COMMENT AND RESPONSE FOR REFEREE 1

We thank the reviewer for carefully reading our work and taking time to comment on some of strengths and perceived weaknesses of the paper. Our responses are represented in the red text in reply to the reviewer comment in black.

(1) Model description It would be helpful if the authors can provide a short overview of model development relative to the previous and already published works/models. Current manuscript referred to these works, but what is exactly new is kind of obscure. Also, brief summary of the capacity/features of previous models from which the current model has been developed will be useful, including programming language, governing equations, and algorithms utilized for numerical solutions (finite difference/volume/element method for equation differencing and Newton iteration if adopted for solution seeking etc.). The current manuscript provides some of above information, but some information is still missing. And it is not 100% clear which part is new to the current model and which part is not new.

The goal of this study is (1) to better understand the impact of episodic events on biogeochemical processes following deposition, and (2) estimate relaxation timescale. In order, to accomplish this, we developed an early diagenetic model called FESDIA based on early OMEXDIA code achievements. The ability to explicitly simulate non-steady early diagenetic processes in systems subject to perturbation events such as massive flood or storm deposition is a novel contribution of FESDIA to early diagenetic models. The novelty introduced by FESDIA compared to OMEXDIA are:

- An explicit description of the anoxic diagenesis beyond the nitrogen cycle including (i) Iron and Sulfur dynamics, (ii) methane production and consumption. In comparison OMEXDIA has a single state variable (ODU: oxygen demand unit) to describe reduced species. This addition required substantial rewrite of the code. This inclusion of the other primary pathways involving Fe, S, CH_4, and potentially Mn species, makes the model suitable for studies in region where organic matter input trigger of anoxic diagenetic processes.
- possibility to include sediment perturbation events such abrupt deposition of sediment. This is a crucial addition that makes the model suited for its intended application.

In this paper, we only discuss part of the FESDIA model concern with implementation of a perturbation event as it relates to some biogeochemical indicators. The model is implemented in Fortran (for speed) and linked to R (for flexibility).

(2) Definition of relaxation time Eq. 22 describes the change rate of species concentration within sediment profile but does not necessarily define deviation from the pre-flood profile. Also, current manuscript lacks figures that directly compare the solute profile development relative to the pre-

event profile. I think Figs. 4-8 need to be improved so that profile development relative to the pre-event profile is more visually obvious.

This relaxation timescale calculation based on the disappearance of the perturbed signal (via successive profiles similarities) may differ from an approach in which the profile returns to a pre-defined "old profile". Because the exactness of pre-flood and post-flood profiles is difficult to quantify numerically (Wheatcroft, 1990), and since the return to the former is frequently driven by slow dynamics, the approach used here can provide a window of estimate for which a particular signal fades toward the background of a theoretically pre-perturbed signal.

The solute profile is emphasized relative to their post-flood evolution because we are interested not on the exact return to the preflood profile (which can be rather long-term or never be if the system moves to a new steady state). Instead, we aim to characterize the time taken for a quiescence of the perturbed signal following the event (a much more reliable/quantifiable entity).

(3) Description of model limitations and future development Relevant to the above point, but limitations of model should be discussed more. There are several simplifying assumptions in the model but its influences on e.g., model validation, comparison with observation and estimation for relaxation time are not discussed.

A new section to discuss the model limitation has been added to revised version.

For instance, the authors assume that burial rate/porosity does not change with the flooding, but it is not discussed whether this assumption is defendable or close to what we observe.

Indeed, some coastal sediment burial rates have been shown to vary seasonally (Soetaert et al., 1996; Boudreau, 1994). In the proximal zone of Rhône prodelta, about 75% of sediment deposition occurs generally during the flood (eg., 30 cm/d). Remaining 25% is distributed along the year with a low range daily constant rate (0.03 cm/d). As well, porosity vertical distribution is time independent i.e. only organic matter content and deposition height change in the new layer. In the case of the Rhône river outlet, porosity in the flood deposit will depend on the contributing zones in the watershed and nature of eroded material as well as on rainfall intensity and duration. Resulting porosity in the new layer is barely predictable and could range between 0.65 and 0.85 in the proximal zone of the prodelta (Grenz et al., 2003; Cathalot et al., 2010).

