

Pasquier et al has presented an interesting inverse model of Nd based on OCIM, and performed Nd cycle parameter optimization against observations of seawater Nd and eNd. Such models have been developed for many other biogeochemical tracers in the GEOTRACES era, and it's great to see that it is now applied to Nd. I think the model has a potential for wide use given the computational efficiency compared with GCMs or ESMs, and obvious advantage over box models.

From the modeling perspective, I found the OCIM modeling framework easy to understand as it has matured through its history of modeling biogeochemical tracers. The optimization procedure here still has more to be desired, though the inherent difficulty is understood. Dr. Pasquier should be applauded to have taken considerable effort to develop various open source Julia packages devoted to ocean biogeochemistry under the transport matrix framework of AIBECS.jl, of which GNOM.jl is a special case for Nd.

However, from the Nd cycle perspective, as an observationalist and a geochemist, I disagree with the treatment of the Nd sources in the model. Here are my main criticisms.

1. There is a disregard of the huge literature on observations and measurements of globe Nd sources and the relevant parameters, including dust/ash solubility, riverine/groundwater Nd concentrations and eNd, benthic flux and scavenging. Such that the "optimized" parameters are not evaluated against measurements, leading to values that are unrealistic: it's hard to accept the model "optimized" value being 9 orders of magnitude higher than measurements (in the case of scavenging Kxs), or exact opposite to the trend in observations (in the case of benthic flux).

2. Despite the "optimized" parameters being orders of magnitude different from observations and previous models, this study arrived at global integrated sources similar to previous studies, suggesting that the Nd source parameters can NOT be uniquely determined ONLY by optimizing the seawater Nd and isotopes data. Either that the parameterizations are incorrect, or that a proper inversion should include measurements of the Nd sources in the cost function. In this respect, proper evaluation of the parameter covariance is required before optimization.

3. The parameterizations for Nd sources often make unsubstantiated assumptions on secondary issues that we have no constraint on, while adopt overly simplistic, even incorrect, representations of the first-order variables that we do have fairly good constraints on. For example, in the riverine parametrization, the authors use the detail reconstruction of global river discharge, which is only a secondary parameter in the Nd flux, yet force the river Nd concentration, the first-order parameter, to be constant despite observations that showing it varying by 2~3 orders of magnitude. Another example is that when treating the sediment flux, a complicated and arbitrary parameterization was created for the differential release of Nd from certain lithology (of secondary important, even smaller than analytical uncertainty), while ignoring that the baseline sediment flux eNd (the first-order variable) map of Robinson has large uncertainties.

Overall, without proper considerations of the actual measurements of the Nd source parameters, I disagree that the black box optimization approach can give any meaningful insight into the Nd cycle compare to previous studies.

Following are my comments on the parameterizations of Nd sources, and optimization approach and some other issues.

Dust

- 40 ppm Nd in dust is higher than that in PAAS (34 ppm) and UCC (27 ppm) (Rudnick and Gao, 2014). Why is this number chosen?
- There are observations of dust eNd and solubility (Goldstein et al., 1984; Robinson et al., 2021), which are disregarded. Asian dust on average have an eNd of ~ -10 (Chen and Li, 2011), the “optimized” value of -7.6 seems too high. Australia is mainly made of old cratonic rocks, how could it supply a dust source as high as -4.0 ? Similarly, it’s hard to image that the North American source could be as radiogenic as -4.3 . It’s difficult to accept the optimized values based on our knowledge of local geology. The parameter range used for optimization seems too narrow to me for many regions.
- The “optimized” ($\sim 20\%$, even up to 80% !) solubility is orders of magnitude higher than observations ($\sim 1\%$) (Greaves et al., 1994). Not surprise that this study gives a dust contribution to the global Nd budget orders of magnitude higher than previous models.
- L480. This is a misunderstanding of the dust flux in (Tachikawa et al., 2003). The “known” dust flux used by Tachikawa is only 2.8 Mmol/yr , like the GCM models. In fact, Tachikawa is the main source of dust flux in the GCM models cited, that’s why they are similar. The extremely high 60 Mmol/yr number cited (I think this is a miss citation. It should be 42 Mmol/yr in Tachikawa, I don’t find this 60 Mmol/yr number) is only “fictional”, that Tachikawa suggested that if it is true then we can solve the “missing source” problem. But there’s no evidence that it is true. Thus, it is safe to say the optimized dust flux in this study is one order of magnitude higher than all previous estimates.

