REDUCING THE POTENTIAL FOR ENVIRONMENTAL CONTAMINATION BY PESTICIDES AND OTHER ORGANIC CHEMICALS (W-82)

TABLE OF CONTENTS	PAGE
PROJECT NUMBER	2
TITLE	2
DURATION	2
STATEMENT OF PROBLEM	2
JUSTIFICATION	2
RELATED CURRENT AND PREVIOUS WORK	4
OBJECTIVES	9
PROCEDURES	11
EXPECTED OUTCOMES	16
ORGANIZATION	17
SIGNATURES	17
REFERENCES	18
APPENDIX A: ATTACHMENTS	23
PROJECT LEADERS	24
RESOURCE LISTING	26
CRITICAL REVIEW	28

ORIGINIAL APPENDIX D OF PARTICIPANTS

WESTERN REGIONAL RESEARCH PROJECT W-82

PROJECT NUMBER: W-82

TITLE: REDUCING THE POTENTIAL FOR ENVIRONMENTAL CONTAMINATION BY PESTICIDES AND OTHER ORGANIC CHEMICALS

PROJECT NUMBER: W-82

DURATION: OCTOBER 1, 2000 – SEPTEMBER 30, 2005

ADMINSTRATIVE ADVIORS: Robert Heil and Ray Knighton

STATEMENT OF PROBLEM:

Agricultural activity while essential in meeting the dietary needs of the world can result in the contamination of soil, air, and water resources. Identifying and adequately quantifying the chemical and biological processes that control the behavior of organic chemicals in the environment is imperative towards improving pesticide management, minimizing contamination of our natural resources, and remediating contaminated environments.

JUSTIFICATION:

Use of organic chemicals by all sectors of the economy has resulted in widespread contamination of soil, air, and water. Because chemical technology is generally recognized as having great societal benefit compared to the risks, it is critical that approaches and tools for managing chemical use are developed in order to minimize future contamination and to remediate contaminated environments. Evaluating and quantifying the environmental behavior of organic chemicals is vital towards the development of sound strategies for ensuring sustainable agriculture and protecting natural resource systems.

A major societal concern regarding chemical contamination is pesticide residues in our environment and food. Pesticide use will continue to decline as a result of improved integrated pest management (IPM) systems and development of sustainable, integrated farming systems. Nevertheless, use of pesticides will remain a mainstay within production systems, especially for the many crops for which IPM systems have not been developed. Research is needed on identifying efficient pesticide use practices with minimal environmental impacts. Newly developed pesticides, many of which are applied at rates one-tenth or less of conventional pesticides, are often highly toxic to nontarget crops or aquatic organisms; thus, knowledge is needed of the transport and fate of these substances.

In addition to pesticides, the environmental fate of pharmaceutically active substances is a rising concern. Pharmaceutical substances, which are designed to cause a biological effect and are usually organic in nature, have a high potential for being a major player in endocrine

disruption. Almost half of the fifty million pounds of antibiotics produced annually in the United States is used for agriculture, with the majority being used as feed additives for growth promotion. With increases in the intensive use of antibiotics in concentrated animal feeding operations and land application of manure, there are concerns that excreted pharmaceuticals will migrate in the environment with potential impacts on water supplies and the production of antibiotic-resistant microbial populations. Recent research has shown a high percentage of multiple-antibiotic resistant microbial populations in litter from broiler houses. In large animal feeding operations antibiotics could be highly concentrated in animal wastes and associated lagoons. Little effort to date has been put forth with regards to the potential of these organic substances impacting ground and surface waters, their impact on soil microbial communities, and their overall persistence in the environment. A better understanding of the environmental fate of antibiotics is needed to assess the environmental risk involved in exposing animal wastes to the environment.

Organic chemicals not necessarily identified with the agricultural industry are also of interest. For example, petroleum products or "inert" materials (e.g. solvents, emulsifiers) are present in pesticide formulations and are commonly used on farms. Also, as areas of urban and rural land use increasingly overlap, these often conflicting communities impact one another through their chemical use patterns. The principles governing the behavior of organic chemicals in the environment are the same regardless of how these chemicals are used. Therefore, both urban and rural sectors of the economy can benefit from basic and applied studies of all organic chemicals whether they are used in agro-ecosystems or are associated with hazardous waste sites. Over the last several decades, the agricultural science community has conducted most of the research on the environmental behavior of organic chemicals regardless of their source. Much of the current research on the environmental behavior of industrial chemicals is dependent on basic principles originally derived in the study of pesticides, some of which resulted from research by W-82 members.

The fate and accumulation of organic compound residues are mediated by various, often tightly coupled, processes including advection and diffusion, sorption and desorption, biodegradation, and chemical reactions. These processes occur within and between intimately associated environmental compartments (soil, water, air, and biota). For example, chemical interactions with the soil controls chemical persistence, release into water and air, and bioavailability, which impact pesticide efficacy, degradation, and off-site transport. Because of the complexity of interactions among chemical properties, environmental compartments, and spatial and temporal scales, research involving environmental pollution requires a multi-disciplinary approach. A unique strength of the W-82 committee is the close research collaboration among its members from different scientific disciplines who, outside the committee, do not have sufficient opportunity to communicate with one another. W-82 provides a forum where specialists in mechanisms of chemical behavior, microbial ecology, transport behavior, mathematical modeling, and field assessment techniques can come together, and exchange information gleaned from individual or cooperative research efforts. Such cooperative efforts between research groups representing different expertise and diverse geographical areas are imperative developing appropriate management techniques for minimizing environmental contamination and thus risk.

The goal of W-82 research is to minimize environmental contamination from pesticides and other organic chemicals. W-82 proposes to carry out a cooperative and interdependent research program that elucidates fundamental mechanisms of chemical behavior and applies this

knowledge to different spatial and temporal scales. In combination with mechanistic models, the principles of chemical behavior will be integrated to form management models and propose and assess best management techniques. Research carried out by W-82 will be useful in the continued development of best management practices for minimizing environmental contamination, as well as, the development of efficient and comparatively inexpensive strategies for remediating contaminated environments. Results of the research proposed by W-82 will be applicable to agro-ecosystems in addition to urban systems, and will fulfill USDA needs with regards to protection of water quality and development of integrated agricultural systems as well as the USDA mission for serving the dominant urban sector of our economy. By developing better management techniques the risks of adverse environmental and health effects of our chemical technology will be greatly minimized.

RELATED, CURRENT AND PREVIOUS RESEARCH:

Interfacial Transfer Processes

Our understanding of interfacial transfer processes impacting the distribution of organic chemicals in the environment has progressed greatly over the past two decades. Interfacial transfer includes partitioning between air, water, soil particles, and microorganisms. Sorption by organic matter (humus) and clay minerals are key processes affecting the mobility of pesticides and other chemicals in soils (Meyer and Thurman, 1996; Koskinen and Clay, 1997). Organic matter can be bound to minerals, and humus-coated minerals can play a very important role as sorbents (Kordel et al., 1997; Celis et al., 1997, 1998; 1999a,b; Cox et al., 1998). The interactions of chemicals with sorbed and in some cases dissolved (or colloidal) humus largely determine their ability to move through the soil profile (Meyer and Thurman, 1996; Koskinen and Clay, 1997; Seta and Karathanasis, 1997; Chien et al., 1997). Research is needed that focuses on the interactions of chemicals with such materials. Despite decades of research on these interactions, there are a number of issues that remain unresolved (Pignatello, 2000). These include: the mechanism of sorption by natural organic matter and organoclays, hysteresis, prediction of sorption/desorption kinetics, the relationship between sorption/desorption and bioavailability, and the influence of adjuvants in pesticide formulations on pesticide transport.

Sorption may be affected by the residence time in soil (Cox et al., 1998; McCall and Agin, 1985; Pignatello and Huang, 1991: Shelton and Parkin, 1991). Hysteresis—the asymmetry in the sorption isotherm between the forward (uptake) and reverse (desorption) directions—is a vexing problem that has not been satisfactorily explained. There is a need to account for sorption hysteresis in a quantitative way and to understand it mechanistically in order to improve our ability to predict leaching from aged residues. Partitioning of pesticides between particles and water may not be the same in field moist soils than traditional batch slurry systems; as a consequence, the obtained batch sorption parameters may not adequately reflect sorption processes in field moist or unsaturated soil (Celis and Koskinen, 1999; Hance, 1977; Rochette and Koskinen, 1996; Rochette and Koskinen, 1998).

Recently there has been a great deal of interest in sorption/desorption kinetics in relation to biological availability of contaminants in soils (Pignatello and Xing, 1996; Luthy, et al. 1997; Lee et al., 1998). It is known that intraparticle sorption processes reach equilibrium only slowly; that exhaustive water extraction leaves behind an appreciable fraction that strongly resists further desorption; and that the resistant fraction increases with time as the soil-chemical matrix ages. Evidence has emerged linking this “resistant fraction” to reduced leachability; non-ideal

transport in groundwater; reduced bioavailability to bacteria, ecological receptors and humans; and incomplete remediation at polluted sites. Since bioavailability is strongly dependent on desorption rates, and since remediation costs rise exponentially with the degree of removal, studies are critically needed to understand the underlying causes of resistant desorption. In particular, the interaction of organic chemicals with soil organic matter is proving to be more complex than previously realized. Studies are needed to understand this interaction at a fundamental level. Similarly, it has been observed that rainfall soon after pesticide application (i.e., prior to attainment of equilibrium) results in elevated pesticide concentrations in ground and surface water samples, i.e. a flush effect (Wauchope 1987; Wauchope et al. 1990; Dravillas 1993; Dowdy et al., 1998). Many attempts have been made to model pesticide transport and to quantify the flush effect, but results have been less than adequate.

