Hydrol. Earth Syst. Sci. Discuss., 9, C5631–C5638, 2012
www.hydrol-earth-syst-sci-discuss.net/9/C5631/2012/

Hydrology and Earth System Sciences 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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Received and published: 3 December 2012

Reply to Comments from Reviewer 1

Thank you for your comments on this manuscript. We believe that many of the comments result from a lack of common language and conceptual understanding, and it is exactly this confusion (which we have identified across a range of literature) that we aimed to unravel with this manuscript. We explicitly aimed to keep the manuscript conceptual and at a higher level, to present a common language, definitions and concepts that can then be applied to many different contexts.

In contrast to our aims, this reviewer seems to have a very specific application in mind

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– transport and reaction through heterogeneous porous media. Our framework aims to be more broadly applicable. Some of the language we use will therefore be different to that used in previous studies in specific contexts. We have aimed for, and at times had to develop, clear definitions that are applicable across multiple applications and contexts. The term "reactive transport" has been used previously to describe solutes that react; our framework can be applied more broadly than reactive solutes, and so we have not used this term. The word "effectiveness" is a more general term that encompasses many contexts, for example particle settling, seed viability. The expression can be used for both dissolved and non-dissolved material, reactive and non-reactive material. Also, the term "fluxes" is only relevant when systems are hydraulically or hydrologically connected. Our framework aims to encompass both connected and intermittently disconnected systems, and so again we prefer the use of the term "transfer effectiveness".

We have defined "effectiveness" within the manuscript and have given examples in our Case Studies of how we apply this definition. The concept is clear and unambiguous: transport is "effective" when the exposure time within a defined system is too short for substantial biological or chemical processing. The value of "substantial" will vary from context to context and is set by the researcher.

The term "effectiveness" is an attempt to integrate all removal processes, rather than focusing on specific processes. The paper aims to integrate process understanding into a broader framework. Researchers can translate "effectiveness" into their specific process of interest. This does not detract from the framework. In our Case Study we have indeed used the effectiveness concept to determine % removal, however we note that the effectiveness concept is applicable to a far greater range of contexts than given in our Case Studies. We therefore believe that the use of the broader term is warranted.

It is certainly useful and desirable to undertake mapping and flux estimates, when such detailed data is available. Unfortunately the majority of contexts do not have access to such data. Our conceptual framework utilizes a very different approach. It asks whether a system is able to effectively transport material. We also note that spatial patterns, and heterogeneity are actually the result of the balance between transport and material processing. They are not primary indicators of processes. As an example, we cannot extract nutrient turnover rates from nutrient concentration profiles. The profiles are the net effect of turnover rates and transport.

We strongly agree that there has been definition ambiguity in past literature around the concept of connectivity; it is one of the reasons we are attempting to establish common terminology and concepts. Different people have used different terminology in the past creating a lot of confusion. This is an attempt to consolidate terminology.

The elements of the hydrological cycle were defined on P10491, L6-7.

We have attempted to unravel the definitions of connectivity in the manuscript. The most frequently used definition is in terms of water flow i.e. either hydrological or hydraulic connectivity. However we define connectivity on the basis of effective material transfer and it is this definition that requires the use of the NE approach.

We never assume boundaries are constant with time, we use a control volume approach. Control volumes need to be carefully defined for every context and can change with time. This fact does not detract from the usefulness of the control volume approach and is one is the strengths of the NE framework. It is a tool that forces scientists to very carefully delineate boundaries, both in space and time.

Boundaries and initial conditions are indeed required as one implements the framework. A good way to understand this is by taking an example from the application of the Reynolds number, Re, another non-dimensional number used in environmental flow research. The Reynolds number is the balance between inertial and viscous forces. As you try to define the Re that is specifically relevant to your context or process, you must carefully define your system, its relevant length scales, and then quantify velocities. These velocities may be estimated via order of magnitude estimates, via experimental data or via modeling. The methods used to quantify velocities could be very simple or highly sophisticated. The exploration of space scales and the dependency of velocities on a range of other factors, does not detract from the usefulness of estimating Re. Rather Re provides a framework for conceptualizing a problem and for updating our understanding as results from improved models or experiments become available. Establishing Re for a given context provides some degree of generalization across systems. We envision a similar utility of NE.

We agree with the comment that "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". As stated above however, we can be as broad-scale, small-scale, simple or complex as we wish, depending on our knowledge of our system and the data available. This is the case for whenever we use non-dimensional numbers but this does not detract of the usefulness of non-dimensional numbers.

