Historical development of soil-water physics and solute transport in porous media

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Abstract The science of soil-water physics and contaminant transport in porous media began a little more than a century ago. The first equation to quantify the flow of water is attributed to Darcy. The next major development for unsaturated media was made by Buckingham in 1907. Buckingham quantified the energy state of soil water based on the thermodynamic potential energy. Buckingham then introduced the concept of unsaturated hydraulic conductivity, a function of water content. The water flux as the product of the unsaturated hydraulic conductivity and the total potential gradient has become the accepted Buckingham-Darcy law. Two decades later, Richards applied the continuity equation to Buckingham’s equation and obtained a general partial differential equation describing water flow in unsaturated soils. For combined water and solute transport, it had been recognized since the latter half of the 19th century that salts and water do not move uniformly. It wasn’t until the middle of the 20th century that scientists began to understand the complex processes of diffusion, dispersion, and convection and to develop mathematical formulations for solute transport. Knowledge on water flow and solute transport processes has expanded greatly since the early part of the 20th century to the present.

Keywords Convective-dispersion equation; Darcy’s equation; miscible displacement; Richard’s equation; soil-water potential

Introduction

The science underpinning the study of soil-water physics and solute transport is only slightly more than a century in development. Little literature exists on the scientific development of soil-water physics prior to the latter part of the 20th century. Fortunately, Philip (1974) published a paper that evaluated progress during the prior 50 years, and Gardner (1977, 1986) expanded on the history. Kutílek and Novák (1997) discussed the development of soil physics in eastern and central Europe and Raats (2001) discussed developments in soil-water physics since the 1960s.

Humans have tilled the soil, irrigated it, and drained it for agricultural purposes for at least 6,000 years. The recognition that the soil contains ducts or pores that are important for water movement to plants occurred about 2,000 years ago (Philip, 1974). The vital role of pores and pore geometry was further recognized by Evelyn (1676), but little physics was involved. The Aztecs of pre-Columbian Mesoamerica recognized that after a rain different soils held different amounts of water (Williams, 2006). Further developments in the science of soil had to await the development of the physical sciences in the latter part of the 17th century. Little physics-related soil research occurred, however, until the latter part of the 19th century. In Europe, E. Wollny edited a German journal on agricultural physics over the period 1878–98. In the USA, Hilgard, King, and Whitney pioneered physical studies on semi-arid region soils. The state of soil physics at the end of the 19th century was summarized in a textbook by Warrington (1900). Meanwhile, the physics of soil water was scarcely being considered by the beginning of the 20th century. However, the foundations for what would later be considered to be soil-water physics
were firmly being established in pure physics and mathematics. A timeline of major developments is given by Figure 1.

**Foundations in classical physics**

Isaac Newton (1642–1727) in the late 17th century developed the ideas of gravitational force and concepts of pressure, work, and energy so fundamental in understanding soil-water physics. In one of his writings, Newton mentions the puzzling phenomena of capillary forces lifting a wetting fluid in a tube. Leonardo de Vinci, however, is considered to be the discoverer of capillary phenomena. The concept of surface tension is attributed to a Hungarian mathematician, Johann Andreas von Segner. A decisive step in the understanding of capillarity occurred in 1805 in an essay by Thomas Young in using the concept of surface tension to explain various experimental observations (Pomeau and Villermaux, 2006). A little later, the French mathematician, Pierre Simon Laplace (1749–1827), made a fundamental contribution to understanding capillarity by reasoning that the pressure difference across a liquid-vapor interface is related to the interface curvature (Pomeau and Villermaux, 2006). From these developments and those of others to follow, arose the “capillary rise equation” so important to soil-water physics.

Developments related to the flow of fluids, electricity, and heat need to be mentioned inasmuch as they later form the basis for the equations of water flow in porous media. Jean Leonard Marie Poiseuille (1799–1869) discovered the law on velocity of flow of a liquid through a capillary tube. This equation later became known as the Hagen-Poiseuille Law due to experimental testing by Gotthilf Heinrich Ludwig Hagen (1797–1874). Jean Baptiste Joseph Fourier (1768–1830), a French mathematician, conducted important mathematical work on the theory of heat during the period 1804–1807. The concept of flow of heat in response to a gradient in temperature was a fundamental advancement. Further developments in the flow of electricity by Georg Ohm (1789–1854) and for the diffusion of gases and solutes by Adolf Eugen Fick (1829–1901) resulted in equations similar to the Fourier equation (Kirkham, 2005) and Darcy’s equation to be discussed later. Short bibliographies of Poiseuille, Fick, Ohm, Laplace, Newton, and others are contained in the book by Kirkham (2005).

