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A Rebuttal Paper

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To the paper entitled **"An experimental study of pressure drop characteristics under single-phase flow through packed bed microreactors"** by Lu Zhang | Arne Hommes | Remon Schuring | Jun Yue, Oct. 25th 2024

INTRODUCTION

The paper to which this rebuttal paper is directed, was published in this calendar year 2024 in the AIChe Journal, DOI: 10.1002/aic.18640. I am a career scientist who has devoted his entire professional career to fluid flow in closed conduits. Accordingly, I was not only disappointed, but also, rather surprised, when on reading the paper in question, I noticed that none of my publications were referenced, even though I am the most recent global credible author who has published, comprehensively, on fluid flow in closed conduits. Accordingly, I feel compelled to rebut this paper because it is totally at odds with the teaching of the Laws of Nature.

THE QFFM

The abbreviation, QFFM, stands for the Quinn Fluid Flow Model, which is a comprehensive novel theory of fluid flow in closed conduits. It was published in the year 2019. It supersedes all extant fluid flow models on this subject matter, on the basis of experimental verification applicable to both empty and packed conduits, see Table 1A below.

In this analysis, I will focus only on the measurements with water as the fluid, taken in the paper under scrutiny. In this way, I will avoid the issue of fluid compressibility with the measurements using Nitrogen gas. The study involves four different diameters of packed beds containing glass beads. Those diameters are: 0.16, 0.10, 0.08 and 0.05 cm. Thus, we know that the particles are spherical in shape, rigid in structure and have surfaces that are hydrodynamically smooth. There were 6 different particle sizes used in the study and their diameters were reported as the Sauter mean diameters as follows, 63.7, 115.5, 244, 534.7, 734, 1,244 mm.

Table 1A

No.	Name	F	M	Title	Journal	year
1	Quinn	H	M	Reconciliation of Packed Column Permeability Data Part 1. The Teaching Of Giddings Revisited,	Special Topics & Reviews in Porous Media 1 (1), 79-86.	2010
2	Quinn	H	M	Reconciliation of packed column permeability data Column permeability as a function of particle porosity,"	Journal of Materials, vol. 2014, Article ID 636507, 22 pages,	2014
3	Quinn	H	M	A Reconciliation of Packed Column Permeability Data: Deconvoluting the Ergun Papers	Journal of Materials Volume 2014 | Article ID 548482 doi.org/10.1155/2014/548482	2014
4	Quinn	H	M	Some New Light on the Study of Fluid Flow in Closed Conduits: . An Experimental Protocol to Identify the Value of a Misconstrued Constant	Preprints.org 2019, 2019050367	2019
5	Quinn	H	M	Quinn's Law of Fluid Dynamics; Pressure-driven Fluid Flow through Closed Conduits.	Fluid Mechanics. Vol. 5, No. 2, pp. 39-71. doi:10.11648/j.fm.20190502.12.	2019
6	Quinn	H	M	Quinn's Law of Fluid Dynamics: Supplement # 1 Nikuradze's Inflection Profile Revisited.	Fluid Mechanics. Vol. 6, No. 1, 2020, pp. 1-14. . doi: 10.11648/j.fm.20200601.11	2020
7	Quinn	H	M	Quinn's Law of Fluid Dynamics: Supplement # 2 Reinventing the Ergun Equation.	Fluid Mechanics. Vol. 6,No. 1, 2020, pp. 15-29. doi: 10.11648/j.fm.20200601.12.	2020
8	Quinn	H	M	Quinn's Law of Fluid Dynamics: , Supplement #3 A Unique Solution to the Navier-Stokes Equation	Fluid Mechanics. Vol 6, Issue 2, December 2020 , pp. 30-50. doi: 10.11648/j.fm.20200602.11	2020
9	Quinn	H	M	Critique of recent paper in the Journal of Powder Technology by Buckwald et al. (2020)	Powder Technology 394(2) 10.1016/j.powtec.2021.08.067	2021
10	Quinn	H	M	Quinn's Law of Fluid Dynamics, Supplement #4 Taking the Mystery out of Permeability Measurements in Porous Media,	Fluid Mechanics. Volume 8, Issue 1, June 2022 , pp. 1-15. doi: 10.11648/j.fm.20220801.11	2022
11	Quinn	H	M	A Smoking Gun Scenario Relative to Fluid Dynamics in Closed Conduits,	American Journal of Physical Chemistry. Volume 11, Issue 4, December 2022 , pp. 120-127. doi: 10.11648/j.ajpc.20221104.15	2022
12	Quinn	H	M	A Fluid Dynamic Development Like None Other	European Journal of Applied Sciences Vol. 11 No. 2 (2023): https://doi.org/10.14738/aivp.112.14344	2023
13	Quinn	H	M	The Fluid Dynamics of Conduit Hydrodynamic Entrance Effects Explained A Rebuttal Paper	European Journal of Applied Sciences Vol. 11 No. 3 (2023): DOI:10.14738/aivp.113.14714	2023
14	Quinn	H	M	The Solution Equivalent of the Navier-Stokes Equation in HPLC	SCIREA Journal of Mechanics Volume 4, Issue 1, February 2023	2023

METHODOLOGY

In evaluating this paper, I accept as valid, all measurements of flow rate and pressure drop. This is a reasonable conclusion since it is broadly accepted that volumetric flowmeters and pressure transducers are highly accurate. On the other hand, the measurements of particle diameter and packed column external porosity are universally regarded as fraught with problems. I then use the teaching of the QFFM to back-calculate the values for the average spherical particle diameter equivalent, d_p, as well as the packed column external porosity, e_0.

PERMEABILITY

I begin by showing the correlation achieved by using this methodology between the measured data and the calculated data based upon the QFFM. I present the results here in the form of a permeability plot for each individual particle diameter. In all, there are 6 plots identified as Fig 1A through Fig 1F.

