

MOBILITY IN SOIL. AN EXPERIMENTAL APPROACH

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ABSTRACT

Studies of mobility of contaminants in soil are difficult due to the stochastic nature of the inputs and to the spatial and temporal variations of field parameters. Therefore, a systematic approach which combines laboratory and field experiments with computer modelling is proposed for a realistic assessment of groundwater contamination. This should be coupled with validation studies at contaminated sites.

Transport and fate of chemicals in soil is controlled by a number of complex processes. The description of these processes as a function of time, depth, chemical composition and concentration is quite difficult even under controlled conditions. The difficulties become more severe in the natural systems due to the stochastic nature of the inputs and the temporal as well as spatial variations of field parameters. Consequently, most attempts to describe the behaviour of chemical contaminants are based on a compartmental approaches where the behaviour is studied under a fixed set of conditions i.e. single compound in one type of soil under set moisture and temperature regimes.

Even under these conditions, the approaches are only partially successful, thus their applicability in describing the general behaviour of chemical mixtures is very limited. A systematic approach which combines laboratory and field experiments with computer modelling should be undertaken. The approach should include the study of processes which can be classified into two categories : a) processes that lead to loss of contaminants from the soil (sink processes); b) processes that control the movement of contaminants through the soil (transport processes). The first category includes run-off, photolysis, volatilisation/transformation, hydrolysis, oxidation/reduction processes. The second category encompasses advection, diffusion, hydrodynamic dispersion, and adsorption/desorption processes. All of these are time and depth dependent.

During the past several years a considerable interest has been centered on the environmental behaviour of toxic and persistent compounds such as halogenated aromatics, in particular the chlorinated dibenzodioxins, dibenzofurans, biphenyls and benzenes. Many of these compounds, particularly those with high chlorine substitution, are highly hydrophobic, resistant to chemical transformations and microbial degradations.

Earlier studies on the bulk movement of these hydrophobic compounds which combined field and laboratory data with computer modelling have provided interesting results. For instance the 2,3,7,8-TCDD distribution at Seveso (Italy) based on experimental measurements (obtained at a reasonable statistical confidence level up to the maximum depth of 1.5 meters) has been represented by a distribution pattern which has a conical form (1,2). The concentrations obtained at different times are described by the following equation $y = e ax^3 + bx^2 + cx + d$ where :
x = depth, y = concentration

a = cubic term which describes behaviour at greater depth; b = quadratic term which gives its concavity; c = linear term close to the surface; d = gives the extrapolated TCDD contamination at the surface.

The values of parameters a, b, c, d are time dependent and their behaviour has been studied, specially the linear term which represents concentration gradient in the top soil.

The same methodology has also been applied to the experimental data from Times Beach (Mo, USA) (3,4). The concentration and depth relationship has been interpolated. The distribution functions at Times Beach were found to have similar forms as those at Seveso. From soil column-studies (designed to simulate contamination incidence at Times Beach) distribution functions $y = \alpha e \beta x^2$ (α = normalized factor, β = slope) were obtained which describe the distribution of 2,3,7,8 TCDD when applied with waste oil. In this case the investigation of the time dependence of the parameters α and β allowed to determine the time of oil application. It should be noted that these statistical elaborations are based on bulk movement data and did not require specific data on physico-chemical properties of chemicals and soil. On the other hand, provisional models for pollutant migration from top soil to groundwaters need such experimental data. The subsequent sensitivity analysis can indicate the most important parameters to be studied and to be experimentally determined.

The experimental approach

As a first assessment the mobility of a persistent pollutant may be predicted from the soil/water distribution coefficient, K_d . (5,6) Data can be obtained experimentally by batch experiments (7) on each horizon of a selected soil or from an empirical correlation of K_d with other parameters which can be determined readily. In fact for a variety of compounds linear correlations between sorption to soil organic carbon and physico-chemical data have been established (8) e.g. water solubility, octanol-water partition coefficient, melting point, high pressure liquid chromatography capacity constant and gas chromatography retention index. In the absence of experimental data quantitative structure analysis relations (QSAR) can predict K_d . The use of the soil/water distribution coefficient as the only parameter describing the overall chemical and physicochemical interactions between soil and pollutant has severe limitations. As mentioned earlier for the predictive models a large number of experimental data on physical, chemical properties of pollutants and soil hydro-geological properties of the contaminated site are required. Therefore, a comprehensive approach should include the determination of parameter values through batch equilibrium experiments, laboratory column experiments and by small

laboratory lysimeter tests. Afterwards field lysimeter experiments using undisturbed soil columns may be performed. Finally, real in-field pollutant migration experiments should be carried out.

