Reply to Anon. Rev. 1 (Wang et al.)

Reviewer: The authors aim at interpreting in-situ pump data from the DYFAMED station in the Mediterranean Sea. For this purpose they propose a model including aggregation and disaggregation of particles of two size classes, respiration, degradation and sinking. Data and model values are considered at 7 depth levels. The model parameters are estimated in two steps by an Bayesian approach.

At the time of sampling (May 2005) the particle numbers in the water column are very low and thus I doubt that aggregation would have played a significant role in particle dynamics. Aggregation of particles is an essentially non-linear process (because particles have to meet in order to aggregate) and thus in my opinion a linear term for the description of aggregation is highly inappropriate. Further, the authors make a steady-state assumption without justification or further discussion. Application of the estimated ('optimal') parameters in the various model equations yields no convincing support for the steady-state assumption. The whole text seems to be largely driven by methodological aspects (Bayesian approach) and neglects a discussion of the data and the estimated model parameters (are the estimates plausible/do they make sense?). Given the deficits in the model formulation ('linear aggregation', 'steady-state') and the question whether the data used contain valid information about the processes included in the model, I could not see what a reader can learn from this investigation and thus I do not recommend publication in Biogeosciences.

Our reply: As we stated on p. 3 lines 8-13, the objectives of the study were 1) to introduce a new method for using pump collected chloropigment tracers to infer particle dynamics rate constants, and 2) to compare to previous studies at the same site, and thus, to infer if sampling methods or tracers themselves have more influence on particle aggregation and disaggregation rate constant estimates. Therefore, "The whole text seems to be largely driven by methodological aspects" is not a deficit; developing this method is what the study aimed to do. From our study, readers can learn 1) a new approach to determining rate constants, and 2) a proof that if the same tracers are used in the model, sampling methods have little influence on rate constant estimates.

The reviewer does not believe that the "linear aggregation" and "steady state" assumptions are valid. First, we argue that linear aggregation is not without merit, especially when aggregation is between the same kinds of particles. As the reviewer stated in his/her comments, "particles have to meet in order to aggregate". Therefore aggregation has to be related in some way to the concentration of particles, no matter whether the particle concentration is high or low. By assuming a first-order reaction, we have

$$\text{aggregation} = \alpha[P],$$

where α is an aggregation rate constant, and [P] is the particle concentration. Higher particle concentration results in a higher chance to collide, and thus more aggregation. In most empirical studies, including with the current data, we have no way to build and test a complicated model such as that in Jackson (1990). Also, unlike in Jackson (1990), we are dealing with particles below the euphotic depth, so do not consider algal division and other euphotic zone processes. Many previous models have assumed first-order reaction kinetics and show reasonable results (Murnane et al., 1990; Clegg and Whitfield, 1991; Marchal and Lam, 2012; Lerner et al., 2016, 2017).

Second, we assume “steady state” since we are using only one concentration profile. We do not have enough information to build a non-steady-state model. To achieve our second goal, to compare the influence of sampling techniques and tracers on parameter estimations, we have to make the same assumptions as we did in previous studies (Wang et al., 2016; 2017).

Reviewer: Further remarks:
Abstract: ”The estimated aggregation and disaggregation rate constants were 7.65 + 3.35 – 2.33 (0.13 yr) and 106.09 + 39.13 – 28.59 yr^{-1} (0.01 yr), respectively, which indicates that particle aggregation and disaggregation were extensive at the studied depths (125-750 m) in May after the spring bloom had ended and flux was low.” Disaggregation is faster (lower time constant: 0.01 yr) than aggregation (time constant 0.13 yr) and thus one would expect less and less aggregates.

Our reply: We do not understand the reasoning here. Lower turnover times (0.01 year for disaggregation, and 0.13 years for aggregation) indicate higher reactions. We were not comparing absolute amounts of aggregation vs disaggregation here. To determine whether more particles were disaggregating than aggregating, we would have to determine actual rates, not rate constants. That was not our purpose.

In addition, another study at the same site at the same sampling time indicates high aggregation and disaggregation using totally different empirical methods (Abramson et al., 2010).

Reviewer: p. 1 L19 Stoke’s law ⇒ Stokes’ law (named after George Gabriel Stokes)

Our reply: We will fix this. Thank you.

Reviewer: p. 2 L5-7 ”3) dissolved thorium activity is orders of magnitude higher than particulate activity, thus in models, dissolved thorium influences the adsorption-desorption balance more than particulate thorium does (Wang et al., 2016).”
Not clear to me: ’What is the disadvantage here?’ A problem for models only or also for the real world?

Our reply: This is a model problem when Th is used, as we stated in the paper.

Reviewer: p. 2 L16 ”Wang et al. (2017) compared rate constants ...” Rate constants: for which process(es)?

