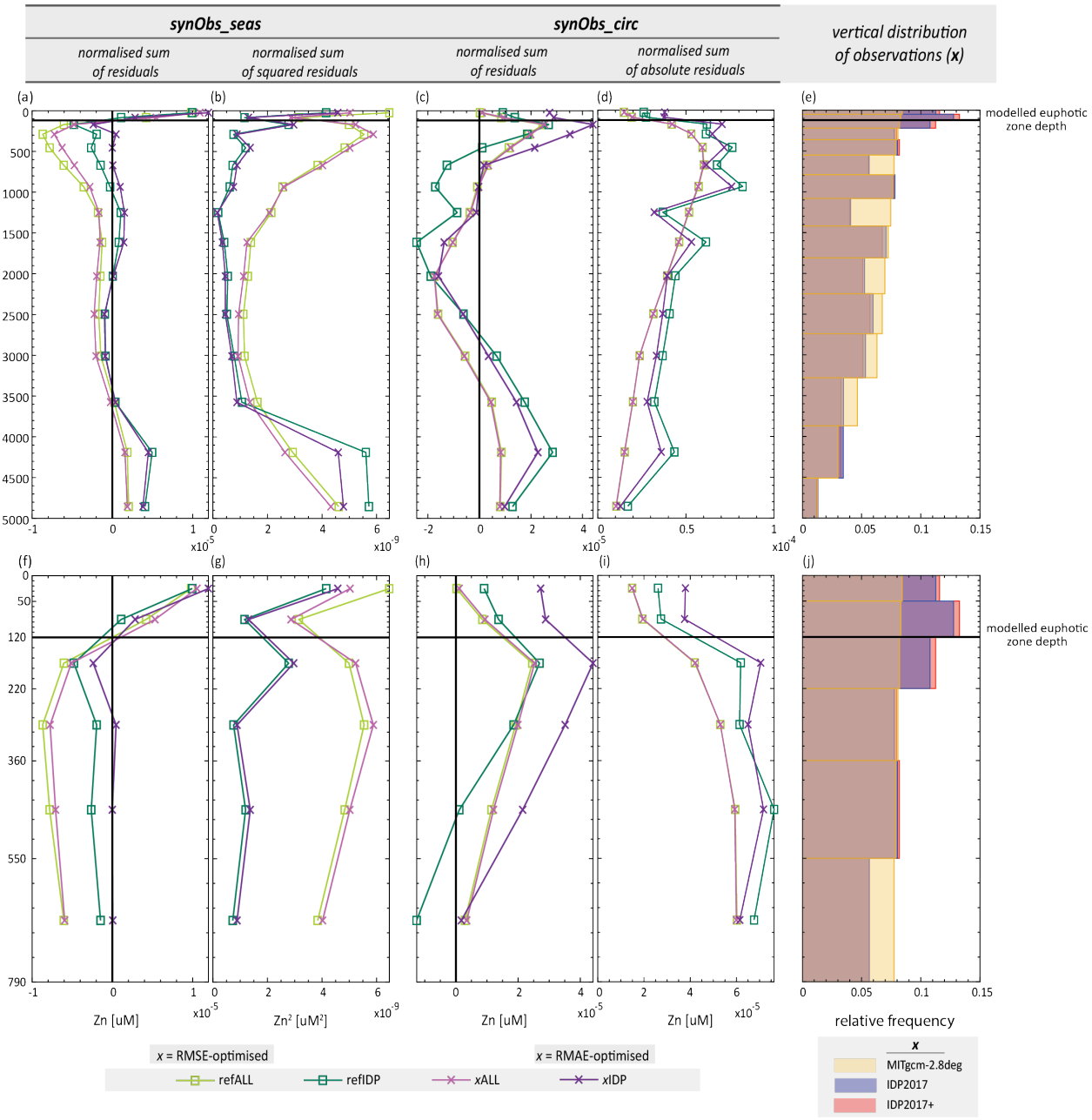
That was a mistake in our Fig.; we have edited the legend.

(3) I am having difficulty in interpreting Figure 10. What do the X-axes represent in panels (e) and (j)? What do the different lines represent in other panels? (e.g., 'refALL', 'refIDP', 'xALL', and 'xIDP')? Are these labels defined in the text or in the figure caption?

We agree that Fig. 10 and its legend can be clarified, and have edited it (see below). In panels (e) and (j), we show relative frequency distributions considering only the vertical distribution of observations – this is now explicitly stated by an axis label. We distinguish between 3 different observational sets (MITgcm-2.8deg and IDP17[+]), as clarified in the legend below this. We have also moved the legend for panels a-d and h-i, and hope the figure is more legible as a result.

(4) The authors discussed additional uncertainties that would rise when applying the optimization to the real observation data in Section 3.7. What about uncertainty in external inputs of Zn (aeolian deposition and coastal sediments, etc.) to the ocean surface? Is it minor compared to the uncertainty associated with model parameterizations of biogenic Zn cycles?

We agree. While not relevant to our optimisations (since there are no external inputs influencing the synthetic observations) these processes may be relevant for optimisations to real data. We have added a short discussion of this in Section 3.7 (see text of this section above).



Caption: "Figure 10. […] Panel **(e)** shows relative frequency distributions of the vertical distribution of three different observational sets, which are the non-reduced observations, i.e. MITgcm-2.8deg, IDP2017, and IDP2017+ and panel **(j)** represents a zoom-in thereof."

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