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Discussions

Interactive comment on "The potential for material processing in hydrological systems – a novel classification approach" *by* C. E. Oldham et al.

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The authors here address important challenges in attempting to unravel complex interactions between water flow, biogeochemical processes, and ecosystem structure in aquatic systems. These dynamics are notoriously complex, and there is a pressing need to understand dominant controls on whole-system biogeochemical and ecological rates and distributions. I think the attempt to develop general classification schemes is worthwhile. However, as much as I appreciate the attempt, I think the proposed quantitative metrics are not well defined and therefore not useful for classification.

I do not fully understand the "effectiveness" concept for ecological connectivity. How does this differ from the actual transport of reactive material, e.g., fluxes or mass loadings? Effectiveness here seems vague, while fluxes or mass loadings can be precisely defined. The first example provided by the authors, Fe(III) delivery, illustrates this issue well. The authors state (correctly) that biogeochemical processes will tend to immobilize Fe(III), and thus that its transport will be less than that of conservative material. I don't understand what about this would make transport effective or not effective. Effective relative to what…Fe(III) transport in other systems? Physiological needs for iron? This lacks context and precise definition, and so I find it vague. It seems preferable to use more precise and quantitative descriptions here (mass immobilized vs. transported, % removal, etc.) rather than superimpose a more vague concept of effectiveness on it. It is easier to understand the effectiveness concept when applied to populations and communities of organisms. But here also I think it would be greatly preferable to employ specific quantitative metrics, such as spatial statistics for patterns, heterogeneity, etc., which provide more primary measures and could then potentially be mapped onto standing stocks and fluxes between reservoirs to better understand the extent of connectivity provided by hydrologic transport.

I think part of the issue is that the prior literature is also vague. In the abstract (line 10) and the introduction (p. 10492, line 23-24), the authors refer to the prior definition of Pringle in terms of the "elements of the hydrologic cycle." I am not certain what this means…is it bodies or reservoirs of water, connectivity provided by flow, or something else? In either case, how exactly are the boundaries and magnitudes of such elements defined? And are these really well defined and constant over time? It would be preferable to provide a more specific description of the physical system here so that system boundaries and fluxes can be more precisely understood. If the authors attempt this strictly, I think they will find that a major part of the problem is trying to identify the system boundaries in the first place.

The issue of system definition carries through to the quantitative analysis. Equation 2, while providing a quantitative framework, isn't really very useful unless the system is specifically defined. Certainly the authors know that eqn. 2 formally doesn't have any solution without both boundary and initial conditions – in that case, how can it be used to classify systems when the systems aren't specified? The authors acknowledge these issues briefly at the top of p. 10494, but I think that they do not fully follow through on the implications. The effective diffusion coefficient in eqn. 2 and eqn. 4 is highly scale dependent. Therefore this parameter itself depends on how the system is specified and whether system boundaries are physically strict (e.g., fully impermeable) or effective (e.g., low connectivity). Very few hydrological system boundaries are strict at the scales being considered here, so effective diffusion coefficients are empirical properties dependent on the assumed discretization of the system. Therefore Deff cannot easily used for classification.

One key challenge with Equation 2 is that the diffusion equation is based on the assumption of Brownian motion, particularly that motion is continuous in space and time, but many real cases are effectively discontinuous/non-local (i.e., alternating between very slow and very fast, so effectively episodic and non-local in space). Such discontinuities are not well represented by effective diffusion coefficients. There is a substantial body of literature available on this in physics that has now been extended to geophysics, hydrological transport, and reactive transport.

The reaction rate constant k in equation 7 and specific consumption rate q in equation 8 are subject to similar considerations but are even more problematic. As the authors note, biogeochemical and ecological processes cause the distribution of reactive chemicals and organisms to differ from conservative solutes. Patterns and rates of transformation at the system scale therefore depend on both transport and true local (e.g., organismal scale) chemical and biological process rates. Thus k and q are emergent properties that contain information on hydrological connectivity as well as biogeochemistry. Fully well-mixed systems will show a maximal rate, but all real systems, and especially the poorly mixed systems considered here will show important transport limitations in the effective chemical and biological rate parameters. So again they are not easily used for classification.

Based on the above, I find that the classification system in section 2 isn't very useful. Certainly it provides some effective measures, but only a very bare, empirical view of the relative behavior of semi-arbitrarily defined effective hydrological reservoirs at particular times that measurements are made. I think this would likely not be useful in practice and could be misleading, as different discretization of the same system or repeated application of the same measures at different times would yield very different results. In other words, the resulting classifications would not be general or reproducible because they depend strongly on the details of internal dynamic that are not captured with this parameter set. In my opinion, more basic measures of spatial patterns and mixing rates (e.g., spatial segregation and the factors that tend to favor or disfavor such segregation) are more likely to be useful for system classification. I think it would be better to consider more directly the extent of heterogeneity in primary variables of interest (distributions of chemical and biological species), which are still descriptive but at least provide a primary measure of patterns instead of an interpreted one (i.e., they aren't dependent on being fit into the assumptions of a diffusion model).

Some of the direct conclusions of the analysis are similarly misleading. The abstract states "Within each regime we use a new non-dimensional number, NE, to compare exposure timescales with reactions timescales. NE is reaction-specific and allows the estimation of relevant spatial scales over which the reactions of interest are taking place." This is not true, as relevant spatial scales in heterogeneous systems cannot be identified only from characteristic times. Similarly the statement of conclusions that "NE can be used to determine the physical constraints on a system required to ensure, for example, that 90% of a biogeochemical is removed" is not guaranteed to be true for heterogeneous systems and is in fact unlikely to be true for the highly heterogeneous systems specifically considered here. Internal distributions of transport and reaction, and the covariance between reaction and transport parameters in both space and time, are likely to dominate overall export.

The examples provided in Section 3.2 - 3.3 are all interesting to me and worth considering. However, they do not prove the utility of the authors' classification framework. These examples are explicitly included by the authors in the ms. as illustrations only, and do not provide enough detail to really explore the limitations of the material presented in Section 2. I do think that this type of approach can be useful in getting an overall sense of biogeochemical dynamics at large scales, at which detailed observations of chemical/organisms distributions are generally not available and so only order-of-magnitude analysis is possible. But this has already been done for many cases (e.g., Harvey and Fuller, 1998, "Effect of enhanced manganese oxidation in the hyporheic zone on basin-scale geochemical mass balance" is a nice example). It doesn't make sense to me to try to formalize this sort of approach as a general classification scheme beyond what has been done classically. It is just not fundamental enough to provide detailed understanding of the basic process controls on the important biogeochemical and ecological dynamics, and it does not incorporate recent theoretical advances in stochastic transport theory, reactive transport in heterogeneous systems, spatial segregation of reactions in transport-limited systems.

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