Humus-coated clays and other model sorbents are useful models for studying the complex sorption sites for pesticides and other organics in soils with varying pH and ionic strength conditions (Celis et al., 1997, 1998). Organo-clay formulations have also been used as a means of plume containment at contaminated sites, i.e., and more recently, formulations were developed to reduce alachlor and metolachlor leaching (Polubesova et al., 1997; El-Nahhal et al., 1998, 1999). These formulations were based on montmorillonite preadsorbed with the synthetic organic cations benzyltrimethylammonium (BTMA) and phenyltrimethylammonium (PTMA) to change the clay mineral surface from hydrophilic to hydrophobic, resulting in effective adsorption of hydrophobic herbicides (Xu and Boyd, 1995), thus reducing their soil solution concentration and extending their herbicidal activity. Optimized organo-clay formulations demonstrated slow release of the herbicides to the soil solution, thus restricting herbicide activity to near the top of the soil profile as measured by bioassay (El-Nahhal et al., 1998, 1999). Similar results may be found for herbicides capable of hydrophobic interactions with humus-coated minerals. Additional research with organo-clays is needed to evaluate and optimize the use of such formulations as a means to modulate pesticide release and serve in-situ plume containment strategies, and as model sorbents to improve our ability to predict complex sorption processes.

The use of pesticdes in wetland ecoysytems such as those used in the production of cranberries can be problematic because of the strong likilhood that water flowing through the system will become contaminated. Weed control relies on the use of early season herbicides applied to the soil prior to cranberry shoot growth. Growers have reported efficacy problems with napropamide (Devrinol) as evidenced by the high rates of application required for control. Dichlobenil (Casoron) has performed very unpredictably; in some cases it has caused phytotoxicity to vines and in other situations it has failed to adequately control dodder. Herbicide efficacy can also be greatly affected by adsorption of the active ingredient to soil organic matter (OM), volatilization, and rate of degradation. Early work with dichlobenil indicated that bioactivity was affected by binding with organic matter (OM), which may have also reduced degradation (Miller et al. 1966). Leaving the chemical on the surface without incorporation augmented volatilization; irrigating following application to achieve some incorporation prolonged persistence (Miller et al. 1967). Commonly used pesticides in cranberries, including napropamide, 2,4-D, and carbofuran, have been shown to rapidly degrade upon repeatedly applying the product to soil (Walker and Welch 1991), which has been shown to adversely affect pest control efficacy (Felsot 1989; Wilson and Felsot 1997). In soil from a nearby bog without napropamide use but managed by the same grower, only 10% of the napropamide dissipated. In contrast to napropamide, multiple applications of dichlobenil did not affect degradation rate, which suggests either chemical degradation or volatilization as major loss mechanisms (Felsot

and Patten 1997). Under drying conditions, sorption would increase thereby reducing bioavailability while under flooded conditions, bioavailability would be enhanced by desorption, thus enhancing the probability of uptake by cranberry vines with consequent phytotoxicity. Furthermore, growers periodically put a layer of sand on the bog, which may periodically decrease dichlobenil sorption potential, thereby increasing its volatilization and/or bioavailability. Effective pest management in cranberry requires a better understanding of soil-herbicide interactions in wetland-like systems. An understanding of these interactions will allow better predictions of bioavailability, a key determinant of efficacy as well as likelihood for environmental contamination.

Volatilization of pesticides may contribute significantly to air pollution in regions with intensive agricultural production. For instance, a multiple-year monitoring study conducted by the California Air Resource Board showed high concentrations of pesticides, especially soil fumigants, in ambient air near application sites (Baker et al., 1996). Because some soil fumigants are acutely toxic or carcinogenic, excessive emissions may impose hazardous effects on field workers and nearby residents. Thus, it is clearly important to understand fumigant volatilization after application and develop mitigation measures minimize their emissions. The maximum amount of pesticide losses to the atmosphere is determined by the Henry's law constant, i.e., the air-water partition coefficient, and the rate of mass transfer from the air-water, soil-air interface, or through liners used in fumigant application. Henry's law behavior is well known in simple aqueous systems; however, studies are needed to predict air-water partitioning in complex aqueous solutions that exist in soils, estuarine, and fresh-water systems, especially the influence of "dissolved" and colloidal organic matter (Baker et al., 1996; Breiter et al., 1998). Currently, plastic films are used to tarp the soil surface following soil fumigation; however, these films are relatively permeable to fumigant vapors, allowing a large proportion of the applied mass to escape to the atmosphere. Films with very low permeability are being developed in hopes that with more fumigant containment, fumigant application rates can be decreased and still result in adequate efficacy. The permeability of plastic films must be determined under a variety of environmental conditions to develop management practices that minimize fumigant flux to the atmosphere.

Microbial Degradation & Microbial Communities

Our ability to investigate the soil microbial populations involved in biodegradation has been enhanced with the expansion of molecular tools for describing, and in some cases quantifying, specific microbial populations and communities. A clear understanding of the fate of organic contaminants in the environment depends on an understanding of the biodegradation pathways by which these compounds are transformed, and an understanding of the mechanisms by which genes encoding their metabolism are spread throughout a soil community. Genetic exchange and gene recruitment are likely to be important mechanisms in development of novel pathways for metabolism of newly introduced compounds, such as pesticides. Little is known about how genes encoding pesticide degradation spread among soil bacteria, or of the impact that genetic exchange has on the development of enhanced biodegradation.

Despite numerous years of study, little information is available concerning microbial population and community structure involved in the transformation of pesticides and organic chemicals in soil. Relatively few microorganisms that degrade pollutants have been isolated and characterized. For example, virtually all that is known of the biochemistry and genetics of

bacterial naphthalene metabolism was gained from analysis of a limited number of microorganisms, such as *Pseudomonas sp.* strains, but attention is now turning to metabolism by more diverse gram positive organisms, such as *Rhodococcus* species. *Rhodococcus* strains metabolize a range of aromatic hydrocarbons and chlorinated aromatic compounds (Kastner et al., 1994; Finnerty, 1992; Grund et al., 1992; Uz et al., 1999); however, little is known of the pathways or genetics of metabolism by *Rhodococcus* strains. A similar situation exits for the pesticide atrazine, where despite several years of research there is little information is available concerning the genes involved in the metabolism of atrazine and other *s*-triazine compounds (Nagy et al., 1995; Shao and Behki, 1995). In a model system developed to study atrazine biodegradation, the first three genes involved in atrazine degradation in *Pseudomonas* sp. strain ADP have been cloned, sequenced and expressed (de Souza, et al.,1996; 1997; Mandelbaum et al., 1995; Chakrabarty, 1996). We have shown that atrazine degradation genes are plasmid encoded in strain ADP and have investigated the distribution and transfer of these genes in other atrazine degrading bacteria on a global scale (MN). Similar work is being performed with carbofuran as well (FL). Use of genomics will allow us to gain a better understanding of the way in which genes and biodegradation pathways are assembled on plasmids to degrade new anthropogenic compounds.

It is often assumed that the solution phase of a sorbed chemical can be biodegraded by microorganisms. However, there is a significant diversity in the capabilities of microorganisms to degrade contaminants partitioned in, or sorbed by natural organic matter (NOM) (Guerin and Boyd, 1997; Tang et al., 1998). Physiological explanations for such diversity may include variations in attachment and adhesion to NOM surfaces (Harms and Zehnder, 1994; Holm et al., 1992; Neu, 1996), production of biosurfactants (Herman et al., 1997; Noordman et al., 1998), motility, rates of membrane transport, and/or variation in metabolic pathways or enzyme expression leading to variations in mineralization kinetic parameters (Stringfellow and Aitken, 1994). Enrichment strategies in which contaminants are sorbed to organic solid phases may provide a more relevant selection environment, and may eventually lead to a better understanding of the involvement of microorganisms and microbial processes in the mineralization of contaminants in soils.

Soil microbial communities are extraordinarily diverse and it is unknown what proportion of the community is actively engaged in degradation processes in soil. Analysis of phospholipid fatty acids (PLFA) can be used to quantitatively describe and "fingerprint" the whole soil community. (Kerger et al., 1986; Dowling et al., 1986, Ringelberg et al, 1989, Olsson et al., 1995, LeChevalier, 1977). Community level, PLFA analysis can distinguish changes in microbial biomass and community structure in soils polluted with heavy metals and organic pollutants (Pennanen et al., 1996, Smith et al., 1986). Recently, ^{13}C-labeled PLFAs have been used resulting from incorporation of stable isotope-labeled C substrates into soil, to provide a way to define the groups of organisms utilizing those substrates (CA).

Soil fumigants are degraded in soil both chemically and microbially. It was reported that in soils that had received repeated applications of fumigants, fumigant degradation was significantly enhanced (Smelt et al., 1989). Understanding biodegradation of fumigants will not only improve our knowledge on the environmental fate of soil fumigants, but may also lead to isolation and identification of fumigant degraders. Application of such biodegraders may, ideally, be used to reduce fumigant contamination of air and/or groundwater. Our early study also showed that amendment of some organic wastes in soil resulted in drastically enhanced fumigant degradation, and the enhancement was mainly due to stimulated microbial degradation

(Gan et al., 1998a). It is important to screen and identify organic amendments that are capable of degrading fumigants. Such amendments may be applied in practice for reducing fumigant emissions.