In certain cases column experiments simulate field conditions better than batch methods, even if the rate of movement is generally higher than observed in the real environment. In any case, the soil column technique offers the opportunity for screening different soils. Saturated flow experiments using upstream or downstream conditions and constant concentrations of the pollutant in the inlet solution allow the determination of the distribution coefficient under dynamic conditions. Isotopic tracer techniques (radioactive or stable) can distinguish between natural background content of the soil and the added contaminant. The use of tritiated water or a conservative radioactive compound provides an accurate mean to determine water movement and distribution relative to the contaminant. By applying solid or liquid wastes on the top of the soil columns the leachate migration can be followed by analysing the fractionated outlet and segmenting the column.

In nature, however, pollutant transport from top soil strata to the groundwater occurs mainly under unsaturated conditions. Because it is much easier to simulate saturated flow (positive pressure head) conditions in experimental soil columns than unsaturated flow conditions, experimental devices which can be used to maintain defined unsaturated conditions in laboratory soil columns should be employed. An analysis of unsaturated soil columns can be done by gamma ray attenuation measurements using a dual energy gamma source (^{137}Cs , ^{241}Am) which allows corrections for varying density of swelling or shrinking soils. When the soil in the column is disturbed, a number of gross natural variations of the soil (such as cracks, root channels, etc) cannot be considered and included in the interpretation of the column results.

Laboratory lysimeter experiments employing undisturbed soil columns from the test sites, can give further information on the influence of vegetation, microbiological and bacteriological activity. Use of climate controlled chambers is advantageous. However, undisturbed soil lysimeter experiments normally give varied results due to the large spatial inhomogeneity of soil. Therefore, to obtain more reliable data it is necessary to operate several laboratory lysimeters simultaneously. A more realistic approach is to run undisturbed soil column experiments at the site.

Field experiments are totally site specific but a number of general conclusions can be drawn from these. The selected site first has to be characterized for geological, pedological and climatological parameters. Before starting a field experiment it is reasonable to perform, at least for one season, checks on the selected uncontaminated site. After application of the contaminant in pure form or in real waste material to the surface soil, collection of soil water samples may be started. The samples may be collected contemporaneously by a series of small porous samplers operated by electronic tensiometer controlled vacuum pumps. Samplers are generally made from porous membrane cylinders of teflon (inorganic pollutants), ceramic and/or metal (organic pollutants). These samplers can be placed with a minimum of disturbance into the soil. Depth and distance of the samplers is chosen according to statistical considerations. This type of soil water samplers can normally be employed up to a maximum depth of about 2 meters. For the study of pollutant migration at greater depths bore hole techniques, e.g. the

three well injection-production test, have to be applied. Groundwater is produced from the two outside wells and injected in the center well. For a short period the pollutant, together with chloride or another nonreactive tracer, is then added to the injection stream. The breakthrough of these compounds is then controlled in the two production wells by continuous groundwater sampling and chemical analysis. This procedure shortens the time required for monitoring migration. Natural gradient bore hole tests potentially provide better representation of natural conditions in the aquifer, but they require some years to cover significant distances. Normally these time periods are not acceptable, especially in the case of real contaminated sites where a decision for purification and recovering is depending on the result of the migration studies. In the light of the above discussion it is clear that a comprehensive approach, which encompasses laboratory/field experiments and computer modelling, needs to be undertaken for realistic assessment of groundwater contamination. This should be coupled with validation studies at some contaminated sites.

Such an approach is being proposed through an international collaborative effort.

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