Ash

- L180. Explosive eruptions mainly produce felsic/rhyolitic ash that can travel long distance. Mafic eruptions are generally effusive and the resulting ash doesn’t travel far (Scudder et al., 2016).
- L181. I don’t find any mention of volcanic ash in Chien. So where are the flux data from?
- L190. Ash eNd is varied. See the global volcanic arc eNd data of (Kelemen et al., 2014). It seems the authors have chosen an initial value of $+10$, which already is the upper limit of volcanic materials. The “optimized” ash eNd of $+13$ is clearly unrealistic.
- The “optimized” ash solubility of 76% is extreme. Using the ash leaching experiment of (Du et al., 2016) we can get a ash Nd solubility of $\sim 1\%$. The argument that ash is more soluble than dust is somewhat an urban legend without clear evidence. In situ observations also show no effect of a recent Icelandic ash fall on surface seawater eNd (Lambelet et al., 2016). Regardless, 76% seems unrealistic.

Sediment

- The sediment source parametrization is completely unsubstantiated and published sediment flux data have been disregarded. I see no rationale to support the assumption that benthic flux at the deep ocean is zero, which is clearly contradicted by observations (Abbott et al., 2015; Du et al., 2020) that instead show the highest benthic flux is from the deep ocean. The recent study from the abyssal plain of the Pacific (even at $\sim 5000 \text{ m}$) still show a benthic flux of $3.5 \text{ pmol/cm}^2/\text{yr}$ (Haley et al., 2021), similar to what’s observed on the Oregon shelf. Based on the existing data (Du et al., 2020), benthic flux either doesn’t change with depth, or increases with depth, opposite to the

parameterization here. It has also been show that benthic Nd flux doesn't correlate with POC flux or bottom water O₂ (Du et al., 2018).

- L210. If linear increasing is allowed, then z₀ should be allowed to be negative, rather than forced to be positive.
- L219. This is another urban legend with unclear evidence.
- L226. Evidence? Check (Blaser et al., 2019) which estimated the benthic flux from freshly eroded Heinrich Layers. It doesn't seem there is reason to believe it to be much higher than elsewhere.
- L240. This parameterization seems unnecessary if the resulting difference is only 0.03 epsilon, given typical analytical uncertainty of ~0.4 epsilon, and the reproducibility of sediment digestion/leach is ~1 epsilon or more. The uncertainty of the gridded product of Robinson is probably more than 5 epsilons because of the poor spatial coverage of the raw data and the inconsistency in the types of raw data (leach residual, bulk sediment, size fractions etc.). Thus, the biggest uncertainty to capture is that of the Robinson dataset, which should be allowed to vary 5~10 epsilon in the model.

River

- I disagree with the parameterization of riverine input. In my option the necessary source data should come from: (Bayon et al., 2015; Goldstein and Jacobsen, 1987, 1988a, b; Goldstein et al., 1984). I would rather use the measured riverine Nd source, however limited, than using clearly incorrect parameterization, however globally detailed. This is an example of ignoring the first order issue, i.e., the variations in riverine Nd concentrations, while overly concerned with the secondary issue, i.e., the river discharge.
- L249. Riverine Nd concentrations vary by 2 orders of magnitude (Goldstein and Jacobsen, 1987).
- L255. How is the smoothing done? What is the spatial scale? The effect of riverine input doesn't extend beyond the estuary. What is in fig2d seems too extreme to be true.
- Where is the estuary removal term in the parametrization?
- L256. I disagree with the use Robinson data for this purpose. Remember the margin eNd of Robinson is an interpolated product based on rough geological map with very limited outcrop rock eNd, which has nothing to do with rivers. The relationship between riverine eNd and the eNd of the rocks from the drainage basin is a complicated matter (Bayon et al., 2015). There are measurements of riverine dissolved and particulate eNd from (Bayon et al., 2015; Goldstein and Jacobsen, 1987, 1988a, b; Goldstein et al., 1984) that should be used.

