On the meaning of porosity, hydrodynamic porosity and transport

Jesus Carrera

GHS, IDAEA, CSIC, Barcelona, Spain

Review of; “Hydrodynamic Porosity: A Paradigm Shift in Flow and Contaminant Transport Through Porous Media, Parts I and II”, by August H. Young and Zbigniew J. Kabala

1. Introduction

This document contains a review of two papers entitled “Hydrodynamic Porosity: A Paradigm Shift in Flow and Contaminant Transport Through Porous Media” (parts I and II, Young and Kabala, 2023a and b, respectively). In these papers, the authors introduce the concept of hydrodynamic porosity, as the fraction of porous medium connected through flow lines traversing the medium. Further, they find a relatively simple expression for the relationship between hydrodynamic and (total) porosity, as a function of Reynolds number. While this is a relevant contribution, the authors make a number of statements that are, not only ungranted, but also misleading. Together with other contributions by other authors in “fashion” journals, these statements add confusion to the transport field. The spirit of HESS discussions (Copernicus, n.d.) calls for “public commenting on the submitted preprint **by the referees, authors, and other members of the scientific community ... stimulate further deliberation...**”. In this spirit, prior to the actual assessment of the papers, I prefer to clarify some basic issues that are generally accepted the scientific community. Therefore, prior to the actual review in Sections 4-8, I discuss basic concepts on porous media (section 2), and (anomalous) transport (section 3).

2. Porosity, specific surface and permeability are well defined and not transport attributes

A porous medium consists of one or several fluid phases (unless specified otherwise, I will assume a single fluid, water) and, usually, several solid phases (minerals, biofilms, organic matter, etc.). The space between solid phases is called “void space”. Porosity is the ratio of void volume to total volume of a porous medium. Specific surface is the interfacial surface between solid phases and void. Given the multiplicity of phases, the concept of specific surface is also the subject of debate. Note that these definitions simply describe the geometric structure of the medium. They are relevant for all phenomena that may occur in the medium, but in principle, they are independent of those phenomena. They may change through coupling. For example, porosity may change in response to changes in stress, which is key factor in mechanical coupling and in the definition of storage, or in response to dissolution/precipitation, which is one of the motivations of reactive transport. But, on a first approximation, changes are assumed small (or taking place over very long times), so that they are neglected.

Permeability is more complex. In principle, it represents momentum conservation (viscous forces exerted by the medium counter pressure forces exerted by the fluid). Darcy’s Law results from the fact that that viscous forces are proportional to velocity. The critical issue here is that, for Darcy’s law to be valid, the geometry of flow lines must remain unchanged when the flux is changed, which requires flow to be “slow laminar” ($Re<1$, where Re is Reynolds Number, though often accepted $Re<10$). For higher Reynolds numbers, it is usually accepted that the energy loss is more than proportional to the flux. Young and Kabala (2023a) provide a lucid discussion on this topic in Section 4.2.1.

The summary of this section is that the key hydrodynamic parameters of porous media (porosity, specific surface and permeability) are well defined. They are not attributes of any specific phenomenon, but descriptors of the pore geometry. Certainly, using them is not "oversimplistic".

3. On solute transport and the adjectives of porosity

The above concepts are not sufficient to simulate complex phenomena, especially because of the heterogeneous nature of geologic materials. Therefore, researchers have been forced to introduce additional parameters. In particular, numerous adjectives have been added to porosity in an effort to improve the accuracy of transport formulations. A small fraction of these adjectives are mentioned by Young and Kabala (2023a) in Section 2.1. So, I agree with them that the term effective porosity would be ambiguous. However, what I want to emphasize here is that the main goal of these alternative definitions is not so much to study the structure of porosity or to distinguish between mobile and immobile water as to describe solute transport. In fact, the original work of Van Genuchten and Wierenga (1976) was motivated by the anomalous transport features they frequently observed in breakthrough curves. While they mentioned unsaturated flow conditions, which enhance the role of low permeability aggregates, their work has been extended to saturated flow conditions by numerous authors. Carrera et al. (2022) review the various alternative formulations that have been produced and that, directly or indirectly, can be considered extensions of the mobile-immobile model. They argue for the need of this model to account for observed chemical localization, which is essential for remediation. Therefore, I do not consider these models to be outdated, but adequate to represent the numerous departures from "normal" (i.e., Fickian) transport.