Fig 1A

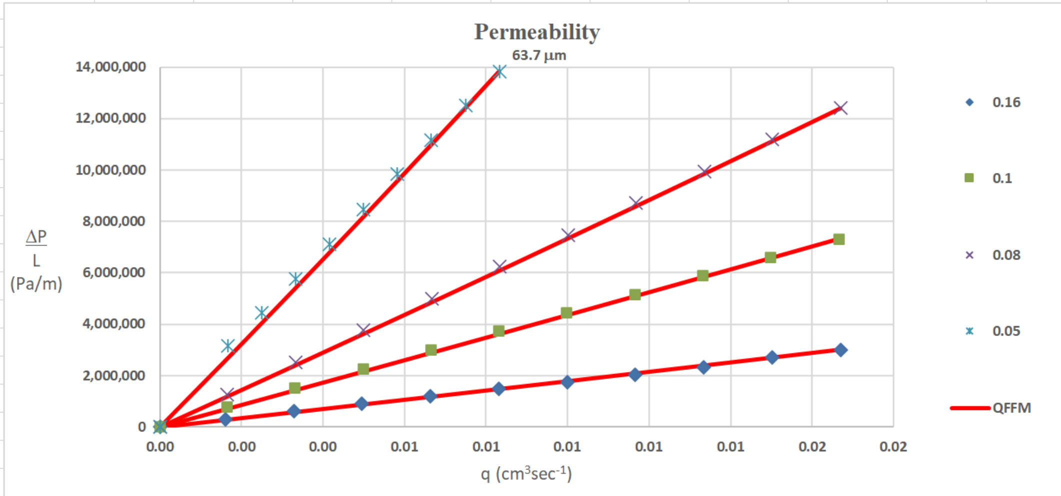


Fig 1B

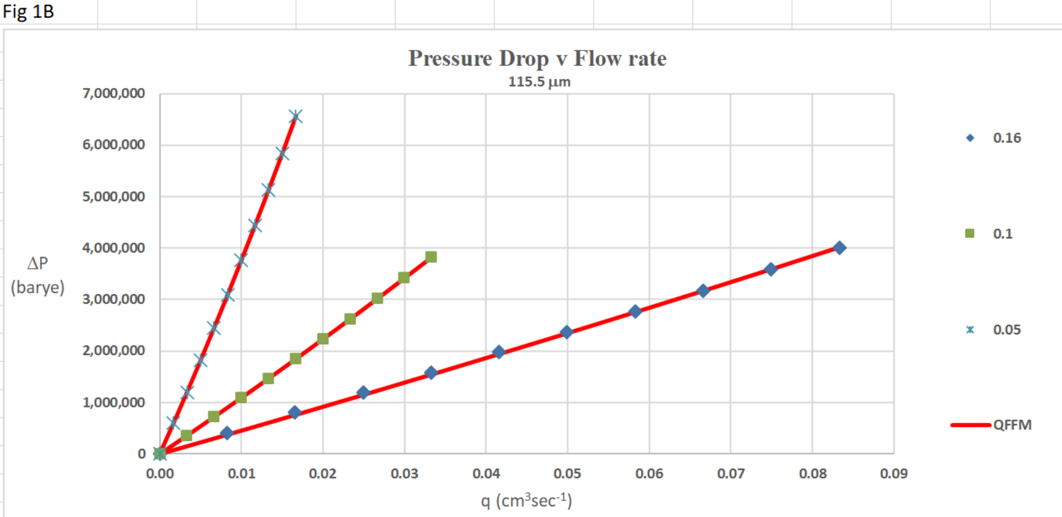


Fig 1C

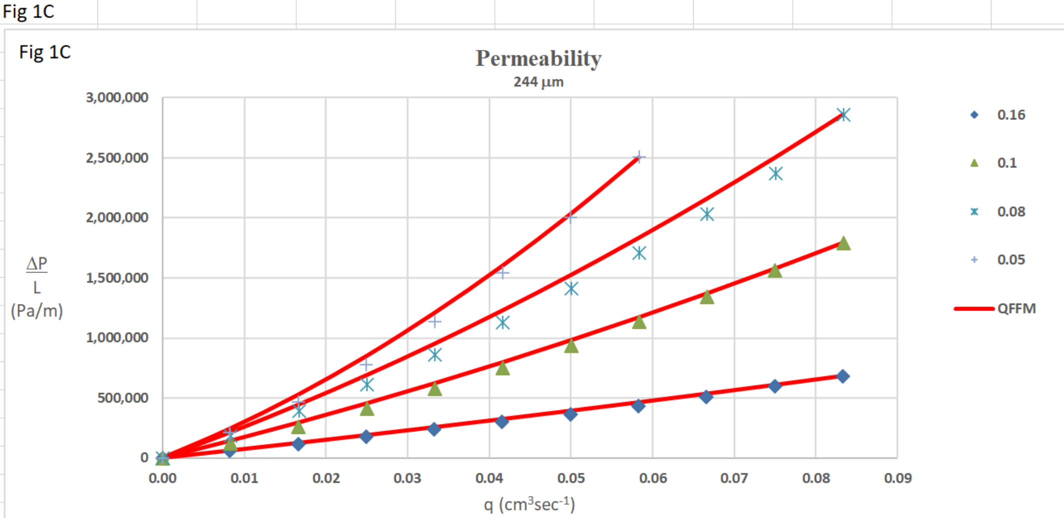