Groundwater

- Same problems as the riverine parameterization: groundwater Nd concertation is not constant, and it varies by 7 orders of magnitude! I see no reason to use the Robinson eNd dataset either. We already have the measured Nd flux and eNd from (Johannesson and Burdige, 2007), why would you use parameterizations that's unsubstantiated.
- What is the removal parameterization of the groundwater flux upon entering the ocean?

Hydrothermal

- Hydrothermal system is a net SINK of Nd (Stichel et al., 2018; Basak et al., 2021). The release of hydrothermal Nd makes local seawater eNd only slightly more radiogenic (within analytical uncertainty), while the seawater Nd concentration decreases significantly because of scavenging by

hydrothermal particles. Therefore, both a source and a sink should be implemented for the hydrothermal system, with the sink being much larger than the source.

Scavenging

- L316. I disagree. For some reason the most important Nd scavengers, Fe-Mn oxides (Schijf et al., 2015; Sholkovitz et al., 1994), are ignored like previous models. And I don't see how scavenging by Fe-Mn oxides can be treated as "precipitation" as it will depend on oxide concentrations that are redox sensitive.
- The "optimized" Kxs are unrealistic. Previous models generally use values around 10^6 (Siddall et al., 2008; Arsouze et al., 2009). Lab measurements also give results on similar orders of magnitude (Schijf et al., 2015). Yet here the optimized values are $10^{13} \sim 10^{15}$!
- I think that such high optimized Kxs are the results of overly high dust flux in the model (see my comments up), so extremely intense scavenging is needed to bring surface Nd concentration down.

Optimization

- There are so many parameters to be optimized. What is the covariance structure of them? For example, the dust solubility will probably covary with the scavenging Kxs. Therefore, can we get a unique solution of the parameter values? These questions need to be studied before performing optimization. That so many of the optimized parameters are unrealistic give me little confidence in the optimization.
- L335. It is nice to include penalty terms for the parameters, but I think the choice of parameter ranges are not very reasonable given that the optimized values are often unrealistic.
- The optimization should be penalized against not just measured seawater Nd/eNd, but also measurements of Nd sources.
- A volumetric weighing should be included in the cost function, so the optimized results are not biased toward the surface ocean because of high data density. The Nd community is generally more concerned about the deep ocean than the surface, in contrast to other communities, for several reasons: 1 Surface eNd is heterogeneous and local, thus contains little global information. 2. Surface input generally varies a lot in the past, so surface eNd isn't useful for watermass tracing. 3. We can only reconstruct deep ocean eNd in the past, as there is no archive of surface water eNd.
- Additionally, basinal weighing could be useful given that the data is so concentrated in the Atlantic.
- How does the choice of the prior affect the results? For many parameters we know very little but the orders of magnitude. Isn't a uniform prior more reasonable than lognormal/logit priors when we essentially have no information on the actual distributions?
- L400. This needs to be demonstrated. And are the ranges of the randomized initial values large enough that we can really believe the results are global?

Minor points

- The unit of eNd is epsilon, not per 10 thousand. This is a radiogenic isotopic ratio, not a stable isotopic ratio. The expression of epsilon looks like delta for stable isotopes, but the reporting convention is different. I have never seen the per 10 thousand unit for eNd in literature.

- L129. Not because there's no stable isotopic fractionation, but because by convention when reporting radiogenic Nd isotopes stable fractionation effect, be it natural or instrumental, is removed by normalizing to constant $^{146}\text{Nd}/^{144}\text{Nd}$ ratio. That is, even if you model isotopic fractionation, you have to remove it when you convert to ϵNd units.
- L408. Needs demonstration.
- Fig9/10. It's very difficult to see the "fit". Instead show the model-date misfit rather than just overlay the data on top of the model.
- L484. If the optimized solubility is not supported by observations, then there is no reason to believe it.
- L487. I disagree with this philosophy of "consistent with previous model" but ignoring actual observations of the fluxes (Du et al., 2020; Goldstein and Jacobsen, 1987; Tachikawa et al., 2003; Johannesson and Burdige, 2007).
- L522. Seem exaggeration. What about the MIT-ECCO, which has also been used as TMM?
- L545. Perhaps not exactly the same calculation, but similar concept of partitioning N/S sources already exist (Gu et al., 2020)
- L571. The most prominent caveat is the disregard of observations of Nd sources.
- L543. Disagree, parameter choices often unrealistic.

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