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**Figure 1** Evolution of the science of soil-water physics and solute transport
Soil-water potential

European scientists attempted to characterize the water regime of soils starting in the early 19th century. The term “water capacity”, an estimate of the amount of water retained in the soil after a rain, was investigated by Schubler (1830), Mayer (1874), and Heinrich (1886). It is generally accepted that Lyman James Briggs (1874–1963) was the first to clearly classify soil water into categories based on how strongly the water was held by the soil. He published a bulletin (Briggs, 1897) where the water in soil was considered to be of three types: gravitational water, capillary water, and hygroscopic water (Philip, 1974). Similar classifications were being proposed in the Russian and German literature (Mitscherlich, 1905; Lebedev, 1919) as discussed by Kutilek and Novak (1997). Although these classifications of soil water were useful means to think about soil water, the categories were arbitrary and artificial and could not be easily quantified.

A fundamental step in understanding how water is held by soil was made by Edgar Buckingham (1867–1940). Buckingham brought a revolutionary concept to the study of soil water. Philip (1974) states the contribution clearly: “Buckingham’s grasp of thermodynamics enabled him to appreciate that, regardless of any qualitative schema of discrete classes of soil water, a continuity of energy states was involved and the whole moisture range was amenable to a unified treatment.” He defined the total potential of soil water which consisted of the gravitational potential and what he called the “capillary potential”, now generally called the matric potential. Buckingham carried out the first measurements of matric potential and presented data on the dependence of the matric potential on soil-water content (Buckingham, 1907). Two additional publications on Buckingham’s contributions to soil-water physics are Nimmo and Landa (2005) and Narasimhan (2005).

An important advance in practically measuring the matric potential with an instrument, to be later known as a tensiometer, was made by Livingston (1908) and later by Gardner et al. (1922), Israelson (1926), and Richards (1942). It is likely that the Russian, Kornev (1924), was not acquainted with the publications of Buckingham and others when he used a term equivalent to suction for the negative pressure of soil water, constructed a “capillarimeter” identical to the tensiometer, and proposed a curve which is today known as a soil-water characteristic curve (Kutilek and Novak, 1997). Haines (1927, 1930) is credited with first recognizing the phenomena of hysteresis.

Soil-water movement

To begin a discussion of soil-water movement, one usually starts with the contributions of Henry Philibert Gaspard Darcy (1803–1858), a civil engineer in Dijon France. His fundamental contribution to the physics of water movement actually occurred near the end of his life with the publication of a book (Darcy, 1856). The porous medium used was saturated sand, and the experiments were conducted by two engineers, Ritter and Baumgarten using wastewater from a hospital in Dijon (Swartzendruber, 2005). In simplest terms, Darcy’s equation stated that the water flux in 1-D flow is directly proportional to the hydraulic gradient and a proportionality constant called the hydraulic conductivity, a composite property of the flowing liquid and the porous medium. Probably independently, Von Weitschowsky (1884) in Germany empirically found the relation now known as Darcy’s law (Kutilek and Novak, 1997). It appears that several decades passed before it was used for theory development in flow through saturated soils. The study of water table heights in drained land using Darcy’s Law was pursued by Jules Dupuit, Philipp Forchheimer, and J. Boussinesq resulting in the Colding equation in 1872 and then the equations developed by Childs in the UK, Houghoudt in the Netherlands, and Kirkham in the USA (Youngs, 2005). Details on the origin of these equations related to soil drainage are given by van der Ploeg et al. (1997, 1999) and Raats and van der Ploeg (2005).
The foundation for the theory of flow of water in unsaturated porous material was again laid by Buckingham (1907). He recognized that water flow in unsaturated soil would be highly dependent upon water content. Buckingham apparently used analogies to Ohm’s law, Fourier’s law, and Hagen-Poiseuille flow through a tube for unsaturated flow, but did not mention Darcy’s law either because he did not know about it (Sposito, 1986) or he did not recognize that his capillary potential was equivalent to a hydraulic head in saturated water flow (Nimmo and Landa, 2005). In either case, he introduced the concept of “conductivity”, dependent upon water content, that today we would call the unsaturated hydraulic conductivity (Philip, 1974). This equation, similar to Darcy’s law, is sometimes referred to as the Buckingham-Darcy equation or simply the Buckingham law (Narasimhan, 2005). Buckingham also went on to define the moisture diffusivity which is the product of the unsaturated hydraulic conductivity and the slope of the soil-water characteristic curve.