Fig 1D

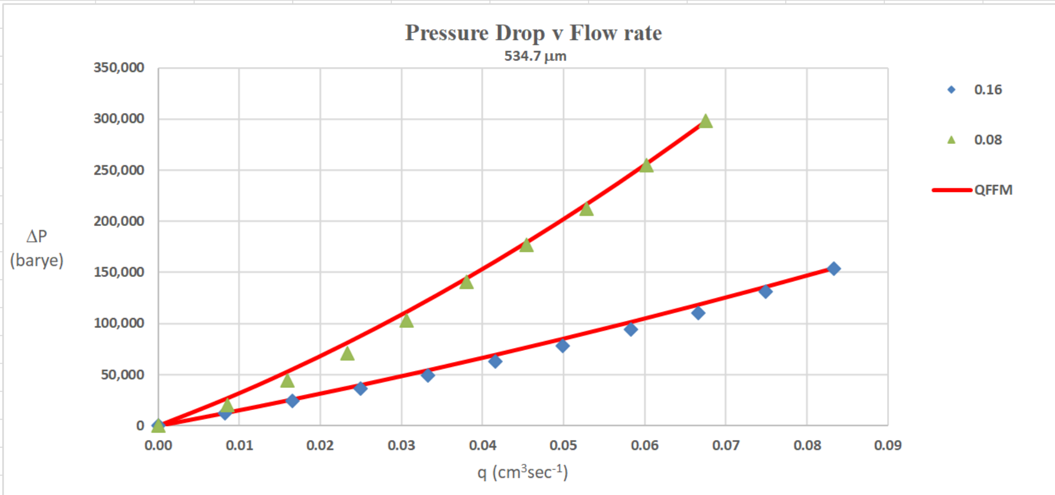


Fig 1E

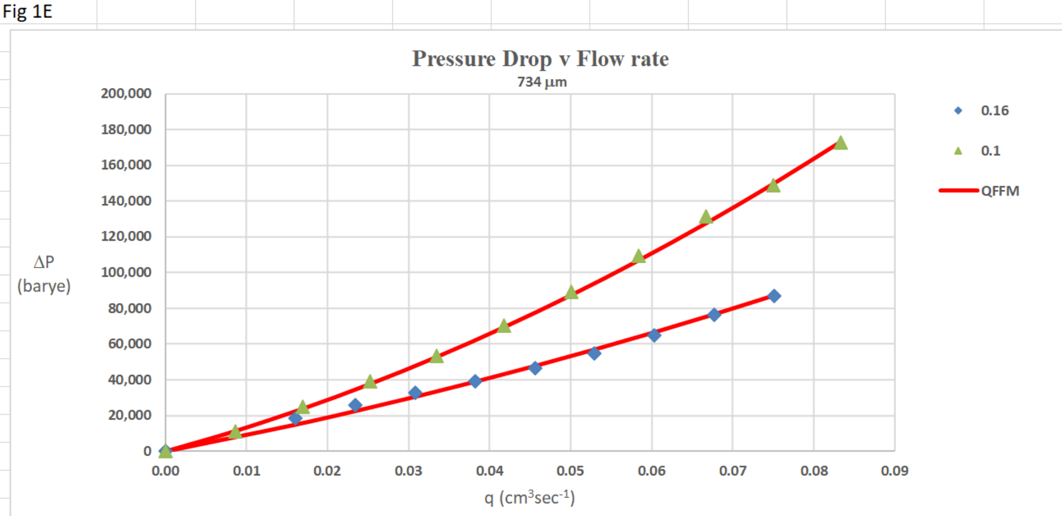
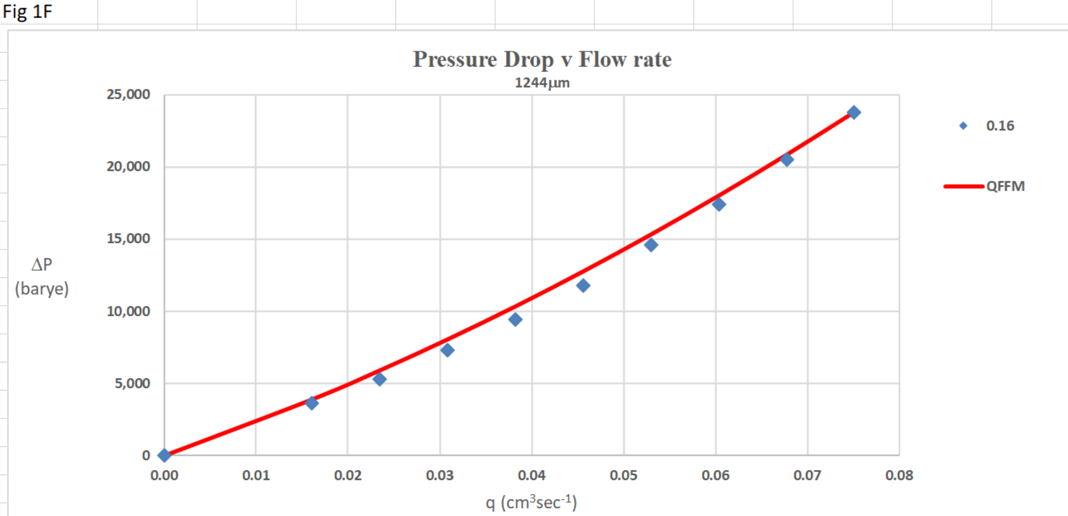


