TRANSPORT OF MAN-MADE POLLUTANTS IN THE ENVIRONMENT AND THEIR FATE

Bhaskar Nath
Free University of Brussels, Belgium, and European Centre for Pollution Research, London, UK

Keywords: environmental; pollutants; man-made; chemicals; fate; transport; physical; chemical; biological; transformations; advection; Fick's laws; advection-diffusion; advection-dispersion; plug-flow; persistence; half-life; soil permeability; atmospheric; circulation; mixing;

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Summary

For maintaining earth's environmental integrity and protecting human and animal health, it is important to know the fate of pollutants such as man-made chemicals that are being produced and used in ever greater quantities. In other words, we need to know what happens to anthropogenic pollutants over time after they are released into the environment. Common sense tells us that the fate of such pollutants is largely determined by two distinct factors which largely occur simultaneously. First, they are subject to transportation (movement) regardless of which of the three main compartments of the environment — air, water and soil — they are released into. And second, they will normally undergo physical, chemical and/or biological transformations. Clearly, the extent to which they might degrade the environment depends on their biodegradability and whether or not they are physically, chemically or biologically transformed into environmentally harmless or benign substances. These two factors are the subject-matter of this article.

Discussion begins with the different modalities of transport to which the released pollutant may be subject, including advection, advection-diffusion, advection-dispersion, and plug-flow. Simplified mathematical treatment of each of the modalities is given, together with a simple mathematical description of the Fick's Laws of Dispersion concerned with the movement of impurities (pollutants) in a fluid medium which is motionless and chemically inert. Discussion then moves on to the fate of man-made chemical pollutants, including their persistence in air, water and soil; half-life of persistent organic pollutants (POPs); and atmospheric circulation and mixing. The increasingly serious problem of endocrine-disrupting chemicals is also discussed. This is followed by the mathematical explanation of: (a) the pseudo-first-order-reaction that can biologically transform a toxic organic pollutant; and (b) second-order oxidation reaction that can bring about the chemical transformation of a pollutant.

Every care has been taken to make the presentation as simple as possible, especially for beginners, and hopefully they would derive significant benefit from what is presented.

1. Introduction

All three major compartments of the natural environment — air, water and soil — have been and are being relentlessly contaminated and degraded by all kinds of pollutants emanating from mankind's headlong pursuit of economic development through industrialisation. The somewhat dramatic word "Fate" is used to characterise what finally happens to a man-made chemical and to pollutants in general, after its discharge into the natural environment. Is it potentially (or manifestly) capable of harming the environment, humans and wildlife? Does it biodegrade quickly into harmless chemicals? If not, is there a cost-effective strategy with which to neutralise the potential risks it may pose? And so on. Mathematical models that are often used in attempting to predict the mobility of pollutants after having accounted for all the processes to which they may be subject, are generally referred to as "Fate and transport models". And the majority of such processes affecting the fate and transport of pollutants can be described by rate equations to show that the rate of pollutant reduction is a first-order process. The implication of this is that the rate, R, is directly related to the concentration, C_1, of the pollutant as $R = kC_1$ in which k is rate constant. In the case of a second-order process, defined as a process in which pollutant reduction also depends on the concentration, C_2, of a second chemical present in the natural environment, $R = kC_1C_2$. (http://science.jrank.org/pages/ 48019/ pollution-control.html). We are deliberately focusing on man-made chemicals because far too many of them are currently being produced and used, and many of them have serious adverse impacts on the natural environment and health of both humans and animals (Nath, 2008).

Clearly, after a pollutant is released into the environment, it will be affected by various physical, chemical and biological processes and, as a result, it will undergo physical, chemical and biological transformation(s). Physical processes include various modalities of pollutant transport such as advection, diffusion and volatilization for example. Chemical processes include photolysis, hydrolysis, solubility, oxidation and reduction (redox), and hydration, while bio-accumulation, biodegradation and bio-transformation are the primary biological processes.

