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Multi-phase ecological change on Indian subcontinent from the late Miocene to Pleistocene recorded in the Nicobar Fan

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Supplementary Material

The TOC/TN ratio of end-members (particularly terrestrial C_3 and C_4 plants) are not well constrained, and inorganic N adsorption onto mineral surfaces appears to be generating lower-than-expected TOC/TN ratios. These effects could conceivably affect the results of the mixing model we used to correct for contributions from marine organic matter. To address these concerns, we ran sensitivity tests of the mixing model that uses TOC/TN and $\delta^{13}C$ to estimate C_4 plant coverage through time. Different C_3 and C_4 end-member TOC/TN assignments changed the quantitative estimates of C_4 coverage but had little effect on the temporal trends and are therefore unlikely to affect the interpretation that there were two distinct episodes of C_4 expansion beginning at ~7 and ~3.5 Ma as well as a pulse at ~2.5 Ma (Figure S1). The additional spike at ~1.75 Ma in the bootstrapped records of Figure S1is difficult to attribute to physical causes due to the sample sparsity after about 2 Ma.

Adsorption of inorganic N onto mineral surfaces could artificially lower the measured TOC/TN, which would cause the mixing models to erroneously over-correct for marine organic matter. We modelled this case by generating a synthetic dataset with prescribed C_4 coverage and assuming a TOC/TN ratio of 55 (similar to the maximum in our dataset) for the C_3 and C_4 end-members. The TOC/TN values of the resulting synthetic dataset were then divided by a factor ranging from 2–5 to simulate artificially low TOC/TN values. The

resulting synthetic data were then inverted in the same way as the real data using a range of 8–55 for terrigenous C_3 and C_4 end-members (Figure S2). This procedure mimics the effects of adsorbed inorganic N by producing data with TOC/TN ratios that are artificially lowered from the end-member compositions used to calculate C_4 coverage. We would expect adsorbed inorganic N to result in an over-correction for marine organic matter since it would produce TOC/TN values closer to the marine range. This would tend to underestimate C_4 abundance when the $\delta^{13}C$ of the data is less than that of the marine end-member and overestimate C_4 abundance when the $\delta^{13}C$ of the data exceed the marine value. Indeed, we find that the tests underestimate C_4 coverage at low values (i.e. when $\delta^{13}C_{TOC}$ values are lower than marine $\delta^{13}C$) and overestimate C_4 abundance at high values ($\delta^{13}C_{TOC}$ similar to or greater than marine values; Figure S2).

While these results underscore the uncertainty in the absolute value of quantitative vegetation coverage estimates, they also indicate that our interpretations are unlikely to be significantly affected. Prior to the first episode of C_4 expansion ~ 7 Ma, C_4 coverage is low and the $\delta^{13}C_{TOC}$ is less than the marine end-member composition (Figure 3), implying that artificially lowered TOC/TN ratios would result in an underestimate of the true C_4 abundance. Therefore, the magnitude of C_4 coverage during this interval is likely to be a lower bound, and the ~7 Ma C_4 expansion is unlikely to be due to an artifact in data processing. For the second episode of C_4 expansion beginning ~ 3.5 Ma, the $\delta^{13}C_{TOC}$ values of the data are still generally less than that of the marine end-member, meaning that inorganic N adsorption is still likely to underestimate true C_4 abundance. Around the 2.5 Ma transition, the $\delta^{13}C_{TOC}$ values begin to overlap significantly with the range used for marine $\delta^{13}C$, meaning that the mixing model may begin to overestimate C_4 abundance by a small amount. Figure S2 also shows that a combination of artificially lowered TOC/TN and a major change in the fraction of organic matter from marine sources could cause the model output to

indicate a spurious increase in C_4 plants. However, even artificially reducing the TOC/TN by a factor of five does not yield the same change in inferred C_4 coverage as the ~2.5 Ma step and would imply a 4:1 ratio of inorganic to organic N in our samples, which is unrealistically large. Therefore, the increase in modelled C_4 coverage around 2.5 Ma is unlikely to be an artifact of the data processing routine.

Two methods of filtering the data also support our interpretations. Considering only the subset of the data with TOC/TN ratios that exceed typical marine values will both reduce any potential effects of N adsorption as well as the influence of marine organic matter. Figure S3 shows the mixing model results considering only samples with TOC/TN above eight, and the familiar increases in C_4 fraction at ~7 and ~2.5 Ma are again apparent, but the onset of the second phase of C_4 expansion is less clear. Increasing the TOC/TN threshold to 10 gives similar results though with a compromise in temporal resolution. Additionally, Meyers (1997) suggested that marine sediment samples with TOC above ~0.3 wt% C were unlikely to show significant alteration in TOC/TN due to inorganic N adsorption. Figure S3 shows the results of our mixing model when only samples with TOC > 0.3 wt% C are considered, and it is clear that this procedure does not significantly alter our interpretations. These results further support the robustness or our interpretations.