Fig 1F

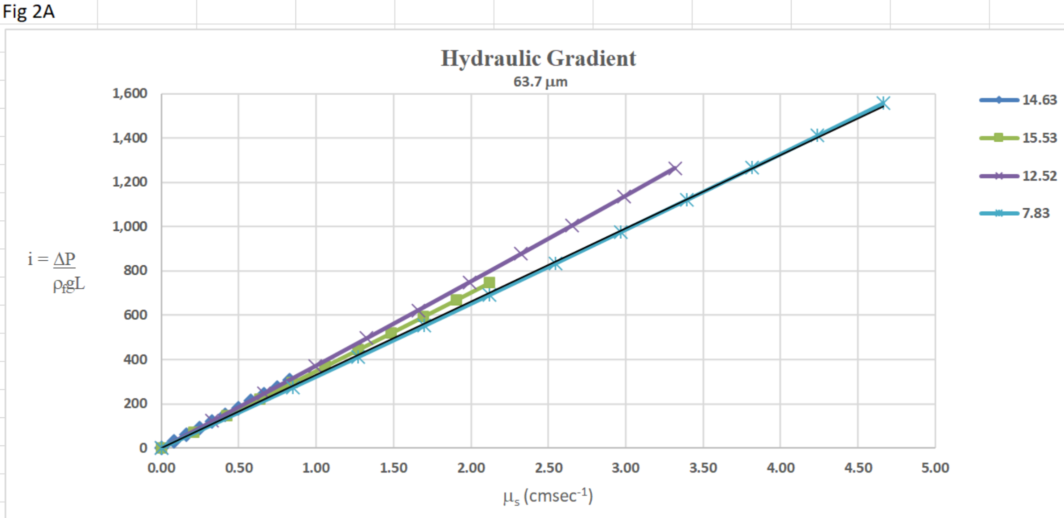


As can be seen from the plots, there is an excellent correlation between the measured and calculated data. These plots, then, represent my *bona fides* with respect to my analysis of this study which is totally driven by the accurately measured data of volumetric flow rate and differential pressure.

HYDRAULIC GRADIENT

The QFFM methodology is based upon the Forchheimer model which balances the measured and calculated data using a quadratic relationship between hydraulic gradient, $i = [DP/(r_f gL)]$ and fluid superficial velocity $m_s = [4q/(pD^2)]$, where q is the measured fluid volumetric flow rate, DP is the measured pressure differential and D is the diameter of the empty column. The linear and quadratic coefficients of the 2nd order polynomial of this relationship, a and b, respectively, also referred to typically as "Forchheimer Coefficients" are in reality, "fudge factors", which guarantee a perfect fit between the measured and modelled data. The hydraulic gradient is calculated based upon two additional universal variables which are the fluid density, r_f, and the acceleration due to gravity, g. Therefore, the Forchheimer model does not depend on either the value of the particle diameter d_p or the external porosity of the packed column e_0 but does incorporate two additional "pegs in the ground" not found in any of the fluid models which pertain to the linear (laminar) flow regime. As an example, we show a plot of the hydraulic gradient in Fig 2A for one of the data sets in this study, 63.7 mm particles.

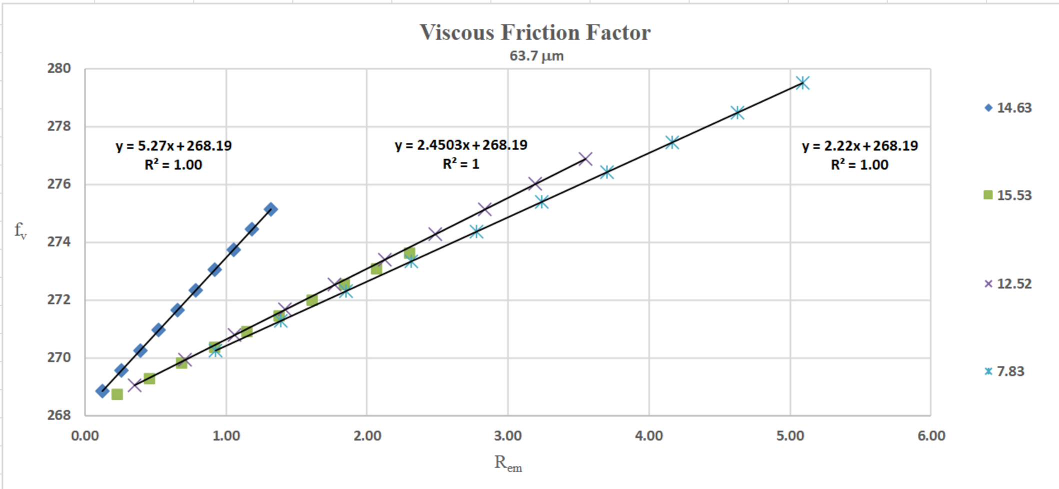
Fig 2A



We point out that, in this plot, the shape of the lines appear to be linear rather than pronounced curves. This is because the values of the modified Reynolds number, R_{em}, are relatively low. When measurements are taken at higher and higher values of the modified Reynolds number, however, these lines become increasingly curved in shape.