Antibiotics are widely used in animal feeding operations and can be expected to appear in animal wastes. Pharmaceutical substances such as antibiotics are designed to cause a biological effect, and have a high potential for being a major player in endocrine disruption; however, little effort has been put forth with regards to their environmental fate (Halling-Sørensen et al., 1998). In the past few years, endocrine disruption has become one of EPA's priority research areas (Kavlock, 1999). The majority of research to date has focused on fate of industrial chemicals and their role in endocrine disruption. However, almost half of the fifty million pounds of antibiotics produced annually in the United States is used for agriculture, with the majority of this used as feed additives for growth promotion (Wade and Barkley, 1991). Antibiotics fed to animals are either metabolized or excreted unchanged in the urine and feces (Chiu et al., 1990; Magnussen et al., 1991). In large animal feeding operations antibiotics could be highly concentrated in animal wastes and associated lagoons. In addition, spreading manure on land as both a fertilizer and a disposal strategy is commonly practiced in animal feeding operations (USDA, 1996). With increases in the intensive use of antibiotics in concentrated animal feeding operations and land application of manure, there is concern that excreted pharmaceuticals will migrate in the environment with potential impacts on water supplies and the production of antibiotic-resistant microbial populations. A better understanding the environmental fate of antibiotics from animal waste application, survival and development of antibiotic-resistant microbial population in the environment, and antibiotic-resistance gene transfer among microbial populations are important to assess the environmental risk involved in exposing animal waste to the environment.

Abiotic Transformation Processes

Almost nothing is known about the role of Mn (IV) oxides in the oxidation of herbicides and other pesticides commonly applied to soils (Wolfe et al., 1990). The prevalent view is that pesticide oxidation in natural soils is usually enzyme-catalyzed, not mineral-surface catalyzed (Bollag and Liu, 1990). However, Lehman et al. (1987) and Lehman and Cheng (1988) have demonstrated that phenolic acids are rapidly oxidized abiotically in natural soils in the presence of Mn oxides. Li and Lee (1999) and Li et al. (2000) have demonstrated the reduction of Mn(III) to Mn(II) during transformation of organic amines in sterile soils. A more comprehensive characterization of the abiotic MnO_2-catalyzed degradation of atrazine demonstrating that dealkylation proceeds at the same rate with or without O_2 and involves no net redox .

Volatilization loss of a fumigant can be reduced if its degradation in soil is accelerated. This has been demonstrated in laboratory column and field micro-plot experiments. Gan et al. (1998b,e) recently discovered that thiosulfate salts can rapidly react with chlorinated compounds including methyl bromide, 1,3-dichloropropene, chloropicrin, methyl iodide and propargyl bromide, and transform these compounds into non-volatile, less toxic ionic species. Because common thiosulfate salts (e.g., ammonium thiosulfate and potassium thiosulfate) are commercial fertilizers, this approach is highly feasible for field application and warrants more study.

CRIS Review of Regional Project Overlap

The proposed research for this regional project combines the study of processes controlling pesticide movement in soil, water, and air with the goal of model development and of providing stakeholders with environmentally sound decision-making strategies that optimize the management of pesticides. A search of currently funded CRIS projects revealed that there are several projects that address pieces of the proposed work, especially the study of transport mechanisms in soil; however, there are no existing projects that encompass all the aspects of the research included in this project.

Interestingly, several of the overlapping projects in the area of transport mechanisms have a W-82 member as the primary investigator. For example, Dr. S. Yates and Dr. J. Gan lead three out of the four existing projects focused on volatilization processes. Dr. J. Pignatello and Dr. W. Inskeep are lead scientists in two out of the six projects on sorption/desorption kinetics. There are also currently three projects that address pesticide movement in soil using a traditional hydrologic approach and soil column leach tests. Additionally, Dr. L. Lee leads one of the two projects on abiotic processes controlling mobility and bioavailability of pesticides in soil. Therefore this regional project team includes a number of outstanding scientists in the area of pesticide transport.

In the area of bioavailability and biotransformation of organic chemical in soil through the study of microbial ecology, most of the projects are focused on remediation of chlorinated and petroleum-based contaminants and not pesticides. And in the area of model predictions of pesticide fate, most funded projects are utilizing existing models and are not developing new models as proposed here.

As a regional project, W-82 has been complementary in focus to the W-45 regional project that is also undergoing renewal. The new project proposal for W-45 is different than W-82 in that its objectives are to 1) identify, develop, and/or validate trace residue analytical methods, immunological procedures, and biomarkers, 2) characterize abiotic and biotic reaction mechanisms, transformation rates, and fate in agricultural and natural ecosystems, 3) determine adverse impacts from agrochemical exposure to cells, organisms, and ecosystems. Therefore, the W-45 project proposal will be more fundamental in nature with an emphasis on a molecular level processes and exposure rates as compared to the W-82 project proposal.

OBJECTIVES:

Objective 1: Characterize and quantify the basic chemical and biological processes controlling the behavior of pesticides, other organic chemicals, and microorganisms in soil, water, and air.

It is the basic processes of sorption/desorption, transformation, transport, volatilization, and plant uptake that determine the potential for an organic substance to contaminate ground and surface water. Members of the W-82 technical committee have made significant contributions towards elucidating these processes and towards developing methodologies for studying these processes. However, a lack of understanding of the fundamental processes affecting organic compounds in soil continues to be a major gap in our efforts to manage pesticide use effectively and to minimize contamination of soil, water, and air. Specific topics that W-82 members have identified as requiring attention in this next project period include: fumigant emissions reduction; bioavailability with regards to defining ecologically acceptable endpoints; pH-dependent chemical behavior; microbial structure-activity relationships; the role of gene transfer in efficacy

and resistance; and long-term atmospheric transport of pesticides. Research on these fundamental processes will play a significant role in obtaining several milestones we consider critical in improving environmental stewardship. Research on the fundamental processes will continue to have a significant role particularly with respect to unifying parameters, which can describe these processes over a range of temporal and spatial time scales.

Objective 2: Integrate chemical and biological process information for use in models applicable across different spatial and temporal scales.

The effect of process and interaction between processes in the soil environment on the persistence and movement of pesticides and other organics are manifested across and within the landscape at different spatial and temporal scales, at different intensities at each scale, and with different consequences. The focus of objective 2 is integration of the information obtained on fundamental processes in objective 1 and development of tools for predicting a specific process, group of processes, and coupling of processes in a way that is applicable for use in interpretive and management models. Results can be incorporated into larger scale models, provide realistic inputs for computer simulation models, and be used to evaluate current models and develop future models. Modeling approaches used in the proposed project will be mainly deterministic, and will vary from mechanistic to functional, and from quantitative to semi-qualitative. This approach will allow the members of the proposed project the opportunity to clearly focus their work on issues appropriate to each scale, while simultaneously contributing information required to build, test, extrapolate and utilize larger-scale models for various research and management purposes. Investigations at larger scales are left to other scientists, or to other cooperation outside the thrust of the proposed project. The proposed project has established objectives consistent with the understanding of physical, chemical and biological processes at each scale. The current and previous W-82 projects have established considerable basic knowledge that will contribute to the completion of the proposed project. This will facilitate the integration of basic process understanding at each scale, and provide useful interpretive and management models.

Objective 3: Provide stakeholders with tools for developing strategies to ensure sustainable agriculture and to protect natural resource systems.

The multi-disciplinary knowledge base resulting from performance of Objectives 1 and 2 are necessary for developing sound strategies for ensuring sustainable agriculture and protecting natural resource systems. The focus of Objective 3 is to deliver this information in a format that can be used by stakeholders. Stakeholders in this context are all individuals or groups that can use the knowledge generated in their decision-making process towards improving environmental stewardship. Stakeholders include farmers, chemical manufacturers and other industry groups, natural resource managers, regulators, policy makers, extension educators, research scientists, agricultural and environmental consultants, and the general public.

PROCEDURES:

Objective 1

Interfacial Transfer Processes

The W-82 committee has had a leading role in elucidating sorption/desorption mechanisms and refining methodologies for studying sorption/desorption over the life span of the committee. However, there are still gaps in our knowledge of sorption-desorption processes

in soil that will have a significant effect on our ability to manage pesticide use, optimize soil remediation strategies, and define ecologically acceptable endpoints. Sorption/desorption of organic sorbates onto soil, subsurface material, soil components, and organo-clays will continue to be characterized using batch equilibration and steady-state flow and flow-interrupt miscible displacement techniques as well as thermodynamic, kinetic, spectroscopic, and solvent-extraction techniques. (CA-Berkeley, CA-Riverside, CA-USDA, CT, IN, IA, KS,; MD-USDA/ARS; MI, MN-USDA/ARS, MT, SD, WA). Such experiments will be conducted under different soil to water ratios from unsaturated to flooding conditions, as temperatures, and residence times, and the presence or absence of adjuvants, soil amendments, surfactants, colloids, or co-solutes. In selected studies, model sorbents including polymers, lignin, resins, and soil extracts will be employed to aid in delineation of mechanisms.

Long-term and short-term sorption kinetics and the relationship to bound residue formation will be determined and characterized after varying incubation periods including use of soils contaminated and aged in the field. Various chromatographic, spectroscopic, and radio-label assay techniques will be used to monitor chemical concentrations and chemical alterations in the soil-water system. Soil organic matter will be characterized by ^{13}C-NMR spectroscopy and gas adsorption techniques coupled with molecular dynamics modeling (CA-Berkeley, CT; CA-Riverside; IO; MN). The effect of water composition on Henry's law constants will be examined by air-stripping and wetted-wall techniques.

Specific to wetland ecosytems, the hypothesis that periodic sanding and flooding of bogs affects dichlobenil sorption and thereby leads to changes in volatilization flux and plant uptake will be tested. Dichlobenil sorption will be characterized in several different soils collected from bogs around the U.S.. Batch equilibration methods will be used with various ratios of water and soil. Bog soil will also be amended with varying amounts of sand to simulate periodic sanding operations. These experiments will indicate how much dichlobenil partitions into soil water, which is known to affect bioavailability. After sorption is characterized, bog soil amended with various amounts of sand will be placed in a bottle with an air inlet and outlet. The outlet will be linked through tubing to a train that samples volatile organics in a polymeric matrix and carbon dioxide in potassium hydroxide. Dichlobenil will be applied to the sand layer and its loss monitored for at least one month, which should be the minimal period needed for either adequate weed control or uptake by cranberries that could result in phytotoxicity. Experimental treatments will include different amounts of sand mixed into the bog soil in combination with moisture level (saturated vs. unsaturated). Two forms of dichlobenil, radiolabelled and granular, will be used in an attempt to understand the interaction of volatilization flux and degradation. Parallel greenhouse studies will help elucidate how sanding and soil moisture level affect cranberry and weed growth.