It is also clear that in order for the natural environment not to be significantly or irreversibly contaminated, the ideal “Fate” of a pollutant would be to quickly biodegrade into simpler, harmless or benign chemicals soon or immediately after its discharge into the environment. Unfortunately, this is not the fate of most of the man-made chemicals, and the chemical properties (including toxicity) of 103,000 or so chemicals that are currently manufactured and used in the European Union illustrate this well (Nath, 2008). An estimated 1,500 of these chemicals are “Substances of very high concern” because they are variously carcinogenic, mutagenic, bio-accumulative, toxic or highly toxic, persistent or highly persistent (Nath, 2008). Many of these ubiquitous chemicals, such as PCBs, DDT and *ortho-N-nonylphenol* for example, are also endocrine-disrupters (also called “hormone mimicking” or “gender-bending” compounds) because they interfere with endocrinal, hormone-controlled bodily functions, including sexuality and reproductive functions of vertebrates including humans with serious implications for generations yet unborn (Colborn *et al*., 1997). Clearly, it would be practically impossible to set standards for such an enormous number of different man-made chemicals and to organise a robust inspection and enforcement regime for their effective control, let alone carry out their fate analysis and life-cycle prediction. In particular, it is as important and probably more important to understand the transport processes of toxins, and therefore the toxicology of pollutants, because of the serious impacts of toxins on living organisms. Not surprisingly, but disturbingly, at present there is little or no safety information in the public domain on by far the vast majority of those chemicals, let alone standards, fate analysis, or toxicology for their effective control (Nath, 2008). This does not augur well for the sustainability of the natural environment or for the organisms, including humans, which rely on environmental integrity and integrity of nature’s life support systems for their well-being and indeed survival.

In some textbooks and environmental literature models of pollutant transport come under the generic umbrella of “Fate analysis”. However, in this article models of pollutant transport, and the processes that determine the fate of pollutants (such as the physical, chemical and biological processes to which pollutants are likely to be subject in the natural environment), are treated separately because, as experience shows, this strategy is more conducive to the learning experience of beginners than would be the case otherwise. Clearly, a reliable and realistic system model must address these processes as appropriate or necessary as well as the appropriate modality (or modalities) of pollution transport in the environment. In the aquatic environment, for example, the main determinants of the fate of chemicals are their reactivity and the rate of physical transport through the environment. In this case, as in others, the usual approach to system modelling is through “mass balance”, and the key elements of modelling are the following (Schnoor, 1996):

a. A clearly defined control volume.
b. Knowledge of inputs and outputs that cross the boundary of the control volume.
c. Knowledge of the transport characteristics within the control volume and across its boundaries.
d. Knowledge of the reaction kinetics within the control volume.

In an article such as this it is not possible to do justice to the subject-matter in terms of scope or depth. Therefore, what is presented is deliberately basic and aimed at interested readers with limited knowledge of the subject. Those wishing to pursue more advanced knowledge of the subject are invited to refer to relevant literature listed under “Bibliography”.

2. Pollutant Transport Phenomena

2.1 Advection

In Environmental Science and Engineering advection is defined as movement which transports the mass of a pollutant, or an impurity entrained in a current, from one place to another (Reible, 1996; Zheng and Bennett, 2002). It is a mode of transport in which a parcel of fluid containing impurities translates along the direction of flow with little or no rotation or turbulence occurring in the flowing fluid. In hydraulics an observable example of advection is the transport of silt downstream by the current of a flowing river. Another familiar example in chemistry and thermodynamics is enthalpy (or any conserved property; see Glossary) which is advected by a fluid such as water or air containing thermal energy. As advection needs current(s) in a flowing fluid to occur, it cannot occur in a solid because a solid material does not flow.

The motion of fluid in advection clearly creates a vector field with components *u*, *v* and w in three dimensions representing velocity components along the *x*, *y* and *z* axes respectively (Bear, 1988; Schnoor, 1996). The movement of silt, caused by the current of a river along the direction of flow, is clearly an example of one-dimensional advection, provided the velocity components at right angles to the direction of flow are negligibly small. However, the concentration of a substance being advected (such as entrained silt or a pollutant) is a scalar quantity.

Sometimes advection is confused with the more comprehensive process of convection; advection is used as a synonym for convection, which it is not. In general, the word “convection” is used to describe a mode of transport in which molecular and eddy diffusion (see Glossary) combine, while the word “advection” is used to describe a mode of transport such as that of silt or a pollutant concentration in a river or along a pipeline. Also, while advective transport is predominantly horizontal, convective transport is mainly vertical, such as vertical mixing that occurs in the boundary layer of earth’s atmosphere for example (Reible, 1999; Vallero, 2007).