Model validation or comparison with the observed data is essentially based on solute profiles, which likely resulted from a good fit of TOC and may be achievable under different assumptions (those that allow changes of burial rate, porosity, bio-mixing and irrigation etc.). If this is the case, the relaxation time is likely quite different under different assumptions. While most of manuscript discussed how relaxation time is calculated under the specific assumption adopted for this study, it is not discussed how the relaxation time is affected by adopted assumptions.

Daily burial rate, porosity, bio-mixing and irrigation coefficients were extracted from the modelling work published in Pastor et al. (2011) and Ait-Ballagh et al. (2021) where only bio-mixing is an adjusted parameter. Other have been estimated from direct measurements and from macrofauna field observation. Adjustment of bioturbation coefficients for different sampling/modelled stations in the Rhône prodelta showed that modelling outputs were not clearly sensitive to these parameters (Ait-Ballagh et al. 2021).

--- SPECIFIC COMMENTS ---

In model description, it may be better if you say what programming language you are using earlier on (even in abstract).

The model is implemented in Fortran (for calculation speed) and linked to R (for flexibility).

L134. Three OM fractions? Thought the authors are using two.

This sentence containing this statement have been clarified in the revised paper by rephrasing:

"*As a result, the model assumed solid phase organic carbon with two reactive fractions with different reactivities and C/N ratios (Westrich and Berner, 1984; Soetaert et al., 1996). The degradation of OM involvesa labile fraction mineralizing faster than the slow decaying carbon. During the timescales considered here, the refractory organic matter class is not reactive. To compare with the observation, we consider an asymptotic OC constant (C_{ref}) for the inert fraction that scales the model calculated TOC output to the observation (Pastor et al., 2011) (see section 2.2.8).*"

L138. Froelich et al. (1979) rather than Froelich (1988) according to Bethke et al. (2011, AJS 311, 183)?

This has been corrected in the revised text.

L166. What does 'a coupled reaction formulation' mean?

The sentence was modified to read as: "*The law of mass action mathematically describes these reactions, with their kinetic rate term influencing the reaction.*"

Eqs. 6, 7. It does not make any sense to use/define 'maximum rates' when one is not using Monod or Michaelis-Menten type of equation.

"maximum rates" and replaced with "rates"

Eq. 7. What is the definition of rH2Soxid?

Text formatting issue with Microsoft when we translated our text from latex to Word. We fixed that in the updated text.

Eqs. 8, 9. What is the assumption behind the formulation of these equations? For instance, how do you obtain Eq. 9 for aqueous NH4+ with accounting for adsorbed NH4+? Can you track NH4+ adsorbed onto solid species along with OM and other solids or do you have to simplify that adsorption is depth-independent and/or time-independent? This can be important if solid materials with unoccupied exchange sites are flooded to sediment depocenter in a short period of time. If such case is possible, one would expect a large sink of NH4+ to the exchange sites? Related to this, do you model PO4 adsorption onto Fe hydroxides or you do not have to do this?

In regard to this comment, the sentence was modified to:

"*With special consideration of ammonium adsorption to sediment particles, the governing equation is given by:*

$$\frac{\partial \phi C}{\partial t} = \frac{-\partial}{\partial z}\left[\frac{-\phi \times D_{sed}}{(1+k_{ads})} \times \frac{\partial C}{\partial z} + w_{\infty} \times \phi_{\infty} \times C\right] + \sum \frac{\phi \times REAC}{(1+k_{ads})} (9)$$

where we assumed that the immobilization of NH_4^+ *is in instantaneous, local equilibrium (i.e. any changes caused by the slow* NH_4^+ *removal process results in an immediate adjustment of the* NH_4^+ *equilibrium; so, can be modelled with a simple chemical species) and* k_{ads} *is the adsorption coefficient. The inclusion of this formulation for the diffusion and reaction term has the effect of slowing down ammonium migration in sediment. Derivation of this formulation is given in Berner, 1980; Soetaert and Herman, 2009.* "

It is possible to calculated and tracked the amount of absorbed NH_4 as a modeled diagnostic which is part of the output of model.