VISCOUS TYPE FRICTION FACTOR f_v - THE Q- MODIFIED ERGUN MODEL

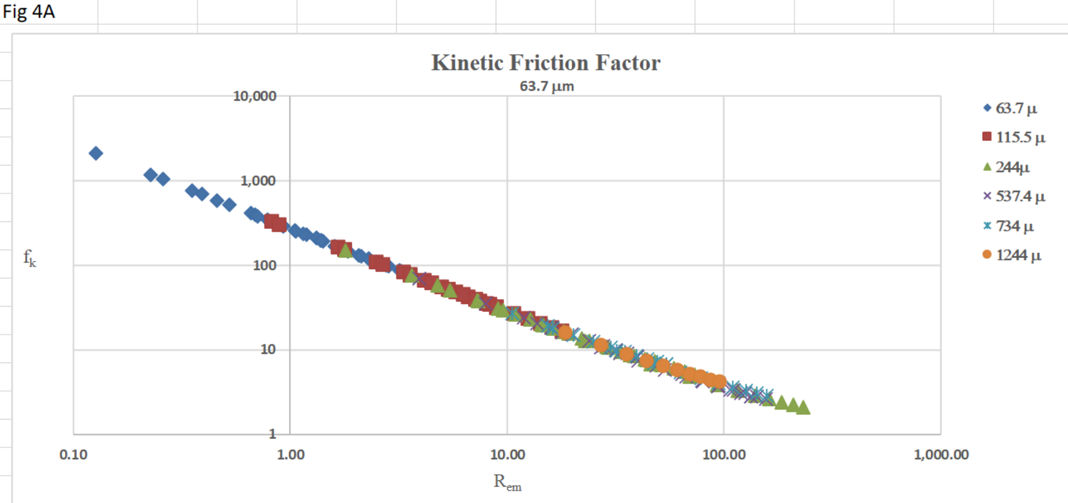
Fig 3A



As shown in Fig 3A, we display the data for the 63.7 mm results as a viscous type friction factor, f_v, versus the modified Reynolds number. This relationship was originally taught by Ergun circa 1951 and it enables the calculation of the Ergun coefficients A and B as the intercept and slope of the plotted lines. In Fig 3A herein, of course, the values of A and B represent the coefficients of the Q-modified Ergun model which means that the original Ergun model is modified according to the teaching of the QFFM. Note that in the Q-modified model, the value of A is always a constant = 268.19, but the value of B is not constant and, rather, is defined by the relationship B = $[l/(2pe_0^3)]$, where l = the wall normalization coefficient. These values are in contrast to the original Ergun model values of 150 and 1.75 for the values of A and B, respectively. Note also, that the QFFM teaching is unique amongst all extant models, in as much as it has a built-in methodology to account for "wall-effect" via its parameter l.

KINETIC TYPE FRICTION FACTOR f_k

Fig 4A

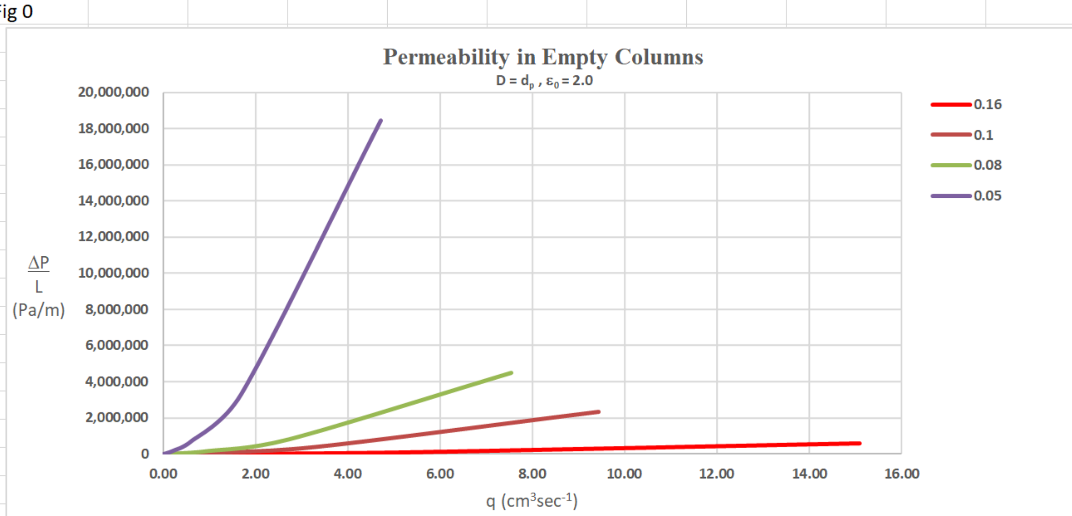


As shown in Fig 4A, using the kinetic type friction factor which was also disclosed by Ergun in his original publication circa 1951, all of the measured data falls on the same straight line. This plot reveals two important elements of this study. Firstly, none of the measurements taken were in the fully developed turbulent flow regime, i.e., the maximum value of the modified Reynolds number did not exceed a value of 250. Secondly, this kinetic type friction factor reveals very little about the underlying parameters driving this study, i.e., it is a useless parameter for this particular study. This, of course, is in contrast to the very valuable information embedded in the plot for the viscous type friction factor, shown above in Fig 3A.

THE WALL-EFFECT-A CRITICAL DISTINCTION EMBEDDED IN THE QFFM

A major objective of this experimental study, as stated by the authors, was to assess the impact of the so called "wall-effect". The QFFM, as stated earlier, is unique, to the extent that it has a built-in mechanism in its teaching to accommodate the wall-effect. So, before I present my analysis of the impact of the wall-effect in this study, I need to establish a frame of reference. That frame of reference is based upon the permeability of empty columns, i.e., columns in which there are no *solid* particles. Note that I refer to the "microreactors" terminology of the authors as "columns". This is because I choose to use the jargon typically used in HPLC, (High Pressure Liquid Chromatography), since this study seems to have a definite influence from this field of study. One of the many novel components of the QFFM is that it is a universal theory of fluid dynamics which pertains to both empty and packed columns. Since the wall-effect is well understood and most pronounced in empty columns, the QFFM is well suited to compare its impact in both types of fluid flow embodiments. Accordingly, I will use the QFFM to establish the frame of reference for the wall-effect in all 4 empty columns corresponding to those used in this study. To do that I initially observe that this study only involves the "primary" wall-effect, i.e., since the particles used in the study have smooth surfaces and are, therefore, hydrodynamically smooth, I need not be concerned with the "secondary" wall-effect. In other words, I need not consider the sand wall roughness effects disclosed by Nikuradze in his classical study of wall roughness. I begin by displaying in Fig 0 below the permeability data pertaining to 4 empty columns having the values for D of 0.16, 0.1, 0.08 and 0.05 cm, based upon the teaching of the QFFM.