Figures

Figure S1

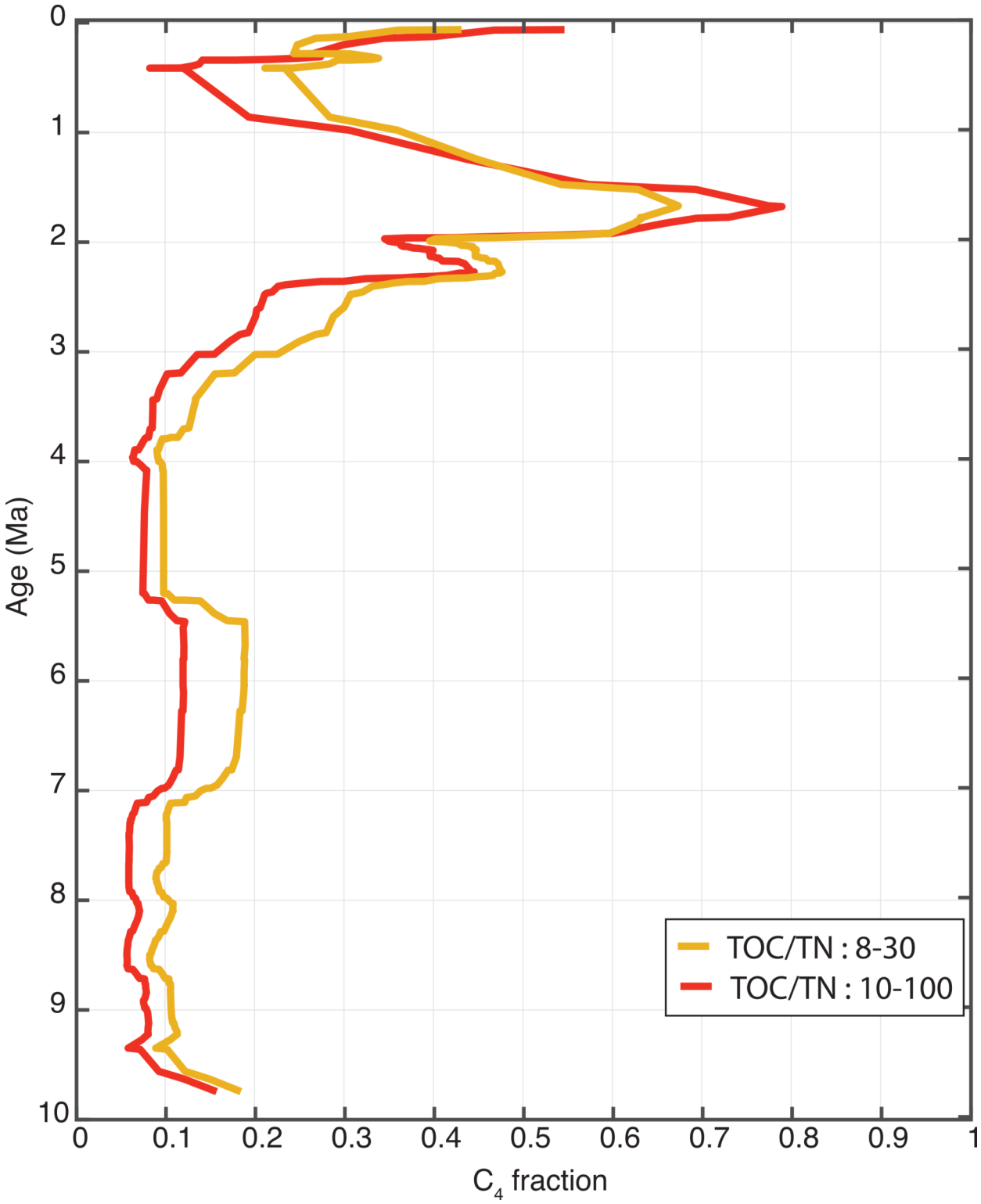

Figure S2

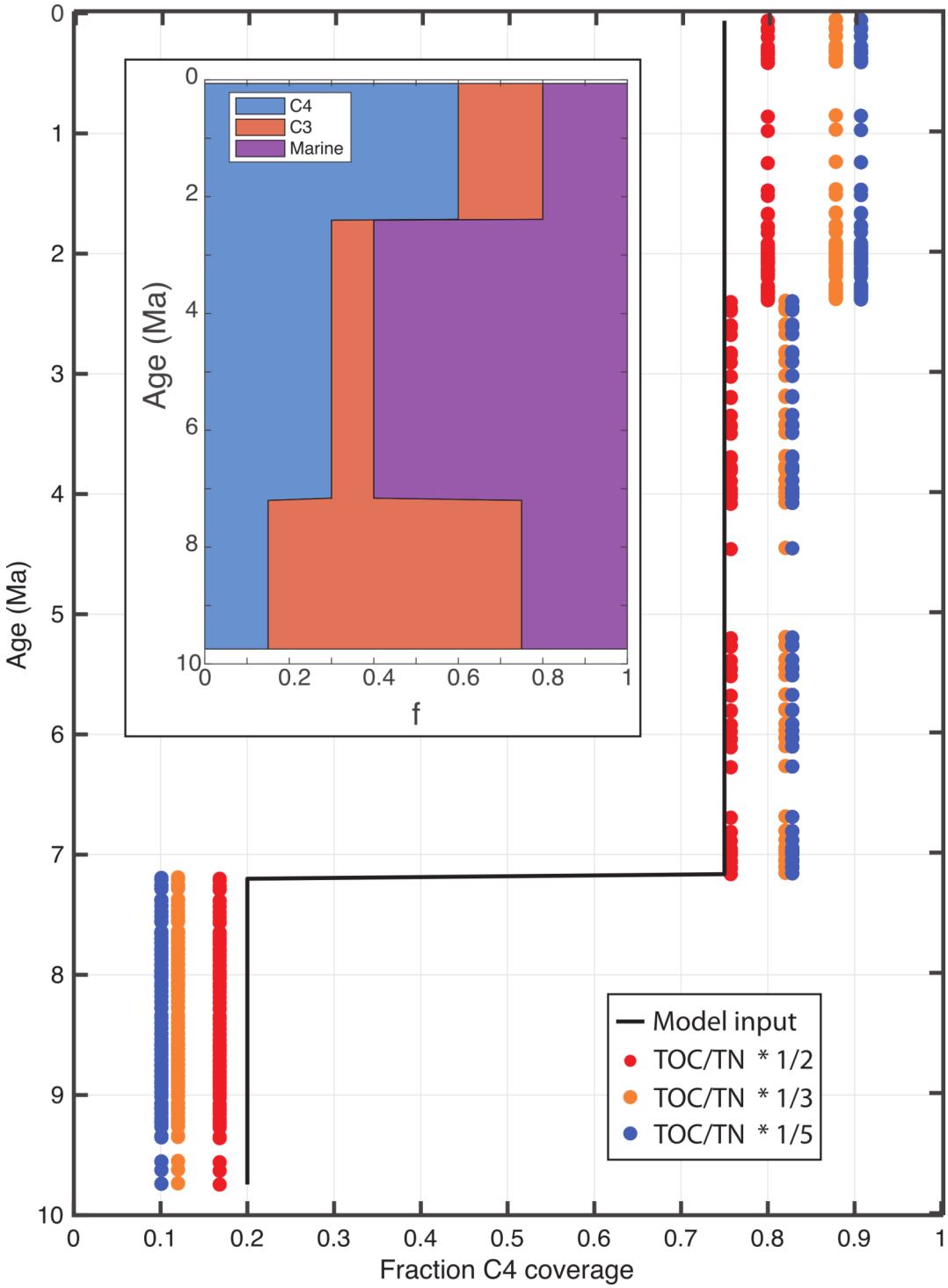
Age (Ma)
Fraction C4 coverage
Model input
TOC/TN * 1/2
TOC/TN * 1/3
TOC/TN * 1/5
C4
C3
Marine
Age (Ma)
f