Our reply: Aggregation and disaggregation rate constants. We made it clear in the paper.

Reviewer: p. 2 L18-20 ”There were clear differences: aggregation and disaggregation rate constants estimated from chloropigments were orders of magnitude higher than those from thorium tracers. These results were thought to be due to the differences in the way thorium and chlorophyll exist on and within particles.”
Not clear, what authors like to tell us.

Our reply: We meant that thorium and chloropigments have different properties, and trace different particle processes. We discussed this more in our previous paper (Wang et al. 2017) and should have repeated it here (and will do so). In that paper, we showed that particulate pigments cycled differently than particulate thorium. Thorium and other surface-active elements would behave very differently from chloropigments as they adsorb and desorb from the surface of the particle. Chloropigments are more likely to be an integral part of the particle.

Reviewer: p. 2 L26-29 this should be dropped: ”... since traditionally it is thought that aggregation is the process of small particles colliding with other particles and becoming larger, and disaggregation is the process of larger particles breaking up into smaller particles. Whereas, particles in fast sinking categories are a combination of large and small particles, because some small particles have high sinking speed, and vice versa …”

Our reply: Why would you like this dropped?

Reviewer: p. 3 L12-13 ”These inter-comparisons enabled us to investigate whether sampling techniques or the tracer itself imposed a stronger constraint on modeled rate constants.” ???

Our reply: We assume you mean that he sentence was unclear? How about: “The inter-comparisons we describe here allowed us to investigate whether sampling techniques (pumps vs. traps) or the tracer itself (chloropigments vs. Th) imposed the stronger constraint on modeled rate constants.”

Reviewer: p. 3 L17 (4320’N, 740’E) ⇒ (43° 20’ N, 7° 40’ E)

Our reply: We will fix this.

Reviewer: p. 4 L16 ”If we discrete ...” ⇒ ”If we divide …”

Our reply: We will fix this.

Reviewer: p. 4 L27-28 ”We assume first-order reaction kinetics for aggregation and disaggregation, which is a gross simplification since the real reaction kinetics is not known.”
If the real reaction kinetics is not known, then it is not known whether first-order reaction kinetics is a gross simplification.
You are correct. We will change this to “which is probably a gross simplification since the real reaction kinetics is not known.”

The kinetics of aggregation of biogenic marine particles has been described by Jackson (1990) and many others: it is essentially non-linear (because particle have to ’meet each other’ in order to aggregate). The kinetics depends on size, sinking speed, stickiness of particles (for details compare, for example, Jackson, 1990).

Our reply: In the model we use, aggregation is a process of smaller particles aggregating to form larger particles. We assume that small particles are single, non-sinking particles, and have the same stickiness. As discussed above, with the data we have, we cannot build a model like that in Jackson (1990).

Also, the first-order kinetic model is reasonable when aggregation is between the same kind of particles. By assuming first reaction kinetics, we have

$$\text{aggregation} = \alpha[\text{P}]$$

Thus, high particle concentration indicates high chance of particle collision, thus high particle aggregation. Previous model studies assuming first reaction kinetics also come to reasonable results (Murnane et al., 1990; Clegg and Whitfield, 1991; Marchal and Lam, 2012; Lerner et al., 2016, 2017).

Reviewer: p. 5 L1 ”We thus use the same mathematical method and conceptual model found in other published studies ...”
Used methods (including first-order reaction kinetics for aggregation) have to be justified based on theories (first principles) and/or observations; reference to ’other published studies’ is not enough.

Our reply: With measurements at only one depth profile. We have no way to build a non-steady-state model. And for the purpose of model comparison, similar steady state assumptions as in previous studies (Wang et al 2016; 2017) were made. We use the same justifications as in those papers, and do not presume to repeat them all.

Reviewer: p. 5-6 ”Here we assume the system is at steady state, just as has been done in Wang et al. (2016, 2017) and almost all similar work, e.g.(Murnane et al., 1990; Clegg

and Whitfield, 1991; Marchal and Lam, 2012; Lerner et al., 2016, 2017), to make the results comparable."
The steady state assumption is not sufficiently justified (again: reference to 'similar work' is not enough).

Our reply: See above reply.

Reviewer: p. 7 L6 "hyperv paramter" ⇒ "hyper parameter"

Our reply: We will fix this.

Reviewer: p. 7 L7-8 "Typically, we obtain the optimal parameters in less than 50 iterations." 'Iterations' is mentioned here for the first time. What is iterated?

Our reply: The model iteratively searches for optimal parameters.

Reviewer: Table 2: typo: $\beta \Rightarrow \beta_1$

Our reply: We will fix this.

Reviewer: **References**

[1] Jackson, G.A. A model of the formation of marine algal flocs by physical coagulation processes. Deep-Sea Res., 37(8):1197–1211, 1990.

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