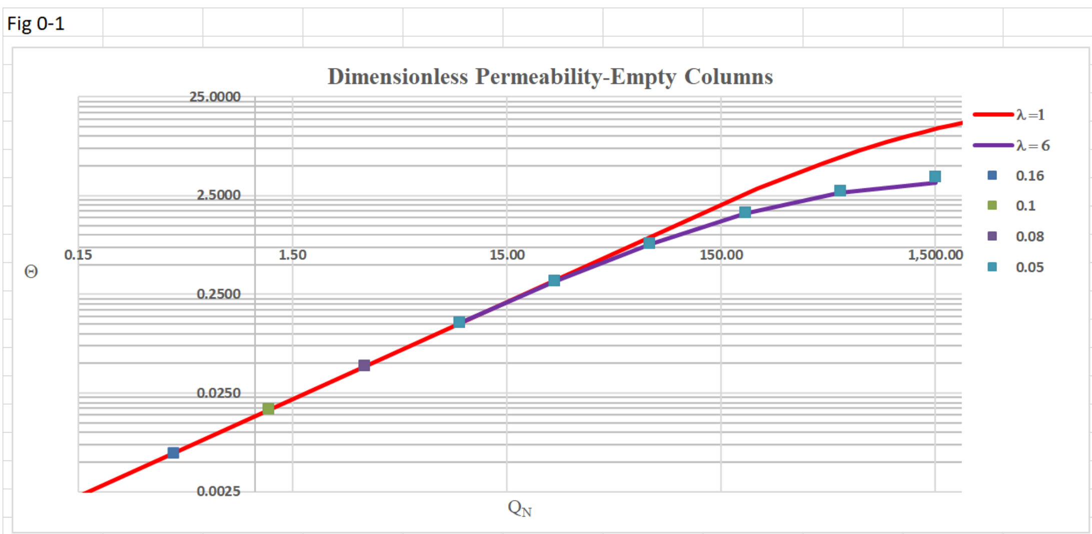
Fig 0



It will be understood that in the teaching of the QFFM, the boundary conditions for empty columns are threefold: D = d_p, e_0 =2.0 and e_p = 1.0, where e_p represents the porosity of the particles within the packed column. Stated in layman's terms these are: the value of the diameter of the empty column is equivalent to the diameter of the hypothetical particles whose porosity value is unity, i.e., the particles are made of free space (no solid skeleton): the external porosity of the empty column is 2.0. Given these three boundary conditions, the QFFM teaches that packed and empty columns are hydrodynamically identical.

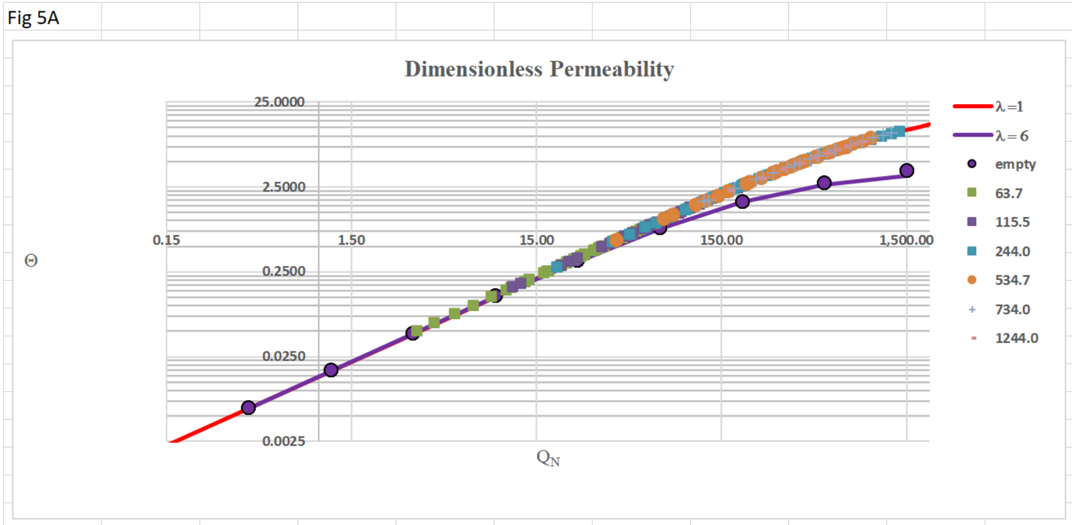
THE WALL-EFFECT IMPACT EXPRESSED AS THE VALUE OF L

As taught by the QFFM, the primary wall-effect is due to both the velocity and viscosity of the fluid in the close proximity to a confining wall and was identified as the viscous boundary layer by Prandyl circa 1930. The parameter l in the QFFM quantifies the magnitude of the impact of this boundary layer on the permeability of any packed or empty column. To isolate the impact of the value of l, therefore, the QFFM uniquely defines the Dimensionless Permeability in a plot of Q versus Q_N, where Q = $4Q_N/f_v$, and $Q_N = R_{em}/e_o^3$. For the 4 empty columns chosen here, we show this plot in Fig 0-1 below.