A new method to measure the permeability of plastic films to fumigant vapors will be developed in order to assess permeability under a variety of environmental conditions needed to develop management practices that minimize fumigant flux to the atmosphere. Unlike methods currently used, the new method will be relatively rapid, easy, and repeatable. The effect of environmental conditions (such as temperature and mixtures of soil fumigant chemicals) on permeability will be assessed. Plastic films have demonstrated significant sorption capacity to fumigant vapors. This method will take sorption into account and provide a means to estimate permeability independent of sorption.

Microbial Degradation & Microbial Communities

Laboratory studies investigating microbial degradation of various organic chemicals will be conducted (AL, CA-Davis, CA-USDA/ARS, FL, IN, MI, MN, MN-USDA/ARS, MT; WA). Degradation studies will include both traditional respiration studies as well as molecular-level tools that have been rapidly advancing the last few years.

Laboratory enrichments with target pesticides and non-pesticidal organic compounds under both batch and transport environments will be monitored for changes in chemical composition over time. Subsequent microbial community analysis will be performed using denaturing gradient gel electrophoresis (DGGE) of PCR amplified 16S rDNA fragments (Muyzer et al., 1993). Sequence analysis of prominent DGGE bands will aid in the analysis of microbial community structure of model environments. These sequences will be compared to any contaminant-degrading isolates harvested from enrichment environments, and to soil populations actively degrading the target compounds. Compounds currently targeted for investigation include 2,4-D, 1,3-D, carbofuran, atrazine, aromatic and aliphatic hydrocarbons, and selected antibiotics used in concentrated swine operations.

Enhanced biodegradation of selected chemicals in the environment due to genetic exchange and genetic recruitment will be investigated by studying the organization and plasmid biology of genes encoding metabolism of target chemicals. Bacteria capable of growth on a particular substrate are being studied with respect to their resident plasmids, including confirmation of the role of these plasmids in metabolism of the substrate and similarities between the plasmids. Biodegradative genes are being cloned by standard procedures.

Differential enhanced degradation (stereoisomer-specific degradation) will be investigated using cis and trans 1,3 D. Bacteria capable of degrading 1,3-D will be isolated from soil by analog enrichment strategies. These strains will be thoroughly characterized by 16S rDNA sequence and phenotypic analyses, and enzymes specific for transformation of 1,3-D will be isolated using chromatographic separation.

Atrazine degradation genes are found in geographically-distinct and genetically-diverse soil bacteria. Genes for atrazine biodegradation are located on a large indigenous plasmid, pADP-1, in *Pseudomonas* sp. ADP. This plasmid will be characterized by Southern hybridization analysis, restriction enzyme analysis, and replicon typing. We will determine the complete genomic sequence of pADP-1. We will also determine whether atrazine catabolic plasmids are present in other bacterial strains and whether transposable elements are involved in gene transfer and the assembly and evolution of pADP-1.

We plan to use the incorporation of radiolabeled and ^{13}C-labeled substrates into community lipids as a means of determining which portion of soil communities are responsible for turnover of organic pollutants. This methodology will be used to examine the potential biodegradation of a fuel additive, methyl tertiary butyl ether, by indigenous microbial communities in vadose-zone and groundwater. DNA fingerprinting methods, such as DGGE and ITS, will also be used to explore and confirm the importance of specific populations in biodegradation.

The levels of chlortetracycline, tylosin, and bacitracin, three commonly used antibiotics in commercial swine feeding operations, will be investigated using biological assays and HPLC, GC, and GC-MS analyses. The levels of chlortetracycline, tylosin, and bacitracin in soils and in runoff from swine manure-amended field will be investigated and antibiotic transformation in different soil environments will be studied. The effects of low levels of antibiotics sorbed on soil

minerals surfaces and the extent of antibiotic-resistant gene transfer between swine manure microbial populations and the native soil microbial populations will be studied (KS, IN).

Abiotic Transformation Processes

The role of soil manganese oxides in whole soils to abiotically transform organic chemicals (e.g., organic amines and triazines) will be investigated using traditional equilibrium techniques as well as batch-rate studies (CA-Berkeley, IN). Organic chemical concentrations and transformation will be tracked using GC and HPLC methods with MS,UV, and/or UV resonance Ramon spectroscopic detection. Changes in extractable Mn with time will be monitored using ammonium acetate extractions and ICP analysis. Changes in structural Mn will be investigated using micro-Xanes spectroscopy. In selected experiments, pure Mn oxide solids observed to form biologically will be prepared to vary with respect to surface area, topography, acid site density, and adsorption sites to assess the role of each in Mn-catalyzed transformation reactions.

The role of thiosulfate-facilitated fumigant transformation in aqueous and soil matrices under representative conditions will be investigated (CA-Riverside, CA-USDA/ARS). Transformation pathways will be studied using chromatographic and mass spectrometric techniques and ^{14}C-labelling. Although thiosulfate-facilitated fumigant transformation is chemical reaction during the initial stage, it is expected that microbially mediated mineralization of fumigants may change drastically. This will be studied by measuring mineralization of ^{14}C-labelled fumigants to $^{14}CO_2$ in soil after thiosulfate amendment. Changes in toxicity and carcinogenicity caused by the transformation will be assesses using well-established methods, including test using luminescent bacteria and Ames test.

Objective 2

Information collected in Objective 1 will be integrated to develop algorithms for predicting a specific process or group of processes that can be incorporated into larger scale models, to develop realistic inputs for computer simulation models, to evaluate current models, and to support future model development. Integrated information and algorithms developed will be used to develop and improve management and remediation strategies to reduce soil, air, and water contamination from pesticides and other toxic organics. Historically, W-82 has been involved with obtaining parameter estimates for quantitative description of the processes described under Objective 1. This includes estimates of sorption, models of the sorption process and changes in sorption due to system properties (CA-Berkeley; CA-Riverside; CT; IN; USDA-MN), coupled sorption and degradation (CA-Davis; FL; MT; WA); multiple processes that impact the distribution and transport of volatile and air-borne compounds (USDA-CA), and interfacial phenomena at the field scale (FL; IN).

Previous research has demonstrated a reduction in Kd for napropamide retention with increasing DOM of the soil solution when a drying cycle is included in the sorption process. This reduction in Kd is positively correlated with enhanced facilitated transport through laboratory columns. This will be pursued as a predictor for the potential of facilitated transport as a result of herbicide application practices (CA-Riverside).

Sorption during transport in unsaturated soils has been shown to vary with water content, however, changes in retardation with water content do not follow predictions that assume a linear sorption isotherm and single value of the equilibrium distribution coefficient, K_d. Physical and chemical heterogeneities and the corresponding effect on sorption and transport will be

considered. Changes in sorption (i.e., K_d values) will be evaluated in the context of physical parameters such as hydraulic conductivity and immobile water content. Algorithms to link changes in sorption to quantifiable physical properties will be explored (WA; IN).

The characteristics of the complexes formed between Hillsdale Lake aquatic and water-soluble soil organic matter and 13C-labeled atrazine will be studied with NMR spectroscopy. In addition, the elemental analysis, titrimetric, and spectroscopic (UV-visible, IR and NMR) data characterizing the Hillsdale Lake systems will be used to build molecular models of the systems under investigation. These theoretical models will be used to predict the relative stability constants of the Hillsdale Lake organic matter complexes and to elucidate the nature of the atrazine-organic matter interactions. By providing a molecular-level understanding of these interactions, this research will provide a scientific basis for the analysis of interactions of other anthropogenic organic pollutants with aquatic or soil organic materials (KS).

Soil fumigants are heavily used in intensive agricultural production. However, many fumigants are highly volatile and acutely toxic or carcinogenic. Cooperative studies (CA-USDA, FL) will be conducted to identify approaches to reduce atmospheric emissions of these fumigants while without compromising their effectiveness for pest control. Surface barriers such as impermeable plastic films, fumigant degrading agents, and integration with other agricultural practices such as soil solarization will be investigated at laboratory and field scales. Growers, chemical manufacturers, extension specialists, and commercial applicators will participate in some of these studies. Quantitative evaluation will be made on environmental improvement as a result of these new practices, e.g., reduction in fumigant emissions, and information will be disseminated to growers and regulators.

Chloroacetanilide herbicides including acetochlor, alachlor, metolachlor and propachlor, are among the most heavily used herbicides in the US and the world. Their extensive use has resulted in frequent detection in groundwater and well waters. As some of these herbicides (acetochlor and alachlor) are carcinogenic, removal and prevention of their contamination in the environment is highly desirable. These herbicides react rapidly with thiosulfate fertilizers, giving non-toxic, water-soluble products. Cooperative studies at the laboratory and field scale (CA-USDA, MN) will be conducted to evaluate the application of this approach to remediate soil and water contaminated with these herbicides, reduce their residues in soil and water as a result of agricultural application, and decontaminate pesticide containers and waste disposal sites.

Organic matter, pH, and soil texture vary across rolling topographies. Therefore, the field scale, landscape position influences pesticide fate in the environment. Dissipation of commonly used corn and soybean herbicides will be studied at and across landscape positions. The information developed in Objective 1 will be used to evaluate the processes observed in field studies. The information gained in these studies will be useful to growers and regulators and help in the development of better site-specific management practices.