Figure S3

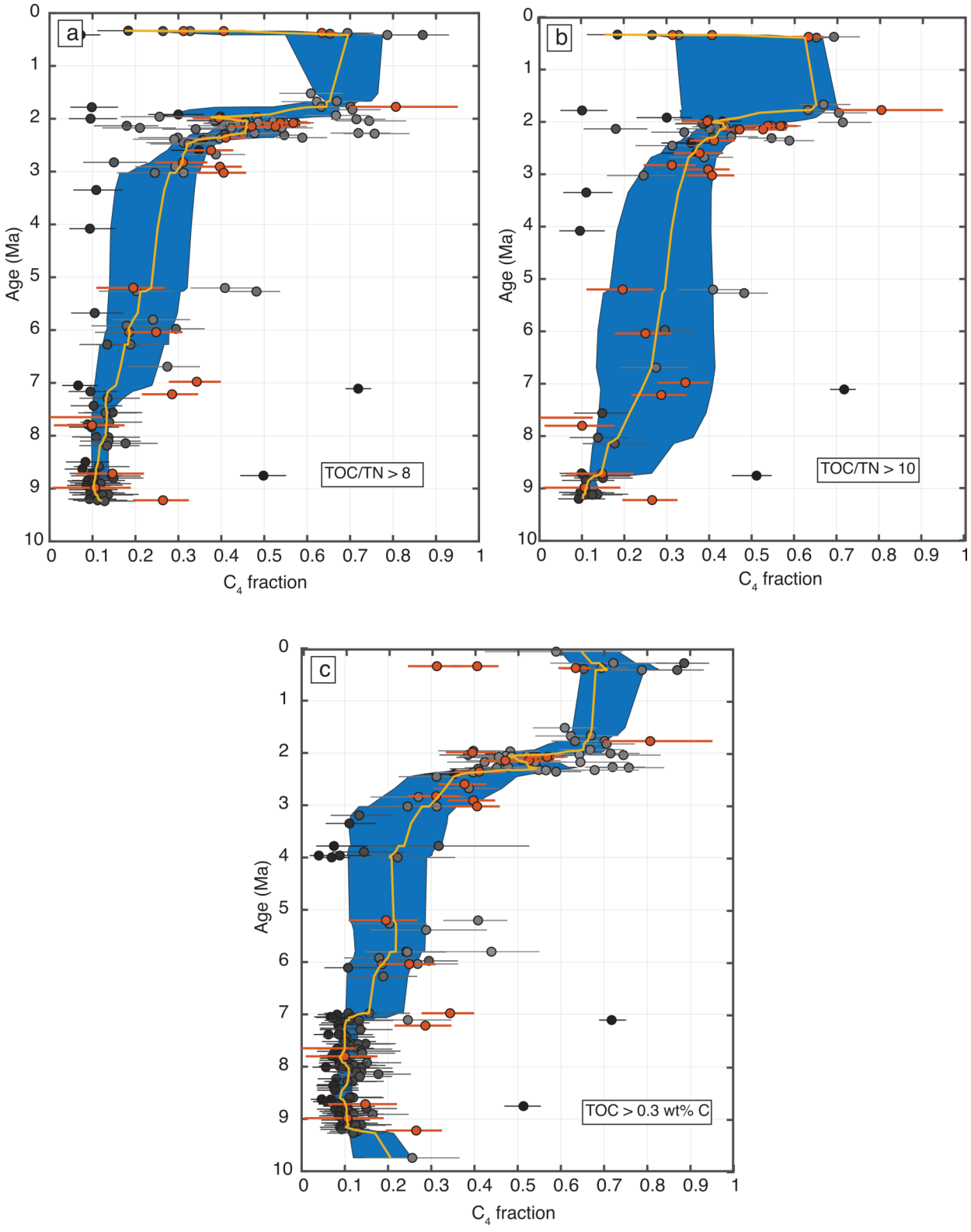
a
TOC/TN > 8
b
TOC/TN > 10
c
TOC > 0.3 wt% C
Age (Ma)
C4 fraction
0
1
2
3
4
5
6
7
8
9
10
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9

Figure captions

Figure S1. Results of sensitivity tests to determine the effects of varying the terrestrial end-member TOC/TN range. Smoothed model outputs (see Section 2) are shown for terrestrial TOC/TN of 8–30 (yellow) and of 10–100 (red). Inter-quartile ranges from Monte Carlo simulations and other information shown in Figures 5 and 6 are omitted here for clarity.

Figure S2. Simulation of the effects of additional inorganic N. The inset shows organic matter sources of a hypothetical dataset. The black line corresponds to the true fractional C_4 coverage while the coloured points demonstrate the model output assuming the TOC/TN ratios of end-members were artificially multiplied by a factor of 0.5 (red points), 0.33 (yellow points), and 0.2 (blue points). These values would imply that adsorbed inorganic N accounts for 0.5, 0.66, and 0.8 of the total N, respectively.

Figure S3. Sensitivity tests using more stringent data filtering criteria. In (a), only samples with TOC/TN > 8 are included, while in (b) only those with TOC/TN > 10 are shown. Based on the higher TOC/TN cutoffs, these data subsets are unlikely to reflect the inclusion of significant marine organic matter. In (c), only samples for which TOC > 0.3 wt% are shown, again reducing any possible reliance on data influenced by the inclusion of marine organic matter.