It took nearly 2 decades before the full implication and importance of Buckingham’s contributions were recognized. Lorenzo Adolph Richards (1904–1993) applied the continuity equation to Buckingham’s extension of Darcy’s law and obtained a general partial differential equation (Richards, 1931) describing water flow in unsaturated, non-swelling soils with the matric potential as the single dependent variable (Philip, 1974). Childs and Collis-George (1948) recognized the diffusion form of the steady 1-D flow equation and presented data for diffusivity as a function of water content for a sand. Klute (1952) rewrote the Richard’s equation for 3-D unsaturated flow in the diffusion form with the volumetric water content as the dependent variable rather than the matric potential. Since Richard’s equation and Klute’s rewrite are highly non linear, the stage was now set for the next phase of the development of soil-water physics, that is the mathematical solutions to water flow problems.

Some advances in understanding soil-water movement were occurring simultaneously in central and eastern Europe during the early to mid 20th century. For example, Zunker (1930) tried to link soil-water physics to the basic equations of hydraulics, apparently formulated the capillary conductivity based on capillary phenomena, and derived an equation for vertical infiltration which was eventually shown to be identical with that of Green and Ampt (1911) (Kutilek and Novak, 1997). The well known empirical equation of infiltration by Kostiakov (1932) appeared. Although some theory development occurred in central and eastern Europe during the early 20th century, there seemed to be little or no awareness of the developments by Buckingham, Richards, and others and little further development of initial theoretical advances. According to Kutilek and Novak (1997), part of the reason for lack of advance was the emphasis on empiricism.

Mathematical solutions to water flow equations

For saturated porous media, many mathematical solutions were derived for 1-D, 2-D, and radial flow of water to wells, auger holes, agricultural drainage ditches, and tile drains. These solutions to Darcy’s equation and the 2-D form (Laplace’s equation) for various types of boundary conditions, and usually for steady state flow, were started by the development of the Colding equation in 1872. In 1937, Symen Barend Hooghoudt (1901–1953) developed a theory for flow to ditches and drains in shallow, homogeneous and heterogeneous soils (Raats and van der Ploeg, 2005). Hooghoudt and his colleagues were responsible for the design of much of the drainage systems of the Netherlands and the export of their ideas to other parts of the world. Don Kirkham (1908–1998) in about 1945 began in earnest the development of numerous mathematical solutions to drainage problems. Many seepage problems are not of simple shape, so Kirkham developed a new
mathematical method called the modified Gram-Schmidt method to address this problem (Nielsen and van der Ploeg, 2005).

For unsaturated porous media, the quantitative physical theory of water movement has depended heavily on the availability of solutions of the nonlinear Fokker-Planck equation and on the nonlinear diffusion equation. Klute (1952) took the lead by developing a solution to the 1-D form of the diffusion equation for horizontal absorption of water. Philip (1954, 1957) extended the approach of Klute to moisture transfer in the vapour and adsorbed phases with the same mathematical formulation. The contributions of John Philip (1927–1999) to the theoretical development of soil-water physics, infiltration, and evaporation are nicely documented in Smiles (2001, 2005). Parlane (1980) also contributed greatly to theory development. The solutions to the flow equation are highly dependent upon the form of the relationships between the water content, the matric potential (head), and the unsaturated hydraulic conductivity. Two groups of parametric expressions describing hydraulic properties of porous media can be defined (Raats, 2001): (a) a group of equations that can be solved analytically, usually by linearization of Richard’s equation using some kind of transformation; and (b) a group of equations that are more amenable to numerical solutions. Since Richards’ seminal paper in 1931, the number of experimental and theoretical studies on complex water flow processes has increased many fold. For details on the many types of solutions of the non linear Richard’s equation, please see the reviews by Philip (1988) and Raats (2001).