Dead end pores are one cause of "anomality" (it is ironic that we term "anomalous" what is consistently observed in reality. However, I doubt they are critically relevant. Diffusion dominates at the pore scale, as shown in Table 1, which compares pore advection and diffusion times for a range of water flues (from around 1cm/d to 1 m/d), and pore sizes (form 1 cm to 10 microns). It is clear that, except for gravels, diffusion dominates. That is, transport is diffusion dominated in the regions with largest specific surface, which are the ones most likely to hold contaminants.

Table 1: Reynolds and Peclet Numbers for a range of water fluxes and pore sizes typical of unconsolidated aquifers.

Flux (m/s)	mean veloc (m/s)	Pore size (m)	Reynolds num vd/nu	adv time (s)	Diff time (s)	Pore Peclet (-)
1E-07	4E-07	1E-02	1E-03	3E+04	5E+04	2.000
1E-06	4E-06	1E-02	1E-02	3E+03	5E+04	20.000
1E-05	4E-05	1E-02	1E-01	3E+02	5E+04	200.000
1E-07	4E-07	1E-03	1E-04	3E+03	5E+02	0.200
1E-06	4E-06	1E-03	1E-03	3E+02	5E+02	2.000
1E-05	4E-05	1E-03	1E-02	3E+01	5E+02	20.000
1E-07	4E-07	1E-04	1E-05	3E+02	5E+00	0.020
1E-06	4E-06	1E-04	1E-04	3E+01	5E+00	0.200
1E-05	4E-05	1E-04	1E-03	3E+00	5E+00	2.000
1E-07	4E-07	1E-05	1E-06	3E+01	5E-02	0.002
1E-06	4E-06	1E-05	1E-05	3E+00	5E-02	0.020
1E-05	4E-05	1E-05	1E-04	3E-01	5E-02	0.200

Further insight can be gained from random walk simulations by Bolster et al. (2009) shown in Figure 1, which illustrate that diffusion causes solutes to equilibrate in the recirculating (i.e., closed flowlines) flow regions (if anything, recirculation accelerates equilibrium).

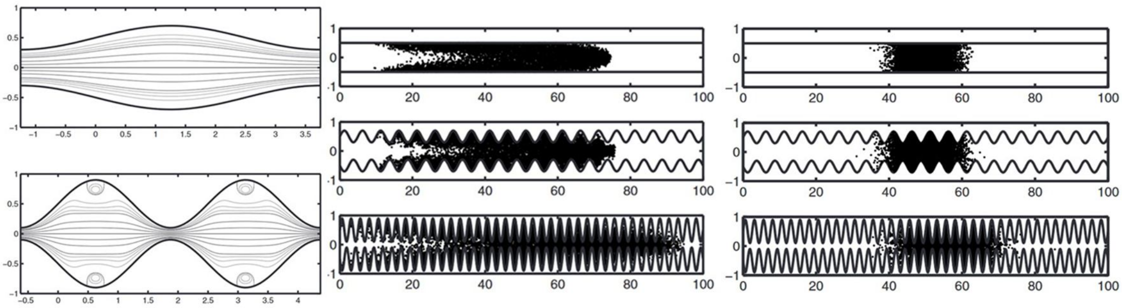

Figure 1: Simulations of Flow through a channel with periodically varying aperture: Flow lines (left) and particles locations resulting from random walk simulations with Pe=1000 (center) and Pe=10 (right) (modified from Bolster et al, 2009)

The ultimate motivation for this long discussion is that what controls the mean arrival is the total porosity. Velocity can be locally very large but diffusion will tend to equilibrate immobile regions at the pore scale, as shown in Figure 1. The key parameter is not so much the mobile porosity as the time it takes for equilibrium. Figure 2 displays breakthrough curves obtained with multi-rate-mass-transfer (multiple immobile zones characterized by a memory function with log-log slope of ½ for varying characteristic diffusion times, Carrera et al., 1998) model for transport along a 9 cm long column. All models are identical except for the mobile-immobile porosities (the total porosity is always 0.4). When the characteristic diffusion time is much smaller than the advection time, the curves are virtually identical. Peak arrival is fast with small mobile porosity only when the characteristic diffusion time is much larger than the advection time. In all cases, the mean arrival time is the same (t=V_w/Q), as demonstrated for this kind models by Haggerty and Gorelick (1995) and Carrera et al. (1998).