To understand the mathematics of advection, consider the movement of mass from ad to *bc* in Figure 1 caused by one-dimensional advective transport along the x-axis. Also, imagine that the mass being transported is that of a chemical pollutant of concentration C which continuously reduces as the fluid, in which the pollutant is entrained, flows along the x-axis. Let A be constant cross-sectional area of the idealised channel, normal to the plane of paper. Then at ad the amount of the pollutant, J_{ad}, being advected per unit of time is given by the equation

$$J_{ad} = Q_{ad} C_{ad} \qquad (1)$$

Here $Q_{ad} = Au_{mean}$ is discharge across ad per unit of time and u_{mean} is constant mean current velocity of the flowing fluid. Similarly, the amount of the pollutant being advected across bc is given by

$$J_{bc} = Q_{bc}C_{bc} \tag{2}$$

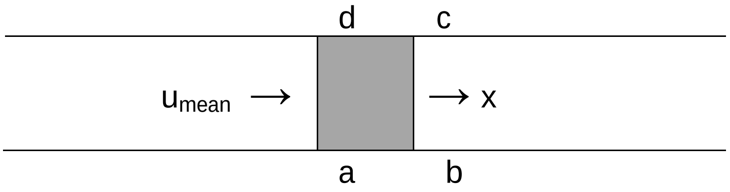

Figure 1 Schematic of advection through an idealised control volume; $ab = dc = \Delta x;\ \Delta x \to 0$.

Then, the amount of the pollutant retained in the control volume $abcd$ during time Δt is given by ($C_{ad} > C_{bc}$)

$$\begin{aligned}\Delta C A \Delta x &= -\left(Q_{bc}C_{bc} - Q_{ad}C_{ad}\right)\Delta t \\ &= -Au_{mean}\left(C_{bc} - C_{ad}\right)\Delta t\end{aligned} \tag{3}$$

Thus, the partial differential equation for advection in one dimension is obtained as

$$\partial C/\partial t = -u_{mean}\left(\partial C/\partial x\right) \tag{4}$$

in which $C(x,t)$) is clearly a function of both space and time. Equation (4) is a mathematical description of advection in one dimension when concentration of the entrained material (e.g. a pollutant or an impurity) is a function of both space and time. The negative sign in equation (3) is because outflow concentration (C_{bc}) of the pollutant being advected is smaller than the inflow concentration, C_{ad}. This sign is not to be used if/when $C_{ad} < C_{bc}$. Also note that the rate of inflow into the control volume of Figure 1 is equal to the rate of outflow from it; that is, $Q_{ad} = Q_{bc} = Au_{mean}$.

In Environmental Science and Engineering it is sometimes necessary to calculate the total mass of the pollutant in question that has passed a given point (or location) over a given time, say T. Let $Q(t)$ denote the discharge passing through the point in unit time, and $C(t)$ denote the concentration of the pollutant entrained in the fluid at that time. Then,

$$\Delta M = Q(t).C(t)\Delta t \tag{5}$$

in which ΔM is the incremental amount of pollutant mass passing through the point during time interval Δt. Clearly, integration of equation (5) between the time-limits of 0 and T gives the total pollutant mass, M, passing through the point during T as

$$M = \int_0^T Q(t).C(t)\,dt \tag{6}$$

Under steady flow conditions Q will normally be constant and not a function of time; as a result equation (6) simplifies to

$$M = \int_0^T C(t)\,dt \tag{7}$$

Exact evaluation of the integral of equation (7) is possible only when $C(t)$ can be expressed as a continuous mathematical function of time. If not, this integral would need to be evaluated numerically; and this can be done very simply by noting that it is equal to the area under the $C(t)$ versus time plot within the time limits of $t = 0$ and $t = T$.

If exceptionally, both $Q(t)$ and $C(t)$ vary with time and if neither can be described by a continuous function of time, then the following numerical method of integration may be employed. Let the time period of interest, T, be divided into a total of N small but equal time intervals of Δt; that is, $T = N\Delta t$. Also, let the measured values of $Q(t)$ and $C(t)$ at the i-th. time interval be Q_i and C_i respectively ($i = 1, 2, 3, \ldots N$). Then the numerical version of equation (6) to be applied to calculate M becomes

$$M = \Delta t \sum_{i=1}^{N} Q_i C_i \tag{8}$$

If for any reason the time intervals have to be of unequal duration, then equation (8) will assume the following more general form

$$M = \sum_{i=1}^{N} Q_i C_i \Delta t \tag{9}$$

in which Δt_i is the duration of the i-th. time interval, and

$$T = \sum_{i=1}^{N} \Delta t_i \tag{10}$$

2.2 Fick's Laws of Diffusion

2.2.1 Fick's First Law of Diffusion

In the mathematics of diffusion (Reible, 1999; Books Group, 2010) there are two Laws, called Fick's laws, attributed to Adolf E. Fick (1829–1901). In 1855 he published his First Law of Diffusion which is based on the movement of impurities (e.g. entrained chemicals) through a fluid which is chemically inert and motionless. He demonstrated that whenever a concentration gradient, $\partial C/\partial x$, of the impurities exists in a finite volume of the fluid matrix, the impurities will tend to move. And by doing so they will seek to distribute themselves more evenly within the matrix and thereby decrease the concentration gradient of the impurities. Over a sufficiently long period of time the distribution of the impurities will become homogeneous within the matrix and net flow of the impurities will cease. In other words, as Fick postulated, flux J of the impurities across a given plane is proportional to the concentration gradient of the impurities, $\partial C/\partial x$, across that plane. That is (Reible, 1999)