Objective 3

The multi-disciplinary knowledge base resulting from performance of Objectives 1 and 2 can be used as tools by stakeholders for developing sound strategies for ensuring sustainable agriculture and protecting natural resource systems. However, often time transfer of such information is slow and limited to a small section of the scientific community. Stakeholders include farmers, chemical manufacturers and other industry groups, natural resource managers, regulators, policy makers, extension educators, agricultural and environmental consultants, and the general public as well as research scientists. W-82 is committed to making a sunstantial effort

in this next 5 year project period to transfer user-friendly information to a variety of stakeholders. The W-82 committee realizes that most of our members spend the majority of their time conducting research and in teaching formal courses in line with their formal appointments, leaving little time to spend transferring information outside the scientific community. How technology transfer will be executed is a function of the specific user-group being targeted. Several technology transfer efforts within the W-82 committee, between regional committees, and to user groups outside the immediate science community.

As a first step towards achieving the goals set by Objective 3, the current W-82 members sought out new or renewed participation in our renewal proposal from individuals that hold partial or complete extension appointments. Several individuals with extension appointments including Jack Watson (Pennsylvania), Mike Hirschi (Illinois), and Allan Felsot (Washington) have agreed to take a more active role in technology transfer on behalf of the W-82 committee as well as encourage and facilitate technology transfer by the rest of the committee. The latter is going to be initiated at out first meeting after approval of our renewal proposal. Dr. Jack Watson, an extension specialist (State Program Leader, Cooperative Extension & Outreach at Pennsylvania State University in Agriculture and Natural Resources), will present to the W-82 committee a seminar summarizing recent Kellogg reports that challenge the present approach to outreach/extension, and suggest mechanisms for engaging users in discussion of the information they need, format they can best use, and their targeted application. We believe this seminar and resulting discussions will facilitate and encourage each committee to find ways they can implement technology transfer at several levels.

Research conducted by W-82 members provides information needed to developing management strategies to reduce soil and water contamination from pesticides and other toxic organics. This information includes identification of key parameters controlling the environmental fate of a particular contaminant or contaminant class, expanding the environmental fate information base, quantifying fate relative to various environmental parameters, development of realistic inputs for computer simulation models, and incorporation of algorithms develop to predict a specific processor group of processes that can be incorporated into larger scale models so that are applicable to end-user problems and needs. For example, many larger scale models have already been developed by or through specific support of various clientele toward a specific purpose; however, often times the models either lack proper algorithms describing various fate processes, data needed to make realistic input estimates, or data to validate model outputs. The information base provided by W-82 will supply process models to describe the retention, transformations, and transport of pesticides under soil, climatic and environmental conditions, landscape settings, and cropping and soil management practices that are agrosystem-specific and site-specific. W-82 will work towards enhancing communication and distribution to various users groups throughout the next 5 year project period. Three examples of areas that W-82 members are planning to implement technology transfer outside our are detailed below.

***Assessment of adequacy of simulation models for estimating environmental concentrations (EECs) used in EPA's re-registration eligibility decision documents (REDS*)**: Under the FQPA mandates, EPA is reassessing all tolerances to ensure they meet current definitions of safety. The FQPA has directed EPA to consider aggregate exposure; thus, EPA must consider exposure from drinking water and residential residues as well as food. Furthermore, during the tolerance reassessment process, EPA also characterizes ecological risk and occupation risk. The main mechanism for characterizing risk relies on estimating environmental concentrations. For

drinking water and ecological risk characterizations, EPA runs pesticide behavior simulation models, rather than relying on empirical databases. The main models are GENEEC and PRZM. GENEEC is a tier one model, whereas PRZM allows more probabilistic analysis. The output is an EEC, or estimated environmental concentration.

When the EECs are compared to residue concentrations in various databases, as well as residues reported in various field experiments, it seems that EPA has significantly overestimated their magnitude. Such overestimation leads to exaggerated risk characterizations that belie the observed impacts following many years of use of modern pesticides. Evidently, the models are not accounting for new information about behavioral processes. For example, PRZM does not take into account volatilization, and that pathway of dissipation is now recognized as being more important than realized twenty years ago.

W-82 as a group will review the surface water and ecological risk characterizations in the REDs. It will provide commentary as the validity of the model inputs and reasonableness of the outputs (i.e., the EECs). W-82 will publish its findings and recommendations for improving the development of EECs and thereby contributing to a more scientifically based risk assessment process. The target audience for such an activity are the EPA, state regulatory agencies (which also carry out their own risk characterizations), and commodity organizations using the crop protection tools undergoing scrutiny.

Development of technical documents for methodology and best management practices (BMPs): Members of W-82 engage both laboratory experiments to test hypotheses of soil and water interactions with contaminants. Members also carry out field studies to validate their hypothesis but also to test management practices (BMPs) that can control contaminant fate. Currently, there are few publications available that have specifically detailed recommended methods for carrying out laboratory and field experiments of soil and water interactions with contaminants. W-82 members can collaborate to produce a monograph of recommended methods. The audience for such a document will be students, regulatory agencies, and private consulting research laboratories.

BMPs for contaminant management has been studied for at least 30 years. Presently there are no comprehensive publications that critically assess the validity of these methods nor document results from field studies. W-82 members are well positioned to comprehensively review the literature as well as draw upon their own research experiences to critically analyze current BMPs. The target audience for such a publication would be regulators, agronomy advisors, county extension agents, and non-governmental environmental organizations.

Reduced-risk fumigant application packages: Some of our W-82 members currently are and will continue to evaluate the contribution of a wide variety of soil and application method related variables to fumigant transport and volatilization with the hopes of improving management strategies to reduce fumigant emissions without losing efficacy. Current focus is on evaluating the use of impermeable plastic films, application of fumigants in subsurface drip, and surface amendment of fumigant degrading materials to reduce fumigant emissions. This work will be accomplished with the participation of growers, commercial applicators, and the agrochemical industry. Also collaboration with pest management specialists will be included to ensure that any developed risk-mitigation strategies are not only improved in environmental safety, but are also effective for controlling pests. are that may be directly transferred to growers. In it anticipated that a few reduced-risk fumigant application packages will be developed in the

proposed project period that may be directly transferred to growers to develop feasible strategies to substantially reduce fumigant volatilization without compromising pest control efficacy.

Combined W-82, W-45, and W-188 meeting: In addition to the activities outlined above, W-82 officers are currently working towards holding a combined regional meeting in 2002 between W-82, W-45, and W-188 regional committees. W-45 focuses more on molecular processes and exposure rates, W-82 focuses more on process models and integration of processes, and W-188 focuses on characterization of scale-dependent flow and transport processes. Naturally, there are some research activities that overlap between these committees, but more importantly, there are several research activities that are implicitly linked between committees as summarized in the CRIS Review section of this proposal. A combined meeting will facilitate technology transfer between committees, which is sure to enhance research efforts within all groups, and may also facilitate a better technology transfer product to outside users. In addition, there are members from each of these committees that serve on the editorial staff for the American Society of Agronomy that are working towards approval of developing a Soil and Water Guide which will facilitate transfer of basic information that should be considered in assessing and maintaining soil and water quality.

EXPECTED OUTCOMES:

The research conducted by W-82 committee members will generate new data that will serve to advance the understanding of environmental fate processes of organic chemicals. W-82 committee members have consistently demonstrated productivity through several hundred publications over the past project periods and are expected to continue the publication of research results in peer-reviewed journal articles, books, book chapters, and technical bulletins as well as commentaries and news briefs. Specific outcomes that are expected include the following: (1) novel laboratory and field measurement techniques for characterizing the behavior of organic chemicals in soils water, air, systems and monitoring of chemicals in the environment; (2) improved predictability of chemical release from soils to water and ecological receptors; (3) a more thorough understanding of microbial community population, structure, diversity, and transfer of genetic information as it applies to contaminant degradation behavior in the field; (4) initial assessment of the persistence, occurrence and mobility of antibiotics from animal production units and the potential for impacting microbial communities; (5) evaluate the environmental fate of new pesticides; and (5) recommendations for pesticide use, application methods, and region-, pesticide-, and crop-specific irrigation practices to minimize environmental contamination without compromising efficacy (e.g., reduced-fumigant application methods; BMP in cranberry productions).

ORGANIZATION

The regional research technical committee consists of members who represent AES, USDA-ARS, and other research units. All voting members of the committee are eligible for office. A chairman and secretary are elected each year. The chairperson will coordinate the regional research activities, arrange annual meetings, and prepare annual reports in consultation with the committee members, the administrative advisor, and the CSRS representative. The secretary will record and distribute the minutes of the annual meeting, perform duties of the chairperson in case of absence, and be promoted to chairperson at the conclusion of the one-year

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APPENDIX A: ATTACHMENTS

- Project Leaders
- Resource Listing
- Critical Review of 1995-2000 Project Period

REDUCING THE POTENTIAL FOR ENVIRONMENTAL CONTAMINATION BY PESTICIDES AND OTHER ORGANIC CHEMICALS (W-82)

PROJECT LEADERS

LOCATION	PRINCIPAL INVESTIGATORS	AREA OF SPECIALIZATION
A. EXPERIMENT STATIONS:		
Alabama	Y. Feng	Soil Chemistry
	G. Wu	
Arizona	R. Maier	Soil Microbiology
California-UCB	G. Sposito	Soil Chemistry
California-UCD	K.M. Scow	Soil Microbiology
California-UCR	W.J. Farmer	Soil Chemistry
Colorado	R. Heil	Adminstrator
Connecticut	J.J. Pignatello	Soil Chemistry
Delaware	Mark Radosovich	Soil Biochemistry
Florida	A. Ogram	Soil Microbiology
	A.G. Hornsby	Soil Physics/Extension
	L.T. Ou	Soil Microbiology
	G. Snyder	Soil Chemistry
Hawaii	C. Ray	Agricultural Engineering
Indiana	L.S. Lee	Environ. Organic Chemistry
	R. Turco	Soil Microbiology
	P.S.C. Rao	Soil Physics
	A.P. Schwab	Soil Chemistry
	C. Nakatsu	Soil Microbiology
	M. Bischoff	Soil Microbiology
Illinois	M.C. Hirschi	Ag. Engineering/Extension
Iowa	V.P. Evanglou	Soil Chemistry
Kansas	K. Xia	Soil Chemistry
	Chang-Jeong	
	H. Keller	
Louisianna	R. Hendricks	Environ. Educ./Entomolgy
Minnesota	M. Sadowsky	Soil Microbiology