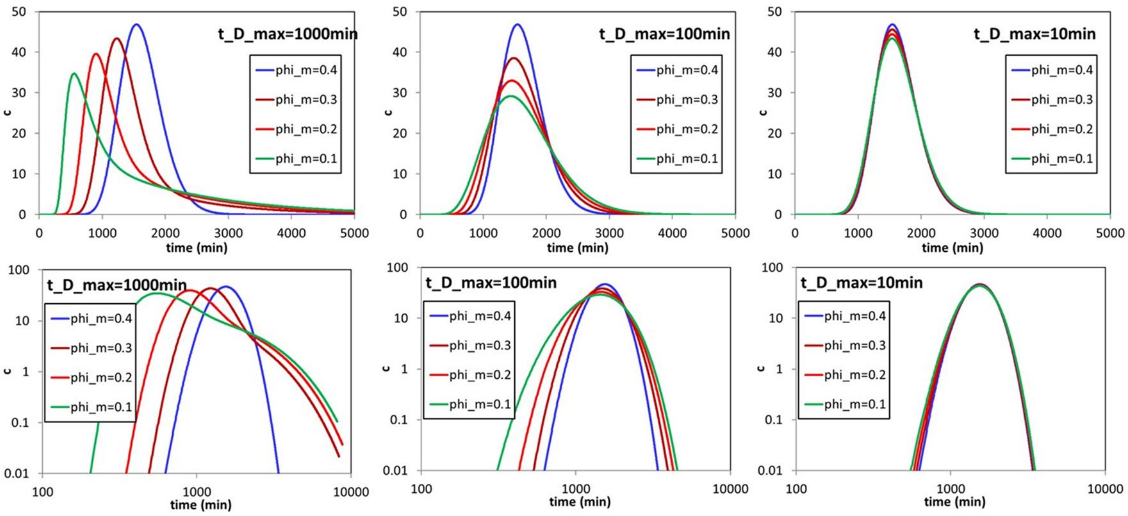

Figure 2: Simulations breakthrough curves of a pulse injection in arithmetic (above) and log-log (below, to illustrate that fast arrival BTCs display longer tails) varying the mobile porosity (total porosity is always 0.4) and the memory function maximum time. Mean arrival time is the same (1624 min) for all curves.

A last comment, while these models were originally derived for diffusion into immobile zones (soil aggregates, rock matrix blocks, or even intragranular discontinuities, Wood et al., 1990), similar models have been developed for “slow advection (Berkowitz and Scher, 1995). Unfortunately, the models are mathematically identical (see discussion by Carrera et al., 2022). Medina and Carrera (1996) showed that advective and diffusive models that calibrate equally well under laboratory or field tracer test conditions may lead to radically different results under natural field conditions (typically, much slower mean flux). Carrera et al. (1998) suggested using tracer tests with different fluxes to distinguish advective and diffusive exchange. I emphasize this because, to me, the most notable result of Young and Kabala (2023a and b) is precisely that exchange displays an advective component even in dead-end pores.

4. Overall assessment of the papers by Young and Kabala (2023a and b)

The first paper (Young and Kabala, 2023a) can be divided in two parts. The first part is devoted to argue that (1) pump and treat has not been very effective because done under constant flow rate, and (2) porosity is not a single value, but depends on velocity. The second part is devoted to show numerical simulations of laminar flow along a straight pore with a square cavity to the side. Simulations are performed by varying the flow rate (Reynolds Number) and the ratio of pore width to cavity depth. Simulations show a closed circulation area (vortex). The authors then separate the pore space in the area where water flows through (continuous flowlines from inflow to outflow boundaries) and the area where it does not (vortex). The ratio of the first to the total area is termed “hydrodynamic porosity” and its fraction of the total porosity is fitted to a constant plus an exponential of the Reynolds Number.

