

# GROUNDWATER VULNERABILITY TO PESTICIDES: AN OVERVIEW OF APPROACHES AND METHODS OF EVALUATION

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## INTRODUCTION

Groundwater contamination is one of the most important environmental quality concerns. The subsurface migration of hazardous wastes from point sources firmly caught the public's attention in 1977 with the well-publicized Love Canal story. Characteristically, point sources are relatively easy to identify and control. Groundwater contamination from nonpoint sources (NPSs) is a greater environmental problem (1). Nonpoint sources, as a result of contaminants entering the environment over an extensive area, are difficult (at best) or impossible to trace to a specific location and have the potential for maintaining a relatively long active presence in entire ecosystems. Historically, point sources have received the greatest attention because of the conspicuous severity of their impacts. In recent years, public, political, and scientific attention have shifted toward NPSs. This shift reflects an awareness of the scope and potential impact of NPS pollution (2,3). Regulation, monitoring, litigation, and remediation have each helped to control or reduce both point and NPSs of groundwater contamination.

The word *pesticide* is a general term used to refer to the chemicals that are employed to control, destroy, mitigate, prevent, or repel pests (plant or animal). There are several classes of pesticides (e.g., algacides, fungicides, germicides, herbicides, insecticides, miticides, molluscicides, rodenticides, and termiticides). In the United States, the Environmental Protection Agency (EPA) regulates pesticides under the authority of the 1996 amendments to the *Federal Insecticide, Fungicide, and Rodenticide Act* (FIFRA) of 1947. All pesticides must be registered and carry a *label* (providing the rules for application rates and frequencies) approved by the EPA. The distribution and trends of pesticides in the atmosphere (4), groundwater (5), surface water (6), and fluvial sediments and aquatic biota (7) have been carefully assessed. For example, there have been well over 100 monitoring studies in recent years, which quantitatively document the occurrence of pesticides in groundwater (5). The impact of widespread pesticide use, in what Freeze (8) calls *the age of awakening* (i.e., environmental perspective) or *the age of social upheaval* (i.e., new right perspective),

was lucidly brought to the public's attention by Rachel Carson in her 1962 book *Silent Spring* (9), which in large part forced the banning of DDT. Barbash and Resek (5) provide a glossary of 600 pesticides that are of interest relative to groundwater impacts. Table 1 identifies 31 pesticides that have received considerable attention.

In many areas of the world, successful agriculture depends on both pesticides and irrigation. The combination of repeated pesticide applications and irrigation over a large NPS area often leads to leaching through the vadose zone and, subsequently, to groundwater contamination. For example, in the semiarid San Joaquin Valley in California, where groundwater contamination concentrations have been among the highest reported, the amount of active pesticide ingredient applied annually is on the order of 50 million kilograms. A spill of concentrated pesticide at a mixing or distribution site is an example of point source pollution. For example, in April 1977, there was an accidental spill of approximately 1900 liters of the pesticide EDB (see Table 1) within approximately 20 meters of a well that provided drinking water to the village of Kunia on the Hawaiian island of Oahu. The area around the spill eventually became a *Superfund* site.

## VULNERABILITY ASSESSMENT

Assessment involves determination of the change in some constituent over time. This change can be measured in either real time or simulated by a model. Real-time measurements reflect the activities of the past, whereas simulations can provide useful glimpses into the future. Both means of assessment are valuable. The distinct advantage of simulation is that it can be used to alter the occurrence of detrimental conditions before they occur. It should be pointed out that simulation cannot replace *real* data.

By definition, a model integrates existing knowledge into a framework of rules, equations, and relationships to quantify the way a system behaves. As long as a model is applied over the range of conditions from which they it was initially developed, it serves as a useful tool for prognostication. Models can range in complexity from the simplest empirical equation to complex sets of partial differential equations that are solvable only by numerical approximation techniques.

The use of models to aid in the assessment of groundwater vulnerability is now a well-established tool, employed both in industry and by regulatory agencies. The various mechanisms that facilitate the migration of pesticides from the surface/near surface to the water table and the methods used to simulate these processes have been the subject of considerable review (13–27). In general, subsurface flow and pesticide transport are, respectively, estimated by Richards' equation and the advection–dispersion equation, or simplifications. The processes that are important to consider in assessing groundwater vulnerability to