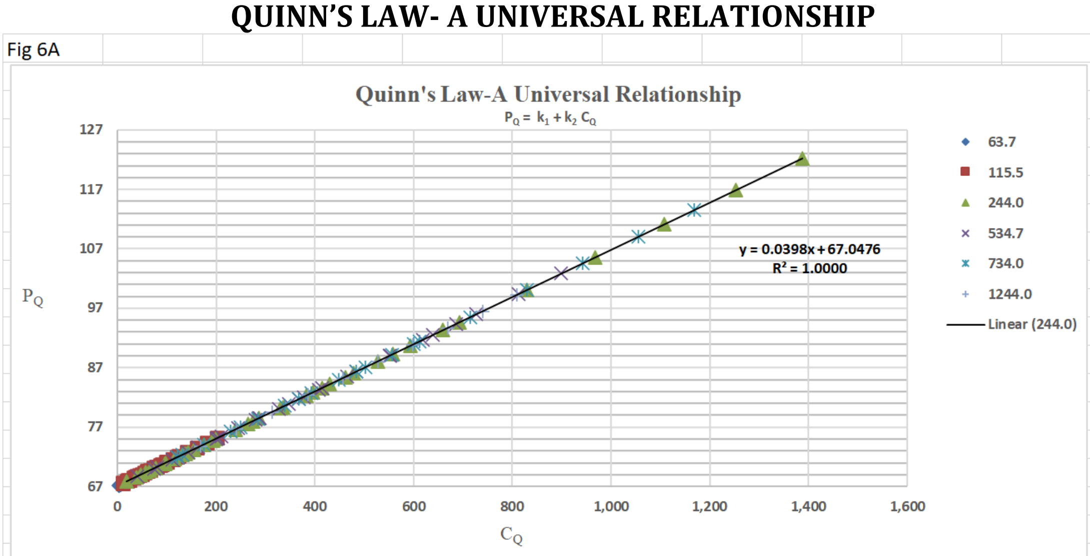
Looking at Fig 0-1, I note that a value of l = 1 represents that of a packed column which is free of all wall-effects, meaning both primary and secondary effects. Note also that the 4 empty columns have identical and significant wall-effect representing a value of l = 6 approx. The shape of the line representing the 4 empty conduits is explained by the fact that the boundary layer is relatively thick at lower values of the parameter Q_N, (Low values of R_{em}) and as the value of the modified Reynolds number increases, the boundary layer is increasingly dissipated resulting in an ever-increasing wall-effect impact on permeability.

THE IMPACT OF WALL-EFFECT ON THE PACKED COLUMNS - A COMPARISON



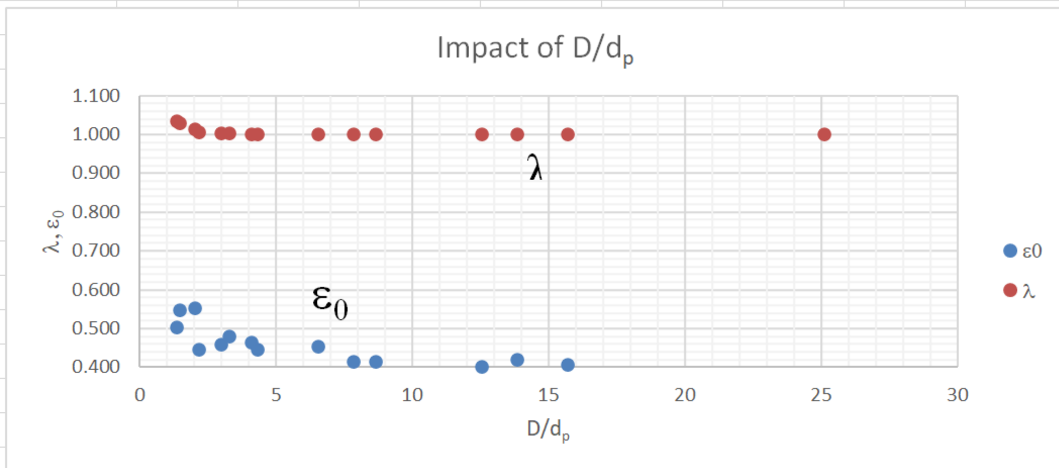
In Fig 5A, I show a comparison of the wall-effect in both the empty and packed columns. Note that there is a negligible impact of the wall effect in the packed columns, i.e., all packed columns in this study have a value of l close to unity. This lack of wall-effect impact in the packed columns, as taught in the QFFM, is due to the tortuosity, $t = n_{pq}D/(Le_0^3)$, of the fluid path in a packed column, where n_{pq} is the volume of the empty column expressed in terms of the volume of a single particle of diameter d_p. Accordingly, it is apparent that in all the packed columns in this study, the tortuosity is very large as compared to the tortuosity in an empty column, which is very small and has the constant value of 3/16, regardless of the dimensions of the empty column. Consequently, since the primary wall effect $W_1 = [b_0^{(1/3)}/t]$, where b_0 represents the instantaneous boundary layer, one can appreciate that in the packed columns in this study, the thickness of the boundary layer is significantly diminished resulting in a negligible impact on permeability.

QUINN'S LAW- A UNIVERSAL RELATIONSHIP



In Fig 6A, I display all the measured data on a plot of P_Q versus C_Q, which is now known as Quinn's Law. Note that all measured data fall on a unique straight line whose intercept and slope represent the values of k_1 and k_2 which are the universal constants in the pressure flow relationship in closed conduits.

Fig 7A



In Fig 7A, I show a plot of the relationship between the parameters e_0, and l versus the ratio of D/d_p. Note that as the value of the ratio of D/d_p decreases the other two parameter values increase.