The second paper (Young and Kabala, 2023b) is an extension of the first one, where the cavities (“dead-end” pores) are extended to other geometries (rectangular, triangular, and semicircular). The resulting hydrodynamic porosity fraction is also fitted to a constant plus an exponential of the Reynolds Number.

The topic of the papers is appropriate for HESS and they are generally well written (see editorial comments below) in the sense that they are understandable. However, the tone is too self-serving, uncritical of their own work and critical of everyone else. Worse, much of the text (basically the 10 first pages) is irrelevant to the actual results (plus questionable, see Section 6 below). Challenging the views and definitions generally accepted by the scientific community is needed and will lead to badly needed “paradigm shifts”, but I am afraid that the challenges are poorly argued and the results do not question current views.

In summary, I think that the point the authors try to make is not supported by their results (actually, it is the opposite, see section 5 below). This, together with the questionable 10 introductory pages of paper I, imply that **the papers cannot be published in their current form**. Still, I think the work is of value (I found their figures fascinating) and many discussions lucid. Therefore, I recommend the authors to (1) revise their papers according to the general comments in Section 5 below, (2) revise the first 10 pages of paper I according to the comments in Section 6, (3) clean-up and clarify the text according to the comments in Sections 7 (paper I) and 8 (paper II).

5. General comments

1. The authors do not show that transport occurs in the hydrodynamic porosity (only advection does). As shown in Section 3, these immobile zones tend to equilibrate with the hydrodynamic porosity in a very short time (ranging from milliseconds to hours, which is small given the typical residence times). What the authors show is that water in “immobile” dead ends is not really immobile. This is paradoxical, because their results imply that equilibrium will occur faster than predicted by diffusion. This implies an

additional dispersion mechanism, discussed by Bolster et al. (2009). Unfortunately, Young and Kabala (2023a and b) do not discuss the velocity, shear, or curl of their vortices. Therefore, it is hard to ascertain this effect, although I suspect it will be very small for the range of Reynolds numbers studied here.

2. The relationship identified between Re and θ_{mob} is neither discovered (it is fitted and hardly discussed why) nor exact. For one thing, θ_{mob} is not just a function of Re (this was my first disappointment). You fix the dependence by fitting θ_{mob} to a set of Re values, having fixed all other parameters (pore geometry, dead end shape, viscosity).
3. It cannot be considered theoretically based. For Darcy's Law to be valid (see discussion in Section 2), the slope should be zero near the origin. But the slope is maximum at the origin with the proposed expression (In fact, the θ_{mob} graphs suggest that indeed θ_{mob} tends to become constant as Re tends to zero).
4. As a result, the fits are good, but not exact. Certainly, the coefficient of determination, R^2, is not "approximately 1" (this is stated in the papers abstracts of the two papers!) as clearly seen in Figures 13 of both papers. In fact, simple inspection of the one in paper II suggests a R2 of 0.99, instead of the 0.9999 reported in table. R^2 is a rather forgiving parameter. We all use it, but exaggerating it is not appropriate. A R^2 of 0.99 to fit 8 points with four parameters is not outstanding (unless the model has a theoretical basis)
5. But the problem is more severe, as it is not clear what is being fitted. At the beginning, ξ is defined as the ratio of θ_{mob} (wouldn't be more clear θ_{hyd}?) to θ. But it is never used afterwards. Instead Figures 13 display $\theta_{mob}/\theta_{MIM}$. I have failed to understand what θ_{MIM} is. It is defined in Equation (13) as $\theta_{MIM} = \xi_{MIM}\,\theta$, where ξ_{MIM} is "determined by the relative magnitudes of the through-channel and cavity volumes for each dead-end pore" (determined, how?, certainly, it is not the ratio, because, if so, Eq. 13 would not make much sense. In this context, the statement "For example, using Eq. (2), we find that for the square cavity, $\xi_{MIM} = 4/5$" leaves me perplexed. In summary, I am not sure what is being fitted. This is frustrating for me, as reviewer, but also to potential readers. So, I have been forced to read the papers accepting that "somehow" the hydrodynamic porosity drops as the Reynold number increases.

