

## GLOBAL PATTERNS OF REACTIVITY AND SOURCES OF ORGANIC MATTER IN MARINE SEDIMENTS: A COUPLED MODEL AND LIPID BIOMARKER APPROACH

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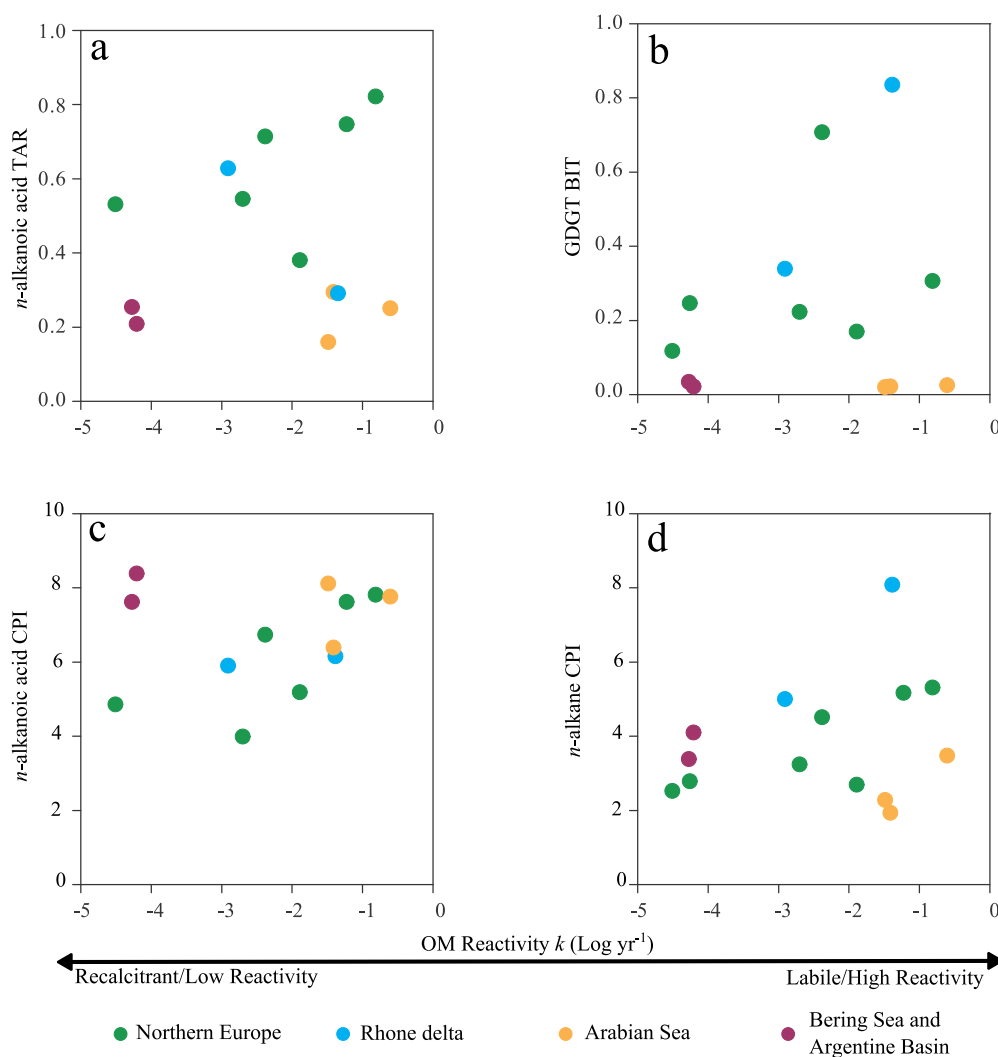
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Understanding the mechanisms dictating organic matter (OM) burial in marine sediments at a global scale is crucial for accurate assessments of carbon and nutrient cycling. The lack of a mechanistic framework that allows us to interrogate the main drivers behind OM degradation and preservation in different geographical and time scales limits our ability to produce robust forecasts of future perturbations on the global carbon cycle and Earth climate. Such a framework would help improve model parameterisation, and thus benefit both data-poor areas and time scales. Arndt et al. (2013) have demonstrated the overall lack of global-scale clear relationships between OM burial patterns and single characteristics of depositional environments. That represents a huge limitation in improving model parameterisation for a better representation of processes controlling OM cycling in marine sediments (Hülse et al., 2018). Therefore, there is a clear need to improve our understanding of OM burial in a broad sense, considering various aspects from OM sources, through transport, to final deposition. We use a multidisciplinary approach, in which we investigate OM sources through lipid biomarker compositions and compare those to quantitatively determined OM reactivity ( $k$ ) by means of Reaction-Transport modelling (RTM). We develop our coupled model-lipid biomarker approach for several depositional environments, comprising coastal, marginal, and deep-sea sites, within a range of depositional rates and sediment redox conditions. Our aim is to establish a global-scale quantitative link between OM reactivity ( $k$ ) and OM sources.

Our coupled model-lipid biomarker analyses reveal broad relationships on the controls of OM reactivity ( $k$ ) and OM sources. Model results show a general, albeit weak relationship between  $k$  and depositional environment characteristics (water depth and sedimentation rates), which complements Arndt et al. (2013) assessments. Regions characterized by low sedimentation rates, low OM fluxes, and located at great depths exhibit low  $k$ -values; whereas shallow sites with high sedimentation rates and fluxes of OM usually reveal high  $k$ -values. However, between those two end-members, we observe large scatter and variability. Similarly, lipid biomarkers ( $n$ -alkyl lipids and GDGTs) record an overall shift from terrestrial-derived to marine-derived sources with increase of water depth and distance from terrestrial sources. However, different molecular ratios record changes in OM sources in different ways across the depositional environment spectrum. Interrogating OM reactivity and sources together (Fig. 1 a–b) reveals no significant relationships ( $k - n$ -alkanoic acid TAR and  $k - GDGT BIT$ :  $r^2 < 0.05$ ;  $p > 0.05$ ). Both lipid biomarker indices show terrestrial-derived lipids associated with both low and high  $k$ -values, which is also true for marine-derived lipids. Thus,  $k$  is not directly related to terrestrial vs. marine sources but to the degree of alteration of OM and the general context of each depositional environment, which becomes more evident at a regional and local-scale context. For instance, deep-sea, well oxygenated sites exhibit marine-derived, yet low reactivity OM, which is associated with pre-ageing during settling, as well as post-depositional processes and mineral matrix association. In contrast, the high OM fluxes, anoxic waters and sediments of the Arabian Sea region are characterized by high reactivity marine-derived OM. In coastal and marginal sites, terrestrial-derived low reactivity OM is associated with increased distance from sources, as well as low sedimentation rates and oxic conditions. OM reactivity

variability is also captured by lipid biomarker indices indicative of OM alteration (Fig 1c – *n*-alkanoic acid CPI; Fig.1d – *n*-alkane CPI), especially in marginal settings (e.g. Northern European margin). Such relationships are independent of water depth, and instead are mainly dictated by distance of terrestrial sources and redox conditions.

Overall, OM sources and reactivity relationships are complex and lipid biomarkers are only robust at explaining *k*-variability in the broad context of each depositional environment. That emphasises the need of multidisciplinary studies to fully capture the main drivers of OM reactivity, which then help to disentangle processes and improve model parameterisation.



**Figure 1** Model-derived OM reactivity  $k$  at the sediment-water interface vs. lipid biomarker diagnostic ratios. (a) *n*-alkanoic acid TAR; (b) GDGT BIT index; (c) *n*-alkanoic acid CPI; (d) *n*-alkane CPI.

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