PROJECT LEADERS (Continued)		
A. EXPERIMENT STATIONS (continued):		
LOCATION	PRINCIPAL INVESTIGATORS	AREA OF SPECIALIZATION
Montana	W.P. Inskeep	Soil Microbiology/Chemistry
Nevada		
Pennsylvania	J.E. Watson	Extension
South Dakota	S. Clay	Soil Chemistry
Washington	A.S. Felsot	Environ. Chemist/Extension
	A.P. Gamerdinger	Soil Physics/Chemistry
B. USDA-ARS		
California	S. Yates	Soil Physics
Maryland	S. Papiernik	Environmental Chemistry
Minnesota	W. Koskinen	Soil Chemistry
C. OTHER PARTICIPANTS		

RESOURCE LISTING						
PARTICIPANT	OBJECTIVES			RESOURCES		
A. EXPERIMENT STATIONS:						
Alabama						
Y. Feng	X			0.1		
G. Wu	X				0.2	
Arizona						
R. Maier	X					
California-UCB						
G. Sposito	X	X				
California-UCD						
K.M. Scow	X			0.3	0.5	
Connecticut						
J.J. Pignatello	X			0.2		
California-UCR						
W.J. Farmer	X	X		0.2		0.3
Colorado						
R. Heil						
Delaware						
M. Radosovich	X			0.1		
Florida						
A. Ogram	X	X		0.2	0.9	0.50
A.G. Hornsby			X	0.5	0	0.50
L.T. Ou	X	X		0.3	0	0.30
G. Snyder	X		X	0.2	0	0
Hawaii						
C. Ray	X			0.2	0.1	
Indiana						
L.S. Lee	X	X	X	0.3		
R. Turco	X			0.1		
P.S.C. Rao	X	X	X	0.1		
A.P. Schwab	X			0.1		
C. Nakatsu	X			0.1		
M. Bischoff	X				0.4	
Illinois						
M.C. Hirschi	X		X			
Iowa						
V.P. Evangelou	X	X		0.2		

RESOURCE LISTING						
(Continued:)	OBJECTIVES			RESOURCES		
Louisianna						
R. Hendricks			X			
Kansas						
K. Xia	X	X		0.2		
Chang-Jeong	X				1.0	
H. Keller	X				0.5	
Minnesota						
M. Sadowsky	X		X	0.1		
Montana						
W.P. Inskeep	X			0.25	0.0	0.25
Pennsylvania						
J.E. Watson			X	0.1		
South Dakota						
Sharon Clay	X			0.1		
Washington						
A.S. Felsot	X		X			
A.P. Gamerdinger	X	X				
B. USDA-ARS						
California						
S. Yates	X	X	X	0.2	0.1	0.2
Marvland						
S. Papiernik	X	X	X	0.1		
Minnesota						
W. Koskinen	X	X		0.1		0.1
C. OTHER PARTICIPANTS						

SY-Scientific man year (project leader); PY-Profesisonal support year (task leader);

TY-Technical support year (technician)

CRITICAL REVIEW OF ACCOMPLISHMENTS UNDER REGIONAL RESEARCH PROJECT W-82

PROJECT PERIOD: **October 1, 1995 - September 31, 2000**

TITLE: PESTICIDES AND OTHER ORGANICS IN SOIL AND THEIR POTENTIAL FOR GROUNDW WATER CONTAMINATION

OBJECTIVES:

1. Characterize mechanisms and quantify processes by which pesticides and other toxic organics interact with soil and water systems.
2. Identify, test and refine models that integrate basic environmental processes across and within spatial and temporal scales.
3. To develop environmentally sound strategies and options that provide guidance for decision makers, based on recognition of user needs, and from the enhanced understanding of natural systems provided by the results of Objectives 1 and 2.

ACTIVE PERSONNEL AND COOPERATING UNIVERSITIES/AGENCIES

Administrative Advisor

R.L. Heil, Colorado State University.

USDA-ARS

J.Seiber, Albany, CA; Jay Gan, Scott Yates* and S. Papiernik, Riverside, California; C.S. Helling* and L. McConnell, Beltsville, Maryland; W.C. Koskinen*, St. Paul Minnesota; T.H. Dao*, El Reno, Oklahoma

Industry

S.S. Harper*, Tennessee Valley Authority.

State Experiment Stations

T.L. Lavy*, University of Arkansas; M.L. Brusseau*, University of Arizona; J. Watson, University of Arizona; M. Ghodrati, University of California, Berkeley; K.M. Scow*, University of California, Davis; G. Sposito*, University of California, Berkeley; W.J. Farmer* and C.E. Castro, University of California, Riverside; J.J. Pignatello*, Conneticut; R.J. Wagenet*, Cornell University; P.S.C. Rao*, A.G. Hornsby, L.T. Ou, and A. Ogram University of Florida; R.E. Green*, University of Hawaii at Manoa; A.P. Schwab*, Kansas State University L.S. Lee, Purdue University; R.F. Turco*; L. V. Evangelou*, Iowa State University; A.P. Schwab*, Kansas State University; S. Boyd*, Michigan State University; H.H. Cheng* and M. Sadowsky, University of Minnesota;; W.P. Inskeep*, Montana State University; R.S. Bowman*, New Mexico Inst. Mining Tech.; J.N. Seiber*, University of Nevada; R.J. Wagenet*, Cornell University; J.S. Selker*, Oregon State University; A.S. Felsot* and Amy Gamerdinger, Washington State University.

WORK ACCOMPLISHED UNDER THE ORIGINAL PROJECT OBJECTIVES

The members of W-82 represent a nationally and internationally recognized group of scientists whose combined efforts have made significant advancements in society's understanding of the relationships among the distribution, transport, and persistence of chemicals. During the past 5 years, W-82 members produced over 350 refereed journal articles and book chapters, several books and edited conference proceedings, a large number of reports and proceedings papers, and several patents. Research accomplishments have centered on (1) expanding our knowledge of the processes by which pesticides and other toxic organics interact in soil-water-air systems, (2) coupling and integrating these processes into useful predictive environmental fate models, and (3) developing or assessing management practices towards pollution prevention and remediation. In addition, W-82 members have presented a large number of invited talks at national and international meetings and symposia are strong indicators of the high visibility of the W-82 members

This review provides an overview of some of the major accomplishments of W82 during the past five years. Space limitations require that this report be more illustrative than comprehensive. The attached list of publications provides details of the broad array of contributions that have resulted from this project. During the past five years, members of W-82 have made a multitude of research contributions under objective 1 which encompasses the fundamental processes controlling the persistence, mobility, and removal of organic chemicals in soils. The majority of research effort on basic mechanisms has been novel and many of the principal investigators are well respected for their contributions and advancements in understanding interactions of organic compounds in soil, water and air. Significant accomplishments were made in the following 5 areas within objective 1: (*i*) sorption/desorption and degradation processes; (*ii*) pesticide transfer to the air and subsequent transport on a regional scale; (*iii*) replacement compounds for methyl) functional and phylogenetic diversity of soil microbial communities; and (*v*) soil remediation. Selected highlights of contributions are detailed below. Many of the results achieved under objective 1 have been used directly for improving predictive capabilities (Objective 2) or for improving management practices that reduce the potential for soil or groundwater contamination (Objective 3).

Sorption-Desorption

An original technique using supercritical carbon dioxide extraction was developed to determine sorption-desorption of herbicides in field-moist soils at selected temperatures was developed (MN-USDA/ARS). Traditionally, sorption-desorption has been characterized using a slurry technique, which often times does not reflect unsaturated conditions in the field. In the case of atrazine, we found greater sorption at low soil water contents than at high water contents. Greater sorption at low moisture contents means that slurry measured sorption coefficients would overestimate the movement of atrazine. This was the first published research on herbicide sorption-desorption in soils at field moisture levels. This research was featured in the "News section" of Environmental Science & Technology (1996, 30:325A). Scientists now have a method for characterizing pesticide sorption-desorption in soil under conditions found in the field. Using pesticide sorption-desorption coefficients measured in field moist-soils at various temperatures, existing solute transport models, such as LEACHP, would be able to better predict movement of pesticides. The results obtained for atrazine using this technique helps explain why transport models over predict leaching in the field.

Investigations conducted over the last several years have focused on the development of an *in situ* diffuse reflectance infrared (DRIFT) spectroscopic technique utilizing chemical isomers as molecular probes of soils and soil constituents (UC-Riverside). We have been able to demonstrate that this technique is uniquely suited to provide information not only with regard to sorption mechanisms but also in the way the status of sorbed molecules changes as a function of time. Impetus for the development of this technology came from the increasing awareness that the sorption/desorption processes for anthropogenic organic chemicals in soil are far more complex and kinetically dependent than previously assumed. Even for very simple chemicals, both processes seem to be comprised at the minimum of a slow and a rapid component. The results of our previous studies with soils are consistent with the suggestions of other researchers that mineral and organic constituents interact in a complex way to form a matrix in which the movement of sorbed chemicals is limited. Our spectroscopic investigations provide molecular level evidence for the kinetic dependence of both sorption and desorption, the existence of a network of pores, the rate-limited accumulation of a persistent sorbed species, and the role of intact organic matter in limiting access to regions of the network. Information gained through infrared spectroscopy is being combined with porosity characterizations using both N_2 and CO_2 as sorptives to provide a dynamic view of the pore system of soil particles as it functions to control the movement of molecular species.