In summary, I see value in the work done, but the presentation needs to be more realistic and accurate.

6. Why the introduction is inappropriate in Young and Kabala (2023a)

I generally agree on the 2 pages discussion on the ubiquity and severity of GW pollution, but it very marginally related to the paper objectives. Instead, it might be more appropriate to review the research community efforts to address solute and reactive transport through porous media.

Lines 90-100: The authors classify as "outdated" what everyone else does even before introducing their concept. And claim that the paradigm shift is related to shifting from "pump and treat with steady flow" to pulsed "pump and treat". It is well known that fluctuating the flow rate in any remediation scheme accelerates remediation (Davidson et al., 2004). But there are numerous explanations for this behavior, ranging from shock waves (Sorek et al., 1992 and 2010) to chaotic mixing, increase in dispersion by transient flow, or ejection by curls in dead end pores, which host pollutants. The latter is well argued by Kahler and Kabala (2016), but it is not addressed at all in this paper. Therefore, it leads to frustration. At first, I thought that this paper was about shock waves. After reading the paper by by Kahler and Kabala (2016), I realized that it was related to transient vortices, only to find that all simulations in both papers are steady-state.

The whole section 2 is devoted to define Effective Porosity as the fraction of the medium devoted to transmit water… at this stage, it is not clear what are the authors referring to. Yet they go on a lengthy criticism of the work by others and an ungranted praise of their own work. I found it amusing, but was frustrated by not really understanding what they are talking about.

Line 207-210: Except for deformable media, porosity is clearly a single scalar value (ratio of voids to total volume) that does not depend on flow. 7 pages into the text and I still do not know what this paper is about (probably something related to porosity). It is true that many adjectives are used with porosity, but you do not need to criticize everyone of them!

Line 213: A very basic concept is that "Darcy/Forchheimer velocity" is not a velocity, but the volumetric water flux. Please, do not introduce a new velocity term here (volumetric velocity?, no one uses this term!)

Line 235: "Given our previous discussion, we know that use of the medium's total porosity is an oversimplification", which discussion? Why oversimplification. Your equation (4) yields the mean velocity regardless. So, it is not any oversimplification. It is just a definition, what may be an oversimplification is its candid use for solute transport. So, I suggest that you define what you mean by "the total volume that is conducive to flow". As a result, Eq. 5 is meaningless at this stage (and we are in page 9).

Line 244: The immobile zone of Van Genuchten (spelling!) and Wierenga (1976) is NOT "defined by isolated volumes of cavities or dead-ended pore space adjacently located to well-connected, mobile regions. They refer to low permeability zones where water velocity is very small. This is especially severe in the unsaturated zone, where water and solutes (the primary goal of their work) can be isolated in highly retentive portions, to be bypassed by fast flows around,…. The good news is that we finally learn what you are talking about!

In summary, please rewrite sections 1-3 into a compact introduction motivating what you are going to do.

7. Editorial comments on paper I

Line 10: conducive, conductive, but the statement sounds awkward

Line 18: "Finally, we show that this exponential dependence can be easily solved for pore-scale flow velocity through use of only a few Picard iterations, even with an initial guess that is 10 orders of magnitude off". True, but irrelevant from a transport point of view. Probably not worth mentioning it in the abstract.
Line 25: I do not understand "domestic and global populations". Do you mean "urban and global"?

Line 29: "6.5 trillion liters" probably OK for fashion journals, but not needed for scientific journals.

Figure 8 and flow lines plots. I have found these figures puzzling and fascinating. Usually, flow lines are plotted at equal flow intervals, which is clearly not the case here (but do not change it, the figures would not be as beautiful). Instead, describe the color code. It appears that warm colors indicate higher velocity, but it would be nice to know how much.

The terminology of depth, width, depth into the cavity, normalized depth, etc. is often confusing and, I believe, inconsistent between the two papers (also inconsistent is the fitting description).

Line 546, as discussed earlier, v is not a velocity, but a flux. While the term “Darcy velocity” is widely used, I believe it is confusing in these papers.

The whole section 6.1 is a bit of an overshoot. The fixed point theorem ensures fast convergence of Picard iterations for functions as flat as yours. However, I would not emphasize it too much OK in the text, but not in the abstracts!!), just in case a mathematician looks at it.