$$J = -D\left(\partial C/\partial x\right) \tag{11}$$

This is the mathematical statement of Fick's First Law of Diffusion. Here J denotes mass flux caused by molecular diffusion, and D is the diffusion constant of the impurity (e.g. a chemical entrained in a solvent) being diffused. D has dimensions of (L^2T^{-1}). In the literature D is also called the "Diffusion coefficient" or simply "Diffusivity". The negative sign in equation (11) indicates that the entrained material is flowing from higher concentration to lower concentration.

It is interesting to note that equation (11) is analogous to several other equations of Mathematical Physics which govern entirely different physical phenomena, including the following that were developed and postulated by others at roughly the same time as when Fick postulated his First Law of diffusion:

- If J in equation (11) is interpreted as intensity of electric current in a certain direction in an electric field, and $\partial C/\partial x$ and D as electric potential gradient and specific electrical conductivity (reciprocal of specific electrical resistance), respectively, along that direction, then equation (11) becomes the differential form of *Ohm's Law* (Woolfson and Woolfson, 2007)(see Glossary).
- If J in equation (11) is interpreted as specific discharge along a certain direction, say along the x-axis, $\partial C/\partial x$ as pressure gradient forcing water to flow along that direction, and D as saturated hydraulic conductivity along the x-axis, then equation (11) becomes the differential form of *Darcy's Law* (see Glossary). Note that this law is essentially a constitutive relationship expressing conservation of momentum (Cedergren, 1997; Terzaghi, 2010).
- If J is defined as local heat flux density, $\partial C/\partial x$ as local temperature gradient, and D thermal conductivity, then equation (11) becomes the differential form of *Fourier's Law* of heat conduction (Carslaw and Jaeger, 1986). Here "Heat flux density" is defined as the amount of heat energy flowing through a unit area in unit time. The local temperature gradient is negative because heat flows from higher temperature to lower temperature.

2.2.2 Fick's Second Law of Diffusion

An important fact, which Fick's First Law does not acknowledge, is that local concentration of impurities as well as their concentration gradients decrease over time. For example, with reference to Figure 1, J_{ad} is the flux of impurities entering the control volume and J_{bc} the flux leaving it. Then, as matter cannot be created or destroyed, and assuming that no impurities are formed or consumed in the control volume, the difference between J_{ad} and J_{bc} must result in a change in the concentration of the impurities within the control volume. This is indeed the essence of Fick's Second Law of Diffusion, which states that over time the change in the concentration of the impurities is equal to the change in local diffusion flux (Books Group, 2010). The mathematical statement of this law is the following equation

$$\partial C/\partial t = -\partial J/\partial x \qquad (12)$$

in which concentration C of the impurities is a function of both time and space. Substituting for J from Fick's First Law (equation (11)) into equation (12), we obtain the following mathematical statement of Fick's Second Law of Diffusion:

$$\partial C/\partial t = \partial\left(D.\partial C/\partial x\right)/\partial x \qquad (13)$$

The diffusion constant, D, is independent of position when the concentration of the impurities is low. Equation (13) then simplifies to

$$\partial C/\partial t = D.\partial^2 C/\partial^2 x \qquad (14)$$

Clearly, equation (13) or equation (14) may be solved to predict how diffusion causes the concentration of the impurities to change with time.

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Nomenclature

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Biographical Sketch

Professor Bhaskar Nath received his Bachelor's degree in Civil Engineering from the Indian Institute of Technology, Kharagpur, India, in 1960, followed by a Ph.D. degree from the University of Wales, UK, in 1964. In 1983 he was awarded a D.Sc. degree by the University of London for his outstanding original research (according to citation) in numerical mathematics. In 2001 he was awarded the Doctor Honoris Causa (Dr.H.C.) by the University of Chemical Technology and Metallurgy, Sofia, Bulgaria, for his contribution to environmental education.

After having taught at the University of London for more than 27 years, Bhaskar Nath is currently: Director of the European Centre for Pollution Research, London; Executive Director of International Centre for Technical Research, London; Editor of Environment, Development and Sustainability published by Springer; visiting professor to several European universities, and consultant to a number of international companies and organisations. His research interests include Numerical Mathematics, Elasto-Hydrodynamics, Philosophy, Environmental Economics, Sustainable Development, and Environmental Education. He has more than 100 scientific publications in these and related areas including 13 books.