The analysis for unsaturated porous media was also extended to nonisothermal systems by Philip and de Vries (1957). Other researchers including the Miller brothers, Childs, Poulouvasilis, Philip, Mualem, and Topp began to develop models and mathematical formulations for hysteresis. By 1975, finite difference solutions of water flow problems were being developed. Since then, faster computers and efficient finite element (Simunek et al., 1996) and control volume (Heinen and de Willigen, 1998) methods made it possible to solve ever more complicated flow problems (Raats, 2001).

**Coupling solute transport and water flow**

The flow of water through porous media is considered as bulk movement which can be described by the Darcy-Buckingham and Richards equations. However, this description becomes inadequate in defining the movement of transient dissolved solutes and pollutants and their chemical processes (Nielsen and Biggar, 1961). Lawes et al. (1881) observed that water and solutes do not travel uniformly within field soils (Kutilek and Nielsen, 1994). Means and Holmes (1901) recognized the complexity of diffusion and convection processes in transporting solutes after rain and irrigation events. Charles Sumner Slichter (1864–1946) may have been the first to attribute dispersion or spreading of a solute to a distribution of flow velocities within the soil pores (Slichter, 1905). Kitagawa (1934) also demonstrated that lateral dispersion of a salt occurred during water flow through sand resulting in something like a normal distribution. It seems that the next experimental and theoretical developments associated with solute transport occurred in the middle of the 20th century by engineers and petroleum geologists (Danckwerts, 1953; Scheidegger, 1954; Rifai et al., 1956; De Josselin de Jong, 1958; Handy, 1959). Paul Day (1956) using exchange resins showed that hydrodynamic dispersion can occur in ion-exchange processes and compared his results with a statistical model of Scheidegger (1954) whose solution was similar to a solution of the diffusion equation (Day, 1956). Nielsen and Biggar (1961) were likely the first to conduct solute transport (miscible displacement) experiments using soil. Nielsen and Biggar (1962) went on to show that the earlier models based on statistical distributions and/or diffusion-type equations were inadequate to describe solute transport and spreading because not all mechanisms were...
considered. They used the analogy of solute moving through a single capillary tube (Bosworth, 1948; Taylor, 1953) and then a differential equation where solute transport occurs due to molecular diffusion and mass flow of the water of the type used by Lapidus and Amundson (1952). This equation is now the well known "convective dispersion equation" used in most quantitative descriptions of solute transport. As with water flow, the number of experimental and theoretical studies of miscible displacement in porous media has grown enormously since the mid 20th century to include adsorption/desorption processes and irreversible sinks and sources (Selim and Ma, 1998), and miscible displacement of gases (Rolston et al., 1969). Numerous analytical solutions of the convective dispersion equation have been derived for a variety of boundary and initial conditions as well as chemical sorption and degradation processes.

Flow of water and transport of solutes in structured soils has received a lot of attention in the latter part of the 20th century (Raats, 2001). Informative reviews of flow in structured soils are given by van Genuchten et al. (1990), Mermut and Norton (1992), and Selim and Ma (1998).

Conclusions
(a) Science of soil-water physics and contaminant transport began about 150 yrs ago.
(b) The first equation to quantify flow of water was developed by Darcy in 1856.
(c) Major developments for unsaturated media were made by Buckingham in 1907.
(d) Richards made the next big advancement by applying the continuity equation to Buckingham’s equation in 1931.
(e) It was not recognized until latter part of the 19th century that water and solutes do not move uniformly.
(f) Understanding of the complex processes of diffusion, dispersion, convection, adsorption and the mathematical formulations of solute transport started in the middle of the 20th century and continue to the present time.
(g) The problems related to water resources, wastewater management, and environmental quality are becoming increasingly complex. Thus, these complex problems require innovative solutions developed by interdisciplinary teams, the major challenge and opportunity for the 21st century.

References