Table 1. Thirty-One Important Pesticides<sup>a</sup>

Common Name	Chemical Name	Use
1,2-Dichloropropane <sup>b,c</sup>	1,2-Dichloropropane	Nematicide
2,4-D <sup>b</sup>	2,4-Dichlorophenoxy acetic acid	Herbicide
2,4-DP <sup>c</sup>	Butoxyethyl ester of ( $\pm$ ) 2-(2,4-dichlorophenoxy)propanoic acid	Herbicide
Alachlor <sup>b</sup>	2-Chloro- <i>N</i> -(2,6-diethylphenyl)- <i>N</i> -(methoxymethyl)acetamide	Herbicide
Aldicarb <sup>b</sup>	2-Methyl-2-(methylthio)-propionaldehyde <i>O</i> -(methylcarbamoyl)oxime	Insecticide
Atrazine <sup>b,c</sup>	2-Chloro-4-ethylamino-6-isopropylamino- <i>S</i> -triazine	Herbicide
Bromacil <sup>b</sup>	5-Bromo-3-( <i>sec</i> -butyl)-6-methyluracil	Herbicide
Carbaryl <sup>b</sup>	1-Naphthyl- <i>N</i> -methylcarbamate	Insecticide
Carbofuran <sup>b</sup>	2,3-Dihydro-2,2-dimethyl-7-benzofuranyl- <i>n</i> -methylcarbamate	Insecticide
Carboxin <sup>b</sup>	5,6-Dihydro-2-methyl-1,4-oxathiin-3-carboxanilide	Fungicide
Chlorothalonil <sup>b</sup>	Tetrachloroisophthalonitrile	Fungicide
Cyanazine <sup>b</sup>	2[[4-Chloro-6(ethylamino)- <i>S</i> -triazin-2-yl]amino]-2-methylpropionitrile	Herbicide
Dalapon <sup>b,d</sup>	2,2 Dichloropropionic acid (sodium salt)	Herbicide
DBCP <sup>c,d</sup>	1,2-Dibromo-3-chloropropane	Nematicide
DCPA <sup>b</sup>	Dimethyl 2,3,5,6-tetrachloro-1,4-benzenedicarboxylate	Herbicide
Diazinon <sup>b,c</sup>	<i>O</i> , <i>O</i> -Diethyl- <i>O</i> -(2-isopropyl-4-methyl-6-pyrimidinyl)phosphorothioate	Insecticide
Dicamba <sup>b,c</sup>	2-Methoxy-3,6-dichlorobenzoic acid	Herbicide
Dinoseb <sup>b,d</sup>	2- <i>sec</i> -Butyl-4,6-dinitrophenol	Herbicide
Diphenamid <sup>b,d</sup>	<i>N,N</i> -Dimethyl- $\alpha$ -phenylbenzeneacetamide	Herbicide
Disulfoton <sup>b</sup>	<i>O,O</i> -Diethyl <i>S</i> -[2-(ethylthio)ethyl]phosphorodithioate	Insecticide
Diuron <sup>b,c</sup>	3-(3,4-Dichlorophenyl)-1,1-dimethylurea	Herbicide
EDB <sup>c,d</sup>	1,2-Dibromoethane	Insecticide
Methomyl <sup>b</sup>	<i>S</i> -Methyl- <i>N</i> -(methylcarbamoyl)oxy)-thioacetamidate	Insecticide
Metolachlor <sup>b</sup>	2-Chloro- <i>N</i> -(2-ethyl-6-methylphenyl)- <i>N</i> -(2-methoxy-1-methylethyl)acetamide	Insecticide
Oxamyl <sup>b</sup>	Methyl 2-(dimethylamino)- <i>N</i> -[[methylamino]carbonyl]oxy]-2-oxoethanimidodithioate	Insecticide
Prometon <sup>c</sup>	6-Methoxy- <i>N,N'</i> -bis(1-methylethyl)-1,3,5-triazine-2,4-diamine	Herbicide
Prometryn <sup>c</sup>	<i>N,N'</i> -Bis(1-Methylethyl)-6-(methylthio)-1,3,5-triazine-2,4-diamine	Herbicide
Propazine <sup>c,d</sup>	6-Chloro- <i>N,N'</i> -bis(1-methylethyl)-1,3,5-triazine-2,4-diamine	Herbicide
Simazine <sup>b,c</sup>	2-Chloro-4,6-bis(ethylamino)- <i>s</i> -triazine	Herbicide
Tebuthiuron <sup>b</sup>	<i>N</i> -[5-(1,1-Dimethyl)-1,3,4-thiadiazol-2-yl]- <i>N,N'</i> -dimethylurea	Herbicide
Trifluralin <sup>b</sup>	2,6-Dinitro- <i>N,N</i> -dipropyl-4-(trifluoromethyl)benzenamine	Herbicide

<sup>a</sup>abstracted from the 341 pesticides in Hornsby et al. (10)

<sup>b</sup>Identified by EPA (11) as high risk for groundwater contamination.

<sup>c</sup>Detected in groundwater in California (12).

<sup>d</sup>Cancelled or severely restricted in the United States.

pesticide contamination are advection, diffusion, dispersion, sorption, transformation, and volatilization (28). The increasing availability of geographic information system (GIS) software to those assessing the potential for NPS groundwater contamination from pesticides has resulted in the generation of regional-scale multicolored vulnerability maps for use in decision management (29).