The code has possibility to incorporate this Fe-P adsorption but that is not the scope of the paper which is focus on the main early diagenetic pathways affected by the flood deposition.

L205. Does porosity 'decay'?

The "porosity decay coefficient with depth" has been replaced with "depth constant for decrease of porosity with depth" (see Rabouille and Gaillard, 1991a)"

L209. According to Eq. 10, the authors seem to assume intraphase biodiffusion (Meysman et al., 2005, GCA 69, 3601). The statement here mentioning an interphase biodiffusion is inconsistent with Eq. 10.

It has been corrected with "*In this work, this bioturbation flux is assumed to be intraphase, with porosity* $\phi(z)$ *remaining constant over time*."

Eq. 14. Irrigation term is not found in the governing equation. Is it included as a reaction term?

Irrigation is included in the REAC term shown in Eq 10 and to that effect a sentence has been added in L207 reading – "*This term includes both biological and chemical reaction within the sediment column as well as non-local bio-irrigation transport term (see next section).*"

Section 2.2.5 & Section 2.2.6. More details are desirable as adding grid for implementing a deposition event must be an important addition to the previous modeling framework. For instance, how you define Zpert, e.g., number of grids and their geometry etc. Some examples, not only schematics may also be useful.

Definition of the Zpert (as the depth of deposition of flood derived sediment layer) were briefly given in the main text. This scalar value is derived from the specified number of grid layer to perturb (Npert). The depth integration of the output from an irregular grid-based generation routine of the ReacTran R packages (which implements many grid types used in early diagenesis modelling) for a number of grid points (Npert) results to Zpert. An example can be found in Figure S1 of the supplement text.

This explanation is added in the section 2.2.5 in the revised text.

L276. 'maximum in the spring and minimum in fall and winter'. This line does not make sense to me.

This sentence has been rephrased with: "*In the latter case, this carbon flux varies over the annual carbon flux* $(\overline{flux}_{org})$ *in the region in question.*"

Eq. 21. What is the units of TOC? Also, how do you derive Eq. 21? Is this simply analytical solution of the governing equation? In any case, it would be helpful if the authors can provide the procedure to obtain Eq. 21 somewhere.

The unit is percent dry wet. A preamble like this has been added to the revised text:

"*TOC (in % dw) is estimated from both carbon fractions* (C_{org}^{fast} *and* C_{org}^{slow}) *assuming a sediment density* (ρ) *of 2.5* $g\ cm^{-3}$ *and conversion from the model unit for detrital carbon fraction of* $mmol\ m^{-3}$ *to unit percent mass. The TOC is not analytical solution to the general governing equation. It is diagnosed from both the fast and slow decaying detritus component of the OM which are calculated from the governing equation that is solved numerically*"

A background refractory concentration Cref is added to Eq 21 to account for the fraction which are not calculated within timescale simulated by the model.

L321. 'were utilizes the R programming language'. Correct English?

This has been corrected to: "*The R programming language is used in the preprocessing routine for model grid generation (Soetaert and Meysman, 2012), porewater chemistry parameter (Soetaert et al., 2010a), steady state calculation (Soetaert, 2014), and time integration (Soetaert et al., 2010b)*".

Section 2.2.9. It would be helpful if the authors can make a short description of what part of "deSolve package" they used, not only directing the reader to the R-forge webpage. More specifically, how the authors numerically solve the governing equations, apart from "method-of-lines" methods? Use of any finite difference/volume/element method?