We agree with the comment 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" and we have discussed this in the manuscript. Within heterogeneous porous media that are hydrologically connected, there will be a distribution of exposure times that create a distribution of NE. The statistical description of the exposure times can be as simple or as complex as required by the researcher. When the duration of the discontinuities increases to the same order of magnitude as material processing timescales, then the system becomes defined by the timescales of disconnection (i.e. it moves into Regime 3 of our framework). This example highlights the beauty of the NE framework: there is a smooth transition between regimes, with common concepts and definitions used across both connected and disconnected regimes.

We disagree with the comment that "k and q are emergent properties that contain in-

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formation 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."

This confusion is exactly the reasons why we recommend the use non-dimensional numbers, specifically NE. They allow us to quantify when a system is transport limited and when not. Whether a system is transport limited or not depends on the balance between exposure and processing timescales. And this will vary depending on the process you are interested in. A system is not a priori transport limited just due to the transport processes, as implied by the Reviewer's comment. This is exactly the thinking we are hoping to pick apart. Concepts of "transport limitation" are typically constrained because a specific reaction is being studied. The statement of limitation must be only related to that reaction, it is not a system-wide limitation.

Also, we believe that there is some confusion here about chemical kinetics. It is actually the rate dB/dt that is emergent as it depends on the supply of B (in eq. 7). Hence it depends on transport, which is exactly what we present in the manuscript. The parameters k and q are rate constants that are characteristic for a certain (bio)geochemical reaction at the molecular scale. Supply limitations are certainly not incorporated into the rate parameters. This common misunderstanding highlights the need for establishing a framework as proposed in our paper.

We fully agree with the comments that the framework "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".

But we believe they provide compelling reasons to use the NE framework. The ex-

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ample of Re is useful again here. There is not a single system Re, but rather a Re is dependent on the scale of interest. For example when we are studying advective flows through a vegetation canopy, we can define multiple Re. This highlights the relative nature of the flows and how scale affects our conceptualization of the important forces. Understanding and investigating the impact of scale on Re does not reduce the usefulness of Re. Indeed, part of its usefulness is that it forces us to define our system of interest, and its scales, very carefully. Similarly, the NE framework forces us to characterize a system in terms of its transport and time dependent ability to process material.

Despite what the reviewer states, we do not assume a diffusion model as the basis for our NE framework. As indicated in our previous comment, the spatial patterns within a system are not primary indicators, rather they are emergent quantities that result as a balance between exposure and processing timescales, and will vary with scale. The framework we have presented attempts to understand what might be causing these patterns. Of course we need good descriptions of the resulting patterns. Another useful example is taken from fluid mechanics. The use of Re in many applications has not prevented more and more sophisticated analysis of turbulence and the patterns caused by turbulence. The use of Re and the quantifying turbulent characteristics are quite different analyses. Statistical analysis of turbulence is used to understand the energetics of turbulence that is ultimately used as input to a better understanding of inertial forces. There will also be a whole area of experimentation investigating the small-scale effects of turbulence on e.g. phytoplankton nutrient uptake. But this forces us to define relevant Re for the scale of interest. So the understanding that the environment is very complex, and attempting to quantify that complexity, does not detract from the usefulness of non-dimensional numbers.

We also note that in parallel with turbulent eddies, the distribution or patterns of chemical species observed at a landscape scale, are likely the result of transport-reaction dynamics occurring at smaller scales (see e.g. Frei et al., 2012).

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We are not exactly sure what is meant by the comment "relevant spatial scales in heterogeneous systems cannot be identified only from characteristic times". But we think that the key point of interest is “of relevance”. We specify that the spatial scale of relevance for the calculation of NE is a) either physically constrained, or b) determined by timescales. Certainly, if one is interested in heterogeneity per se, then the spatial scales are not necessarily determined by any timescales. But we are specifically interested in the balance between processing and exposure. So the relevant exposure spatial scales are determined by the timescales of the processes.

We have presented a framework that can be used to establish hypotheses about time and space scales. And we agree that there is unlikely to be a single NE that represents a system. The “internal distributions” of transport and reaction will produce a distribution of NE, for a process of interest. We fully agree that understanding the distribution of both transport and processing parameters is critical for understanding the range of NE. However the usefulness of the NE concept is exactly that it can be used for any level of understanding (whether at conceptual level, or at basic data level, or at very detailed data level).

We also believe that the statement “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” is more a hypothesis and still needs to be tested. It is perhaps not appropriate as a statement of fact in a review.

We agree that Harvey and Fuller (1998) is a very interesting paper that calculates a non-dimensional number from an effective reaction time scale (which is indeed emerging). In our case we are using chemical rate constants, so our approach is different.

Finally we stress again that the NE framework is not restricted to use in transport-limited systems as defined and as implied by the reviewer. The benefit of the proposed NE approach is that it is useful in a) transport-limited systems b) highly reactive system that are not transport limited, and also in c) intermittently connected systems.

Interactive comment on Hydrol. Earth Syst. Sci. Discuss., 9, 10487, 2012.

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