Transport and fate of contaminants in soils: challenges and developments

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Abstract
Human and environmental risk due to contaminants is quantified as the product of hazard and exposure. For many soil and water contaminants, such as heavy metals or organic compounds, the hazards are well known. The challenge is to determine better the exposure pathways. For emerging contaminants, such as nanoparticles, both the toxicity and exposure pathways are unknown. A pollutant's risk can be due either to the continued residency in the soil, or their subsequent fate in receiving waters. Transport of pollutants through soil defies robust prediction currently due to preferential flow processes along macroporous networks, and this is compounded by spatial heterogeneity in the sources of contamination. However, hard-won biophysical knowledge of chemicals moving through soil is increasingly being incorporated into decision support tools to guide risk assessment policies and to assist with risk-based prevention and remediation practices.

Key Words
Risk assessment, modelling, preferential transport, point-source pollution, decision support tools.

Introduction
Contaminants in our soils and waters can detrimentally impact on ecological health and human well being. To assess the risk of such deleterious outcomes, it is necessary to consider the hazard, or toxicity, of the contaminant, along with the likelihood of environmental or personal exposure to the pollutant. Risk is thus quantified as the product of hazard and exposure. For many soil and water-borne contaminants, such as heavy metals or organic compounds, the level toxicity or hazard has been quantified. The challenge is to quantify better the exposure pathways. For emerging contaminants, such as nanoparticles, little is yet known about hazard levels, and so risk assessment is doubly difficult for there needs to be better quantification of both toxicities and exposure pathways. In this presentation we focus, for both traditional and emerging contaminants, on the pathways of exposure which depend on the transport and fate of pollutants in the soil.

We highlight the challenges we currently face in developing the requisite knowledge to enable apt modelling frameworks to be developed for prediction the transport and fate of contaminants in soil. Nonetheless, we show how by using current understanding it is possible to develop decision support tools for risk assessments and for strategies of remediation based on minimising risks to people and the environment.

A prime exposure pathway, either directly or indirectly, for soil-borne contaminants is via transport in the pore-water solution though the structured and chemically reactive medium of our soils. Soils are also home to plant roots and a myriad of floral and faunal denizens. We highlight the issues around preferential flow, and discuss how the spatially heterogeneous nature of point sources of contamination can make modelling difficult. In predicting pollutant transport, we distinguish between whether the fate is in the soil itself, or in receiving waters. The active role of vegetation in controlling the hydraulic impetus for pollutant transport is highlighted, as is its potential role in risk-based prevention and remediation. Next, we present new results on the transport of nanoparticles through soil. Finally, we provide the results of our recent efforts to capture our measurements and modelling in a decision support tool to assess the health risk of consuming vegetables grown in heavy-metal contaminated soil.

Preferential Flow
Soils are three-phase media, characterised by complex porous structures. New measurement technologies are resolving the impact of these structures on the transport and fate processes that control exposure to pollutants (Deurer *et al.* 2008; Gee *et al.* 2009). Increasingly these new measurements and monitoring devices are revealing the prevalence of water repellency and its linkage to transport processes from the soil

surface down to underlying groundwaters.

Hydrophobicity: The surface, and some sub-surface, zones of many soils when they dry out below a critical water content exhibit water repellency such that they do not spontaneously absorb water during infiltration. This can result in a free-water pond on the surface or zones of positive water potential heads deeper in the profile. Free water can then access the macroporous networks, at least until hydrophobicity weakens (Figure 1).

Macroporous networks: Once accessed, soil water and its entrained chemicals are provided a rapid and far-reaching ride past the otherwise reactive surfaces of soil's matrix. This results in preferential transport towards receiving water bodies, with little adsorption or degradation of pollutants (Figure 1).

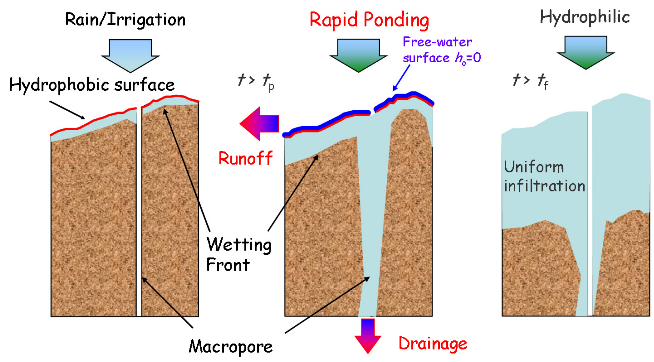

Figure 1. The temporal pattern of wetting and flow in a soil which exhibits transient water repellency (from Clothier *et al.* 2008). During the hydrophobic stage (middle) there is a surface free-water surface that leads to runoff and macropore flow. After repellency wanes (right) wetting occurs through the matrix, depending on the rainfall rate and the soil's conductivity.

Filtering and Buffering - Exogenous and indigenous contaminants: A common pattern of soil wetting and transport shown in Figure 1 has quite different ramifications for pollutant transport. If the contaminant were exogenously incident on the soil surface, then in the middle panel of Figure 1 this would result in deeper-than-expected transport and fate of contaminants. If the pollutant were already indigenous, say mineralised nitrogen, then greater transport would occur under the right-hand conditions of Figure 1.