This is also a featured requested by the reviewer 2. We added in section 2.2.9 a new paragraph:

"*Because the procedure is based on OMEXDIA, complete details of the derivation can be found in that paper and referenced therein (Soetaert et al., 1996). Here we recap the mathematical formulation of the method-of-lines (MOL) algorithm used by FESDIA. Direct differencing of (8) - (10) results to:*

$$\begin{aligned}\frac{\partial C_i}{\partial t} &= \frac{\Phi_{i,i+1} D_{\Phi_{i,i+1}} (C_{i+1} - C_i)}{\Phi_i \Delta x_{i,i+1} \Delta x_i} - \\ & w_\infty \Phi_\infty \frac{\alpha_{i,i+1} C_i + \left(1 - \alpha_{i,i+1}\right) C_{i+1}}{\Phi_i \Delta x_i} - \\ & \frac{\Phi_{i-1,i} D_{\Phi_{i-1,i}} (C_i - C_{i-1})}{\Phi_i \Delta x_{i-1,i} \Delta x_i} + \\ & w_\infty \Phi_\infty \frac{\alpha_{i-1,i} C_{i-1} + \left(1 - \alpha_{i-1,i}\right) C_i}{\Phi_i \Delta x_i} \qquad (22)\end{aligned}$$

for a generic tracer C with a phase properties index Φ *and* D_Φ *denoting porosity and dispersive mixing term respectively for solid or liquid. This equation is calculated such that the variables and parameters are defined both at the centre of each layer* x_i *and at the interface between layers* $(x_{i,i+1}, x_{i,i+1})$*. The position at the centre of the grid is then given as* $x_i = \frac{x_{i-1,i} + x_{i+1,i}}{2}$*.* Δx_i *represents the thickness of the i-layer and* $\Delta x_{i,i+1}$ *is the distance between two consecutive grid layers. A Fiadeiro scheme (Fiadeiro and Veronis, 1977) based on the model's Peclet number (a dimensionless ratio expressing the relative importance of advective over dispersive processes) is used to provide a weighted difference of the transport terms in order to reduce numerical dispersion.*

How is the time-integration of governing equations made (time-implicitly or -explicitly)?

"*…Equations (8)-(10) implemented as Eq. (22) is integrated in time using an implicit solver, called lsodes, that is part of the ODEPACK solvers (Hindmarsh, 1983). This solver uses a backward differentiation method (BDF); it has an adaptive time step, and is designed for solving systems of ordinary differential equations where the Jacobian matrix has an arbitrary sparse structure.*"

L331. What is a "slow" stationary state?

The word and its surrounding sentence have been rewritten as follows:

"*We estimate the relaxation timescale by first calculating the absolute difference* ($\varphi(t)$) *between successive model output after the event, assuming that a slowly evolving state will eventually converge to the pre-perturbed state as time after the disturbance approaches infinity.*"

Eq. 22. Not quite sure this is a legit mathematical expression. phi(tau) < threshold is what I thought is consistent with what the authors described.

It was intended to imply that the relaxation estimate is based on calculating a threshold given by the median of the $\varphi(t)$. Eqn 22 have been updated to explicitly specify the subscript and avoid any misinterpretation:

$$\begin{aligned} \varphi(t) &= \frac{1}{N}\sum_{i=1}^{N}\|X_{t+1}^{i} - X_{t}^{i}\| \\ &\text{In the limit of time (t):} \qquad (23) \\ \tau(t) &\Rightarrow \varphi(t) \leq \varphi(t)_{threshold} \\ &\text{where } \varphi(t)_{threshold} = \overline{\varphi(t)} \approx \text{ seasonal background} \end{aligned}$$

L334. "threshold (i.e given by the median over the entire time duration)." Do you mean that the run is finished when difference becomes less than the median value throughout the simulation and then tau is defined as the model time required for this?

Yes, Using the Eqn 22, the calculation terminates at the infimum (greater lower bound) time point when the curve encounters the threshold. That terminal time point is thus the $\tau(t)$ as given by the equation.