Three categories of models for assessing groundwater vulnerability from pesticides are (1) regression models, (2) index models, and (3) transient-state solute transport models. Regression models generally use multiple linear regressions to relate various causative factors. Index models, which can be property or process based, compute an index of pesticide mobility. Transient-state, process-based models are capable of simulating the concentration/movement of a pesticide in a dynamic flow system.

One of the first and most promoted regional-scale groundwater vulnerability methods for assessing the impact of pesticides was the DRASTIC index (30). The early process-based simulations by Petach et al. (31) were both elegant and ambitious considering the large scale of the pesticide leaching assessments. During the last two decades, a number of process-based models have

been developed for assessing the behavior of pesticides in the near surface (e.g., PRZM (32), LEACHM (33), GLEAMS (34), HYDRUS-1D (35), LEACHP (36), RZWQM (37), PRZM-2 (38), PELMO (39), MACRO (40), HYDRUS-2D (41), and PESTLA (42)). Table 2 lists selected pesticide leaching assessments for California, Hawaii, and Tenerife.

## UNCERTAINTY

There can be considerable uncertainty in assessments of point and NPS pollution to groundwater from pesticides. Three sources of error inherent in modeling are (1) model error, (2) input error, and (3) parameter error. Model error results in the inability of a model to simulate the given process, even with the correct input and parameter estimates. Input error is the result of errors in the source terms. Input error can arise from measurement, juxtaposition, and/or synchronization errors. Parameter error has two possible connotations. For models requiring calibration, parameter error usually is the result of model parameters that are highly interdependent and not unique. For models with physically based parameters, parameter error results from an inability to represent aerial distributions on the basis of a limited number

**Table 2. Summary of Selected Pesticide Leaching Assessments in California, Hawaii, and Tenerife**

Location	Model Name	Model Category <sup>f</sup>	Reference
San Joaquin Valley, California	PSCLR	R	(43)
	AF <sup>a</sup>	I	(44,45)
	PRZM-2 <sup>b</sup>	TSST	(46)
Oahu, Hawaii	TTF <sup>c</sup>	TSST	(47)
	AF	I	(48–56)
Tenerife, Canary Islands, Spain	PRZM <sup>d</sup>	TSST	(57–60)
	AF	I	(61,62)
Canary Islands, Spain	Li <sup>e</sup>	I	(63)
	AF, PRZM-2	I, TSST	(64)

<sup>a</sup>Attenuation factor (65).

<sup>b</sup>Pesticide root zone model - Version 2 (37).

<sup>c</sup>Type transfer function model (66).

<sup>d</sup>Pesticide root zone model (31).

<sup>e</sup>Leaching index (67).

<sup>f</sup>R: regression model; I: index model; and TSST: transient-state solute transport model.

of point measurements. The aggregation of model error, input error, and parameter error is the total (or simulation) error. For multiple-process and comprehensive models, simulation error is complicated further by the propagation of error between model components.

The methods for characterizing uncertainty can, in general, be grouped into three categories (68): (1) first-order analysis, (2) sensitivity analysis, and (3) Monte Carlo analysis. First-order analysis is a simple technique for quantifying the propagation of uncertainty from input parameter to model output. Sensitivity analysis is used to measure the impact that changing one factor has on another. Monte Carlo analysis is a stochastic technique for characterizing the uncertainty in complex hydrologic response model simulations. Loague and Corwin (68) provide examples of first-order uncertainty analysis, sensitivity analysis, and Monte Carlo simulation. Note that most of the groundwater vulnerability assessments listed in Table 2 were designed to identify the uncertainty in the estimates relative to the uncertainty in the chemical, climate, and soil databases.