Point sources and spatial patterns

Contaminants often do not land, or are applied, to soils in a uniform manner. As we show in Figure 2, contaminants need not arrive on, or into the soil, in a homogeneous pattern. For treated posts containing copper, chromium and arsenic (CCA), their point sources of pollution demand special recognition of the spatial configuration of the leakage sources. The dark green spots in Figure 2 are animal urine patches and a risk assessment of their likelihood of contaminating ground and surface waters requires that especial treatment be given to the nitrogen dynamics within those spots. Assuming uniformity leads to erroneous risk assessment.

Fate: Soil or receiving waters?

The assessment of risk needs to consider whether the exposure pathway is via contaminants that become adsorbed and resident in soil, or whether the exposure is through impact on receiving waters, which itself is dependent upon local hydrogeology. These impacts are shown to be quite different, here in the case for CCA leakage from treated support posts in vineyards. The impact of urine spots on groundwaters is also highlighted, and the errors resulting from assuming a uniform distribution is quantified.

Vegetation: Controlling the upper boundary

Soil is not only a reactive physico-chemical medium, it is also hosts plant roots and a teeming microbiological ecosystem. Biological processes, in particular the role of plants in controlling hydraulic processes and the hydrology of soil needs to be closely considered in predicting the fate and transport of contaminants in the rootzone. Further, the biophysical and chemical uptake role of plants needs to be considered as a key part of risk-based assessments of the ability of phytomanagement to control and limit the impacts of soil-resident pollutants.

Figure 2. When assessing the risk of contamination from point sources, such as CCA leakage from fence posts, or nitrate loss from urine spots in pastures, it is necessary to account for the spatial pattern in the landscape of the point sources of contamination.

Nanoparticles

Modern engineering processes have provided a welter of very small-sized particles that are used in a wide range of appliances, cosmetics and devices. These so called nanoparticles are tiny (Figure 3), and their surface area to size ratio is huge such that they could be implicated in disrupting metabolic processes in nature and for humans. Not only is little known about the human health and environmental hazards they pose, there is a dearth of understanding of how they move through soil-plant-faunal systems.

We describe here some preliminary experiments on measuring the transport of fluorescent quantum dots through soils. Such measurements are first needed to provide the knowledge that will be required to develop modelling frameworks for predicting exposures.

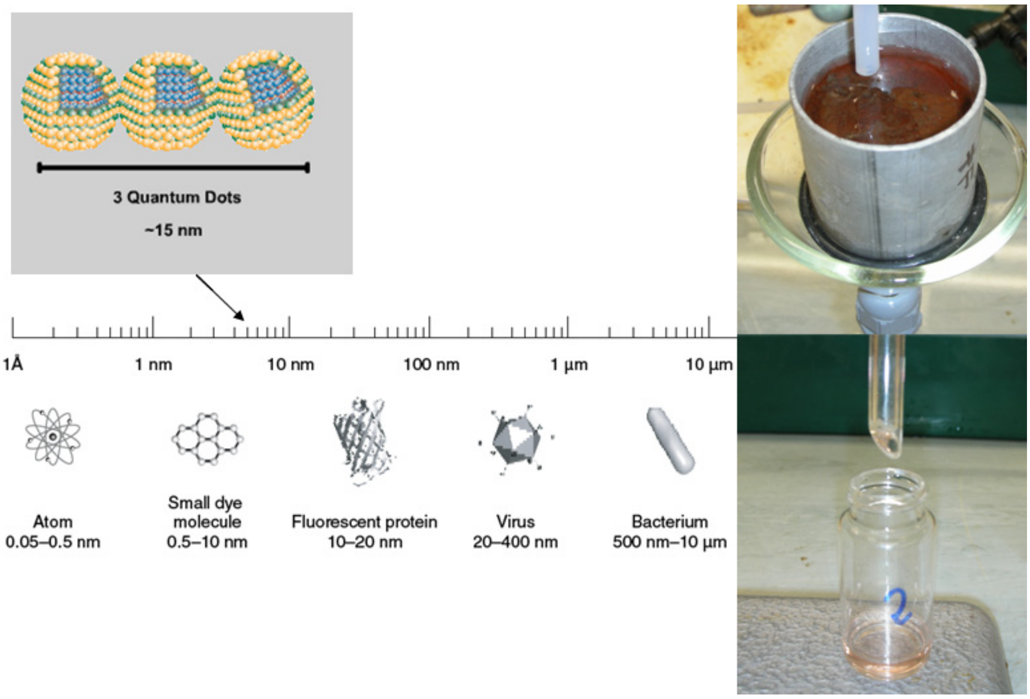

Figure 3. Left. Quantum dots, which fluoresce, comprise a shell and core of different elements. They are very small in relation to other compounds that are likely to be found in soils and plants. Right. One of our simple breakthrough experiments with steady flow through a saturated soil core. Nanoparticles of fluorescent quantum dots of cadmium-tellurium were applied as a pulse in a solution to the soil surface (top) and aliquots of effluent solution (bottom) were then analysed for quantum dots and the conservative bromide tracer.

Decision support tools

Measurements using new devices and novel techniques are shown to be providing confidence for the modelling that is required to develop decision support tools that can be used to guide risk management policies and develop risk-based remedial actions. We demonstrate this for exposure to heavy metals through the consumption of vegetable grown in contaminated soils.

Conclusions

For known contaminants, the challenge is to better understand how risk depends on the exposure pathways resulting from transport and fate processes in the soil. For emerging contaminants, this exigency is heightened by the need to understand better their hazards.

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