The potential leachability of the new widely used insecticide imidacloprid was characterized (MN-USDA/ARS). Based on its solubility, imidacloprid would be predicted to be highly prone to leaching and a potential groundwater contaminant. Imidacloprid sorption-desorption was characterized on a wide range of soils with varied physical and chemical properties and as a function of concentration and aging in soils. It was determined that under normal agricultural use conditions, imidacloprid does not have a potential for significant leaching through soil. This was the first published research on imidacloprid sorption-desorption in soils. Bayer Corporation used the published data to support registration of imidacloprid use in Arizona, Canada, and Malaysia. As the result of this research, Bayer changed their standard protocol for sorption-desorption studies to how the incumbent performed the research.

The magnitude and rates of various soil processes greatly affect the residence time of a contaminant within a reactive soils profile, thus migration to a potable water supply and bioavailability. Our work has focused on identifying the primary soil processes, quantifying the magnitude and rates of these processes, and correlating these processes to easily obtainable soils and chemical properties. For aromatic amines, reversible sorption processes were shown to reach equilibrium within the one-day for all soil-solute combinations while irreversible binding and transformation proceeded for much longer times. Apparent increases in reversible sorption after one day were shown to primarily result from nonlinear sorption coupled to decreasing aqueous solute concentrations from irreversible binding/transformation. Sorption within a one-day contact time was successfully predicted with a distributed parameter model considering only cation exchange and hydrophobic sorption processes. Both irreversible covalent binding and transformation became increasingly more significant with increasing contact times. Reduction in aromatic amine solution concentrations by fast reversible sorption processes retarded irreversible reactions. For a 1-naphthylamine, p-methoxyaniline, and p-methylaniline evidence of mineral-catalyzed dimerization was confirmed with GC/MS and UV-Vis detection. The magnitude and rate of loss of amines from solution was greatest for the amines where dimerization reactions were evident. For a given soil, irreversible binding of aniline and five para-substituted anilines (methyl, chloro, carboxy, methoxy, and nitro) increased with increasing nucleophilic reactivity

as indicated by decreasing Hammett constants. The natural logarithms of the reaction rates (ln k) were linearly correlated with half-wave potentials (E1/2) and Hammett Constants. Preliminary data suggest that soil Mn is responsible for catalyzing oxidative coupling reactions in whole soils.

Laboratory studies were conducted on 45 soils samples varying in contamination concentrations from 10 mg/kg to 20,000 mg/kg total polyaromatic aromatic hydrocarbons (PAHs) obtained from 11 different manufactured gas plant (MGP) sites (IN; FL). Maximum leachate concentrations were estimated using batch equilibration techniques in both aqueous and methanol/water solutions. Mass-transfer constraints were demonstrated with a subset of soils using batch-rate techniques as well as two types of saturated miscible displacement column techniques: flow-interrupt and steady-state pore-water velocity. The sorption parameters determined for the PAHs from MGP soils support the hypotheses that (1) the sorptivity of tar and coal derived organic material commonly found in MGP soils is greater than what is observed for soils containing only natural organic material (NOM soils); and (2) the rate of release of PAHs from weathered MGP soils appears much slower than from NOM soils. For minimally weathered MGP soils, where total PAH analysis reveals that 2-ring PAHs constitute the bulk of the PAHs present in the tar-soil matrix, leachate concentrations may approach equilibrium. Application of results can be useful in defining site-specific ecological soil screening levels and in assessing remediation options at MGP sites.

The use of batch equilibrium sorption coefficients can be an informative tool for predicting the transport of organic chemicals in soil. However, incorporation of sorption coefficients into models can under-estimate herbicide transport when the chemical moves through preferential flow pathways or in conjunction with a carrier, such as dissolved organic matter (DOM) or clay particles. Napropamide [2-(a-naphthoxy)-N, N-diethyl propionamide], atrazine, and prometryne have been shown to form weak soluble complexes with DOM (CA-Riverside; IN). However, the strength of a pesticide-DOM complex may be enhanced if the herbicide is allowed to dry completely at the soil surface prior to irrigation or precipitation. Batch equilibrium and soil column leaching studies were performed to evaluate the effect of a drying event following herbicide application on herbicide-DOM complex formation. Drying was shown to induce herbicide movement with the wetting front even when preferential flow pathways were eliminated. Increased pesticide movement through soil was the result of stable pesticide-DOM complexes in the solution that lacked an adsorption affinity. The presence of pesticide-DOM complexes was inferred from reduced adsorption coefficients measured on extracts of treated soils as compared to those from aqueous solutions. Indeed the reduction in adsorption coefficient was used to quantitatively estimate the amount of facilitated transport. The formation of pesticide-DOM complexes was verified by dialysis tubing techniques. Preventing the applied herbicide solution from drying before irrigation could reduce or eliminate the threat of facilitated transport of napropamide by DOM.

Degradation

Research is being conducted using molecular genetic analysis to determine (1) changes in the microbial community resulting from anthropogenic perturbations; and (2) biochemical and genetic and basis for the biodegradation of pesticides (especially those based on haloaromatic compounds) by bacteria (IN; FL; MN; CA-Davis; MT). Our results demonstrate that molecular

genetic techniques can be used to explore selective conditions that bring out specific microbial populations in a community and to identify those populations.

Microorganisms can evolve or acquire novel catabolic pathways by assembly of existing genes that have undergone mutation or recombination, by acquisition of genes from other hosts through horizontal gene transfer, or some combination of these mechanisms. A number of studies have investigated transfer of genes in the environment by plasmids but knowledge pertaining to other mobile genetic elements, such as transposons, is limited. The arrangement of 2,4-D catabolic genes (*tfdA-F*) harbored on the large plasmid (~200kb), pTFD41, in *Ralstonia* sp. TFD41 was investigated to determine the mechanism involved in the global distribution of these genes. Hybridization and nucleotide sequencing analysis has revealed that *tfdA* (alpha-ketoglutarate 2,4-D dioxygenase gene), *tfdR* (regulatory gene), are flanked by directly repeated copies of IS1071. IS1071 has been shown to flank a variety of non-homologous catabolic genes with global distribution. This finding suggests that IS1071 has been involved in the dissemination of the *tfdA* and *tfdR* genes. We hypothesize that some complex chemicals that are readily degraded in soil by bacteria do so because they carry the required genes on transposons. This genetic arrangement is selected in the environment because transfer and expression of transposon encoded genes increases their potential host range because they are able to integrate into the existing host genome. By gaining an understanding of the mechanisms for disseminating genes that eliminate xenobiotic chemicals in nature, we can modify or isolate organisms with these traits to remediate soils contaminated with more recalcitrant chemicals.

Four soil bacterial strains capable of completely mineralizing carbofuran were isolated (FL; AL). Three of these strains (CFO6, TA, and CD) are classified as *Sphingomonas* species, while one (H1T-1) is most closely related to *Paracoccus* according to 16S rDNA sequence analysis. Three strains (TA, CD, and H1T-1) were isolated after different numbers of carbofuran applications to plots at a study site in north central Florida. The first strain isolated, CFO6, was isolated from a soil in western Washington State , but is very similar, if not identical, to a strain (HPL) isolated from the Florida study site after two carbofuran applications. Strains TA, CD, and H1T were isolated after several yearly carbofuran applications to this soil. Harbor plasmids that cross-hybridize were found in all four isolates indicating shared sequences. Such genomics information is key to our understanding of enhanced or accelerated biodegradation.

A bacterium (strain PM1) was isolated from a compost-filled biofilter that was treating volatile emissions from a Los Angeles sewage treatment plant that is able to use MTBE as a sole carbon and energy source (CA-Davis). PM-1 is able to mineralize a large portion of the MTBE to CO2 and its cell yield is approximately 0.2 mg cells/mg MTBE. Strain PM1 is capable of degrading a wide range of concentrations of MTBE. The gasoline additive, methyl tertiary butyl ether (MTBE), has become a widespread pollutant in many aquifers and surface waters. MTBE is very mobile, not easily treated by conventional remediation methods, and appears somewhat resistant to biodegradation in the field. To assess the potential for use of strain PM1 in bioaugmentation to remove MTBE contamination, PM 1 was inoculated into groundwater sediment samples obtained from an MTBE-contaminated area at Port Hueneme, California. Strain PM1 was able to rapidly degrade added MTBE under aerobic conditions, and the rate of MTBE degradation increased with each new addition of MTBE to the sediment.

The use of the gaseous fumigant-nematicide methyl bromide in agriculture is scheduled to be phased out in the year 2001. 1,3-Dichloropropene (1,3-D) in combination with chloropicrin and an herbicide is considered to be a viable alternative to methyl bromide for some crops. 1,3-Dichloropropene consists of two isomers, cis- and trans-1,3-D, via hydrolysis. 1,3-D is of

particular interest because it is chiral. Chemical hydrolysis was determined to be responsible for the non-selective degradation of cis- and trans- 1,3-D in soil not previously treated with 1,22-D. However, repeated annual field applications over a span of 12 year of racemic 1,3-D to a Florida sandy soil resulted in enhanced degradation of both cis- and trans-1,3-D. Prior to 1995, this site had been treated with the racemic 1,3-D six times over a span of 12 years. Not only was degradation of 1,3-D enhanced, but trans-1,3D was degraded significantly more rapidly than cis-1,3-D. This phenomenon was termed "differential enhanced degradation."

Emissions of fumigant 1,3-dichloropropene (1,3-D) can result in degradation of air quality in certain areas. The high emission potential of 1,3-D is caused by its rapid diffusion and comparatively slow degradation in soil. One approach to reduce the emission is to enhance the surface soil's capacity to degrade 1,3-D. The capability of two organic amendments, a composted manure (CM) and a biosolid-manure mix (BM), to accelerate 1,3-D degradation in soil was investigated. Degradation of 1,3-D was significantly enhanced in amended soils compared to the unamended soil, and the degree of acceleration varied with the type as well as the rate of amendment. CM was more effective in stimulating 1,3-D degradation than the less decomposed BM. The reduction of 1,3-D emissions by surface application of organic amendments was illustrated column studies as well.