DATA SUMMARY
Table 1

Reported					QFFM									Forchheimer		Q-Modified Ergun	
D	d_{sauter}	D/d_{sauter}	ε_0	μ_s	Column	M_p	L	d_p	D/d_p	Ω_p	ε_0	R_{em}	λ	a	b	A	B
cm	cm	none	none	cm/sec	No.	g	cm	cm		none	none		none	none	nonr	none	none
0.16	0.12440	1.29	0.570	0.04-0.5	1	0.250	10	0.120	1.33	0.965	0.483	18.0-95	1.03	0.45	0.06	268.19	1.45
0.16		1.29	0.558		2												
0.10	0.07340	1.36	0.624	0.04-0.5	3	0.094	10	0.074	1.36	1.003	0.503	16.0-157.0	1.03	0.98	0.07	268.19	1.29
0.08	0.05347	1.50	0.632	0.04-0.5	4	0.055	10	0.048	1.68	0.890	0.546	18.0-156.0	1.03	1.53	0.06	268.19	1.00
0.10	0.05347	1.87	0.587	0.04-0.5	5		10										
0.05	0.02440	2.05	0.601	0.04-0.5	6	0.021	10	0.024	2.05	1.001	0.552	23.0-230.0	1.01	5.46	0.11	268.19	0.96
0.05		2.05	0.637		7												
0.16	0.07340	2.18	0.435	0.04-0.5	8	0.268	10	0.073	2.18	1.000	0.445	10.0-54.0	1.01	1.78	0.16	268.19	1.82
0.16	0.05347	2.99	0.390	0.04-0.5	9	0.261	10	0.052	3.08	0.972	0.459	4.0-40.0	1.00	3.05	0.18	268.19	1.65
0.08	0.02440	3.28	0.435	0.04-0.5	10	0.063	10	0.023	3.48	0.942	0.480	7.0-73	1.00	12.63	0.30	268.19	1.44
0.10	0.02440	4.10	0.385	0.04-0.5	11	0.101	10	0.024	4.13	0.992	0.464	5.0-48.0	1.00	13.42	0.36	268.19	1.59
0.05	0.01155	4.33	0.423	0.04-0.5	12	0.026	10	0.012	4.28	1.011	0.445	2.0-18.0	1.00	70.37	1.00	268.19	1.81
0.16	0.02440	6.56	0.355	0.04-0.5	13	0.264	10	0.024	6.66	0.985	0.454	2.0-18.1	1.00	15.05	0.42	268.19	1.70
0.08	0.01155	6.93	0.374	0.04-0.5	14		10										
0.05	0.00637	7.85	0.384	0.04-0.22	15	0.028	10	0.006	7.83	1.002	0.415	0.9-5	1.00	320.42	2.90	268.19	2.22
0.05		7.85	0.407		16												
0.10	0.01155	8.66	0.354	0.04-0.5	17	0.110	10	0.012	8.10	1.070	0.415	1.0-9.0	1.00	85.65	1.50	268.19	2.22
0.08	0.00637	12.56	0.358	0.04-0.5	18	0.072	10	0.006	12.52	1.003	0.402	0.3-3.5	1.00	368.74	3.60	268.19	2.45
0.08		12.56	0.381		19												
0.16	0.01155	13.85	0.324	0.04-0.5	20	0.280	10	0.012	13.72	1.009	0.419	0.8-8.3	1.00	92.55	1.50	268.19	2.17
0.10	0.00637	15.70	0.321	0.04-0.5	21	0.112	10	0.006	15.53	1.011	0.407	0.2-2.3	1.00	344.78	3.30	268.19	2.36
0.10		15.70	0.315		22												
0.16	0.00637	25.12	0.314	0.04-0.5	23	0.333	10	0.011	14.63	1.717	0.311	0.1-1.3	1.00	359.50	11.22	268.19	5.27
0.16		25.12	0.319		24												

In Table 1, I display all our calculations based upon the teaching of the QFFM which are contained in the center columns of the Table. The reported data of the paper is displayed in the columns on the left-hand side of the Table and on the righthand side is contained the Forchheimer and Q-modified Ergun coefficients calculated via the QFFM.

COMMENTARY

In order to facilitate our analysis discussion, I have assigned a number to each of the packed columns reported in the paper. Thus, the Table displays a total of 24 columns and they are arranged in order of increasing ratio D/d. In other words, column number 1 has a D/d ratio of 1.29 and column number 24 has a D/d ratio of 25.12.

I discard column numbers 2,5,7, 14, 16,19,22 and 24, because I could not figure out where in the supplemental data online the specific underlying data for these columns was reported. Additionally, I also discard the data for column number 23 since it had an apparent value of external porosity of 0.311 which is prohibitively low. It is my experience in over 40 years of practice that one cannot pack rigid particles to an external porosity of less than 0.36. Accordingly, I suggest that there is something wrong with the reported data for this column and, therefore, I discard it from my analysis. This means I discard 9 columns from the 24 reported in the paper which leaves 15 columns worthy of analysis in this study.

CONCLUSIONS

Of the data reported for the 15 columns in this study, I conclude as follows:

1. There is no measurable impact on permeability due to wall-effect
2. The increased external porosity of the packed columns due to reduced values of the ratio D/d_p, is not a wall-effect *per se*, because it can also be caused by irregularly shaped particles, for instance, and, therefore, is not unique to the confining walls of the packed column.
3. The permeability measurements in this study were mostly confined to the transitional region of the fluid flow regime and contained no measurements representing fully developed turbulence.
4. The particle Sauter mean diameter value, d, is not the correct value to use in permeability considerations. This is because one cannot measure it accurately, when the particles are not perfectly spherical.
5. The spherical particle diameter equivalent value, d_p, is the correct value to use in permeability considerations. This is because it is based entirely on volume consumed in the packed bed which can be accurately calculated based upon the known volume of a perfect sphere.
6. When the particles are perfectly spherical, as in the case in this study, the value of d and d_p should be identical. As can be seen in Table 1, these values are not identical which means that the experimental technique used to measure the diameter of the particles by the paper authors was not sufficiently accurate.
7. The value of the particle sphericity parameter, W_p, identified in Table 1, is the parameter that connects the value of d and d_p. When the particles are perfectly spherical the value of W_p should be unity, but it is not.
8. The values of W_p demonstrate the error of measurements of particle diameter in this study.

9. Similarly, the discrepancy in the values of the external porosity, e_0, between that reported by the authors and that calculated by the QFFM, identifies the lack of precision of the experimental technique used by the authors to measure its value.
10. It is unfortunate that the authors did not report their measured values for either the mass of the particles, M_p, in each column, or the length L of the columns in the study.