## FRESNO CASE STUDY, AN EXAMPLE

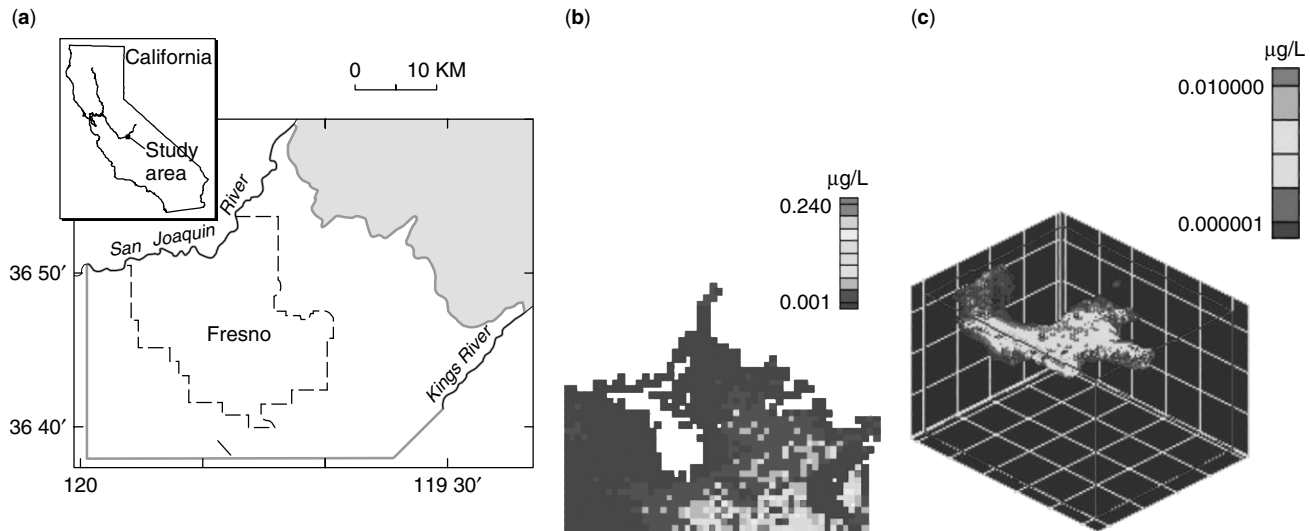
Between the late 1950s and the time of its statewide cancellation in August of 1977, there was widespread use of DBCP (see Table 1) throughout the San Joaquin Valley in California. More than two decades after its cancellation, DBCP-contaminated groundwater persisted as a problem in the San Joaquin Valley. The objective of the Fresno case study (46,69) was to address, from a simulation perspective, if *label recommended* NPS applications were likely to be the principal source of the DBCP groundwater contamination in Fresno County (note that DBCP was a legacy when the study was conducted). The relatively unique characteristic of the Fresno case study is that both the unsaturated near surface and the saturated subsurface were considered for a 2184 km<sup>2</sup> area.

The numerical model used for 1-D simulations (46) of dissolved phase DBCP concentration profiles in the unsaturated zone was PRZM-2 (37). The potential fate and transport of DBCP between the surface and the water table for multiple NPS applications were quantitatively estimated from 1172 separate 35-year simulations. The aggregate of the DBCP concentrations loaded to the water table made up the annual loading files for the 3-D saturated transient transport simulations. The numerical models used for the 3-D simulations (69) of saturated subsurface fluid flow and DBCP transport, for the same 35-year period, are MODFLOW (70) and MT3D (71), respectively. A 76,440-element mesh was used for the saturated simulations.

The simulation results from the Fresno case study lead to several conclusions (46,69): (1) the areas most likely to facilitate DBCP leaching through the entire unsaturated soil profile were targeted; (2) the first appearance of DBCP above the detectable limit at the water table was simulated as most likely to have occurred between 1961 and 1965; (3) the estimated DBCP concentrations reaching the saturated subsurface exceed the maximum contaminant level (MCL) at several locations at different times; (4) the first appearance above the MCL was between 1965 and 1970 (note that by 1990, the concentrations are below the MCL); (5) relative to the size of the study area, the extent and duration of the estimated DBCP contamination was small; (6) DBCP concentrations are a function of spatial and temporal variations in the application rates, the application frequency, the unsaturated profile thickness, the soil-hydraulic properties, and the near-surface sorption; and (7) the DBCP plume evolves (grows and retracts) with time due to the loading rates at the water table. Figure 1 shows 1971 snapshots of the simulated DBCP loading to the water table and the 3-D DBCP plume in the saturated subsurface. Note that the Fresno case study assessments were critically evaluated and extended (72,73).

## EPILOGUE

The ethics, efficacy, and economics of widespread pesticide applications have all received considerable attention in recent years. Without question, it is important to assess the vulnerability of groundwater to contamination from widespread applications of pesticides. For more than 10 years, the U.S. Geological Survey's National Water-Quality Assessment (NAWQA) program has played a key role in this assessment; more than 50 major river basins and aquifers were studied (74). The overall picture emerging from the NAWQA program is that pesticides typically occur at low levels, punctuated by seasonal pulses in concentration (75). Specific to groundwater, pesticides were most frequently found in shallow systems in urban and agricultural areas (74). The patterns of pesticide detection (over space and in time) identify problem areas and chemicals, warn of potential problems, and provide valuable feedback on to improve conditions (75). The use of mathematical models for assessing groundwater vulnerability from pesticides, especially after ground truth comparisons, fills in information gaps, identifies critical



**Figure 1.** Results from the Fresno case study. (a) Location of study area. (b) Plan view snapshot of the simulated DBCP concentration loading at the water table for 1971 (46). (c) 3-D snapshot of the simulated DBCP concentration plume for 1971 (69).

areas and chemicals for future monitoring, and provides the “what if” capability needed for both regulation and remediation.

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