The examples in Section 6.2 are very unfortunate. A velocity of 2800 m/s is higher that the velocity of sound. You cannot displace water at those velocities anywhere, much less in a porous medium. Please, revise that, just in case a hydrologist looks at it.

8. Editorial comments on paper II

Line 25: Equation 1 is a bit careless. Some terms are not clearly defined (v_{pore}?, it is a velocity, but it is not clear which), others are defined twice (a?), and c is defined as dimensionless (it should be s/m) and I am utterly confused about the units of d.

Lines 42-44: The last statement of the paragraph is bit mysterious: “Further, researchers can expand…”. What one would expect at the end of the introduction is a description of the specific objectives of your work.

Figure 1: I would say that what you display is a “washed” porous medium. Unwashed porous media typically contains lots of fines (power law distribution)

Figure 8 caption: I am not sure what you mean by “landscape orientation”. I assume you mean “plan view”, but this is a 2D object. Therefore, talking about orientation is confusing.
…

9. References

Bolster, D., Dentz, M., & Le Borgne, T. (2009). Solute dispersion in channels with periodically varying apertures. *Physics of Fluids*, *21*(5).

Berkowitz, B., & Scher, H. (1995). On characterization of anomalous dispersion in porous and fractured media. Water Resources Research, 31(6), 1461-1466.

Carrera, J., Sánchez-Vila, X., Benet, I., Medina, A., Galarza, G., Guimerà, J. (1998). On matrix diffusion: formulations, solution methods and qualitative effects. *Hydrogeology Journal*, *6*, 178-190.

Carrera, J., Saaltink, M. W., Soler-Sagarra, J., Wang, J., & Valhondo, C. (2022). Reactive transport: a review of basic concepts with emphasis on biochemical processes. *Energies*, *15*(3), 925.

Davidson, B., Spanos, T., & Zschuppe, R. (2004). Pressure pulse technology: an enhanced fluid flow and delivery mechanism. In *Fourth International Conference on Remediation of Chlorinated and Recalcitrant Compounds, Monterey, CA*.

Haggerty, R., & Gorelick, S. M. (1995). Multiple-rate mass transfer for modeling diffusion and surface reactions in media with pore-scale heterogeneity. *Water Resources Research*, *31*(10), 2383-2400.

Kahler, D. M., & Kabala, Z. J. (2016). Acceleration of groundwater remediation by deep sweeps and vortex ejections induced by rapidly pulsed pumping. Water Resources Research, 52(5), 3930-3940.

Medina, A., & Carrera, J. (1996). Coupled estimation of flow and solute transport parameters. Water Resources Research, 32(10).

Sorek, S., Bear, J., Ben-Dor, G., & Mazor, G. (1992). Shock waves in saturated thermoelastic porous media. *Transport in porous media*, 9, 3-13.

Sorek, S., Ronen, D., & Gitis, V. (2010). Scale-dependent macroscopic balance equations governing transport through porous media: Theory and observations. *Transport in porous media*, *81*, 61-72.

Youtube(n.d.) https://www.youtube.com/watch?app=desktop&v=d3CfXQbX_ag

Copernicus(n.d.)https://www.hydrology-and-earth-system-sciences.net/peer_review/interactive_review_process.html

Van Genuchten, M. T., & Wierenga, P. J. (1976). Mass transfer studies in sorbing porous media I. Analytical solutions. *Soil Science Society of America Journal*, *40*(4), 473-480.

Wood, W. W., Kraemer, T. F., & Hearn Jr, P. P. (1990). Intragranular diffusion: An important mechanism influencing solute transport in clastic aquifers?. *Science*, *247*(4950), 1569-1572.

Young, A. H., & Kabala, Z. J. (2023a). Hydrodynamic Porosity: A Paradigm Shift in Flow and Contaminant Transport Through Porous Media, Part I. *HESS Discussions*

Young, A. H., & Kabala, Z. J. (2023b). Hydrodynamic Porosity: A Paradigm Shift in Flow and Contaminant Transport Through Porous Media, Part II. *HESS Discussions*