L340. I probably do not fully understand the ensemble of simulations here to estimate the uncertainty in tau. What parameter do you randomly re-sampled exactly? Median of the reference run through time? If so, the runs for determining the uncertainty in tau is conducted until (randomly-chosen) prescribed median is crossed? But this does not necessitate re-running of the model as the boundary conditions are not changed?

This section has been rewritten to and the pertinent parameter being varied is highlighted in bold:

"*In this case, we employ a modified bootstrapping technique to estimate the uncertainty in the relaxation timescale by resampling on the* ***cutoff point*** *introduced in Eq. 23 (i.e. median,* $\overline{\varphi(t)}$ *of a given reference simulation). This calculation takes advantage of the fact that the timeseries will be dominated by the* ***slowly varying seasonal cycle over a long time period away from the point of perturbation****, with the influence of the perturbation fading to the background.* ***The variation of this reference timeseries over time reflects the uncertainty in this median threshold point.*** *This variance, along with the reference cutoff value, can be used to generate n random perturbations varying about the normative threshold value. We can proceed to create a histogram of the replicate threshold****(s)*** *distribution. The histogram of* *this distribution is depicted schematically in the left margin of Fig. 3. The relaxation time in each realization of the threshold is calculated ($\hat{\tau}_i$). The median absolute deviation from this ensemble of relaxation times is then used to calculate the level of uncertainty in the statistics of interest (timescale of relaxation - ($\hat{\tau}$)). Figure 3 depicts this concept schematically.*

It should be noted that this method eliminates the need to rerun the deterministic model for each iteration, reducing the computational burden of this technique."

L383. "a thickness scale of 1 cm to 30 cm in 5 cm increments". This line does not make sense to me. What exactly did you use for thickness in sensitivity analysis?

This sentence has been changed to include the full value in the revised text.: "*...a thickness variation ranging from 1cm to 30 cm. A 5 cm increment was used for the sensitivity analysis*"

Section 2.2.11.2. If the tested values are not too many, it would be better to list exact values you used for sensitivity analysis.

The revised text includes the tested values utilized for sensitivity analysis.

L507. Please specify what "RiOmar" stands for.

It is stand for river-dominated ocean margins. It is mentioned in the Introduction and discussion.

L621. above → below?

The preceding sentence have been changed to: *"...resulting in a shorter recovery time for the porewater profile to reorganize within the SML*"

This thereby explicitly specify where the shorter recovery time occurs.

--- TECHNICAL COMMENTS ---

Table 1. What does unequal mark on $Fe(OH)_3$ mean? Is this typo? At least notion should be consistent with that in main text.

It is a typo. It has been deleted in the revised paper.

L139. Eq. 3 → Eq. 2?

This has been changed in revised manuscript.

L165. Typo in the second line of Eq. 5.

The typo has been corrected in revised manuscript.

L168. Right parenthesis in the last line of Eq. 6 is missing.

This have been fixed.

L212. Where → where?

This has been corrected in the revised text.

L214. specify à specified?

“Specify” has been replaced with “specified” in the revised manuscript.

L230. i.e à i.e.?

This is fixed in the revised manuscript.

L232. occur → occurs?

The word is corrected in the revised text.

L246. Figure. 2 à Fig. 2 or Figure 2?

This Figure labelling in the text has been made consistent.

L318. “method-on-lines” à “method-of-lines”?

Indeed, this have been corrected to “method-of-lines” in the revised manuscript.

L409. dissolved DIC à DIC?

This has been fixed and corrected.

L415. as thus → as follows?

This has been corrected in the revised manuscript.

L418. Table. 3 à Table 3

A “.” have added to Table in the revised manuscript.

L433. “Solid” should not be superscript

The superscript has been be removed in the revised text.

L434. “Solid” should not be superscript

Same as above.

L535. Improve → improved

It has been corrected accordingly in the revised manuscript.

L581. introduction à introduction of or introducing

It has been corrected accordingly.

L660. Thickness → thickness

Corrected.

L694. design → designed

Corrected.

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