Soil Remediation

Phytoremediation is the use of higher plants to remove contaminants from soil. This technology was demonstrated on soils contaminated with petroleum at three locations in the United States (KS; IN). For a crude oil contaminated soil near the Gulf of Mexico in Texas, the plant species tested were St. Augustine grass, sorghum-sudan grass, and annual rye. Dissipation of the contaminant during the two years of the experiment ranged from 36 to 50% for the vegetated plots compared to 21% in the unvegetated plots. In Virginia, phytoremediation using tall fescue, white clover, and bermuda grass was applied to diesel fuel contaminated soil and compared to unvegetated, contaminated soils. Concentrations decreased 32% in the control plots compared to 40 to 50% in the vegetated plots during the 30 months of the experiment (all differences were statistically significant, $P<0.05$). Soil in California contaminated with heavy, weathered motor oil was tested with phytoremediation using a mix of grass species or a combination of native grasses and legumes. These vegetation treatments resulted in decreases in concentrations of total petroleum hydrocarbons of 37% in 24 months compared to 27% in the unvegetated control. Phytoremediation was observe to perform better than unvegetated controls and rates of decline continue as long as the plants are present. Phytoremediation was shown to be an attractive technology because of its ecologically-friendly approach and low cost, but it is significantly slower than most alternative remediation strategies.

The presence of non-aqueous phase liquids (NAPLs) such as fuels, oils, and organic solvents in the subsurface, strongly sorbed contaminants, highly recalcitrant organic chemicals limit the success of both bioremediation and traditional pump-and-treat remediation technologies. Although excavation and disposal of such soils at landfills is an option, concerns regarding long-term liability might preclude this option. The use of *in-situ* flushing techniques for removal of DNAPLs and recalcitrant compounds was extensively investigated at both laboratory and field-scale (IN; FL; AZ). Data were gathered at several field-scale tests of in-situ flushing technique for enhanced dissolution/extraction of NAPL source zones. Also evaluated were innovative *in-situ* source zone characterization techniques involving partitioning and

interfacial tracers to quantify the NAPL volume and spatial distribution as well as the NAPl-water interfacial area. These tracer tests were used in conjunction with soil coring to evaluate the spatial patterns in NAPL volume and areas in NAPL interfaces, both before and after implementing some in-situ technique for site cleanup, in order to evaluate the effectiveness of remediation achieved and the possible impacts of the cleanup technology on hydrodynamic and bio-geochemical processes.

Aqueous Transport

A multiplexed fiber optic miniprobe (FOMP) system capable of measuring solute breakthrough curves (BTCs) in situ and in realtime at a single "point" within the soil matrix was developed (CA-Riverside). The system allows continuous operation of up to 20 FOMPs simultaneously. The multiplexed system was used to investigate the efficacy of FOMP measurements, as well as the point-to-point spatial and temporal variability of solute transport parameters in a large silica sand column. The results indicate that even at this "point" scale, the convective-dispersive flow through the sand matrix is relatively uniform. The BTCs in these experiments were described relatively well using the convection-dispersion equation (CDE). Experimental evidence suggest that the FOMPs are likely measuring resident concentration rather than flux-averaged concentration. We tested and compared three nondestructive methods to monitor solute transport in a large, repacked clay loam soil column. The methods compared included time domain reflectometry, FOMP, and continuous small tube pore water samplers. All three probes performed well and produced BTCs which were adequately described by the CDE. The average effective pore water velocity estimated by fitting the data to the CDE was similar for all probes; however, the effective dispersion coefficients estimated using data from each type of probe were systematically different.

A novel method using centripetal acceleration for conducting unsaturated transport experiments under steady-state flow conditions are necessary for evaluating the mechanisms that determine contaminant transport through the vadose zone (WA). The use of centripetal acceleration to measure steady-state hydraulic properties has been demonstrated during the last decade by USGS researchers; however, this approach has not been widely adopted by the soil science community. The experimental system was modified by researchers at Pacific Northwest National Laboratory (PNNL) to include an ultracentrifuge, rotor, and rotating seal. The modified centrifuge is referred to as the "unsaturated flow apparatus" or UFA (Beckman Scientific). The rotating seal has two inlet ports which are plumbed to separate pumps, allowing simultaneous fluid delivery by two independent flow paths, to two soil samples or columns. The ability to achieve and maintain stable average water contents and average flow velocities was validated in miscible displacement experiments with non-reactive and reactive tracers.

Pesticide Emissions & Atmospheric Transport

Atmospheric inputs of pesticides transported from California's Central Valley to the Sierra Nevada mountains (California, USA) were investigated by collecting winter-spring precipitation (rain and snow) from Sequoia National Park and from the Lake Tahoe Basin. Pesticides currently used in California's Central Valley were detected in snow and rain samples from two elevations in Sequoia National Park (SNP) in the southern Sierras and in the Lake Tahoe Basin, elevation 2,200 m. To determine the extent of summertime atmospheric transport of pesticides to this region, high volume air, dry deposition, and surface water samples were

collected in the Central Valley and at different elevations in California's Sequoia National Park. Results revealed that the highest residue concentrations were for compounds with heavy summertime agricultural use. A significant drop in pesticide concentrations in both air and water samples was observed within a few hundred meters elevation from the Valley; however, levels remained relatively constant between ~500-2000 m. Water concentrations from two areas above 3000 m contained levels 10 to 100 times lower than the other areas. While more work is required to determine actual annual flux values to the region, results from this project do indicate that pesticides volatilized from the Central Valley are a consistent source of these toxicants to the Sierra Nevada mountains. Similar studies were conducted for assessing PCB congener concentrations in air, water, fish, and snow were determined in the Lake Tahoe Basin and in the water and fish of an adjacent but more remote rural comparison lake, Marlette Lake, to examine whether atmospheric transport and deposition are primary sources of contamination to this alpine region of the Sierra Nevada mountains. PCB was found in all compartments sampled, particularly snow and air, and the similar pattern of congeners in surface waters and in fish from Lake Tahoe and Marlette Lake indicate an atmospheric source of contamination.

A field study was conducted in September 1995 to measure the ambient atmospheric concentrations of methyl bromide (MeBr) in the Salinas Valley, California. Air concentrations of MeBr were measured at 11 sites located on the adjacent mountains, valley floor and at the Pacific Ocean coast over a 4-day period. ISCST3 (Industrial Source Complex-Short Term 3) dispersion model simulations were performed when several fumigated fields were emitting MeBr vapors. The model-predicted values were compared with measured values with the objective to characterize this tool for estimating area-wide concentrations of MeBr and in turn assessing potential exposure to this fumigant for the inhabitants of the valley. Good correlation ($R^2 \approx 0.7$) were observed between predicted and measured concentrations. Meteorological factors were identified as being responsible for the variation between measured and modeled concentrations in addition to variation in source emission rates. The ISCST3 model was found successful for modeling downwind concentrations due to multiple agricultural emissions in a complex terrain.

Atmospheric degradation of chlorofluorocarbon (CFC) replacement compounds, namely HFC-134a, HCFC-123, and HCFC-124, results in the formation of trifluoroacetic acid (TFA). A simple and sensitive analytical procedure was developed to determine the concentration of TFA in plant, soil, and water samples. The analysis involves extraction of TFA by sulfuric acid and ethanol followed by derivatization to the methyl ester of TFA (MTFA). This was accomplished within a single vial without complex extraction procedures. The highly volatile MTFA is then analyzed using headspace gas chromatography. The spike recovery trials from all media ranged from a low of 86.7% to a high of 121.4%. The minimum detectable limit (MDL) for the method was 34 ng/g for dry plant material, 0.20 ng/g for soil, and 6.5 ng/L for water. This is the first practical method for low-level analysis of TFA in plants and soil, and it is a significant improvement over previous methods for TFA in water.

Little is known about fumigant degradation in soil under high temperature or low moisture conditions that prevail near the soil surface during fumigation. The effect of soil temperature and moisture on the rate of degradation of 1,3-dichloropropene (1,3-D), methyl isothiocyanate (MITC) and chloropicrin (CP) was determined for extended soil temperature and moisture ranges. Results clearly indicated the need to consider specific responses of fumigant degradation to temperature and moisture variations when describing their transport in the environment, and should prove to be useful for designing fumigation practices that reduce atmospheric emissions.

Methyl bromide plays an important role in stratospheric ozone depletion, but its sources are not well defined. *Brassica* plants were shown to take up Br− from soil, produce methyl bromide and release it into the air. Emission of methyl bromide was detected from plants grown in natural soils containing 0.06-0.31 mg kg^{-1} Br−; the emission increases with increasing soil Br− level. Given the ubiquitous distribution of Br− in soil, methyl bromide production by terrestrial higher plants is likely a large source for atmospheric methyl bromide.

Plastic tarps are often used in soil fumigation to contain the chemical in the soil to increase efficacy and decrease emissions of fumigant vapors. Plastic films were shown to have a significant capacity to sorb fumigant vapors and that the sorption is largely reversible (CA-Riverside, USDA/ARS; FL). Three agricultural films (polyethylene and two high-barrier films) were tested with four soil fumigants (methyl bromide, chloropicrin, 1,3-dichloropropene, and propargyl bromide, a potential alternative to methyl bromide). Significant sorption of all fumigants to all the films at field-relevant concentrations was observed. Sorption isotherms were linear over an order of magnitude increase in vapor concentration, indicating that the film may be a large sink for fumigant vapors. Sorption of most fumigants was very rapid, with the bulk of the sorption occurring within the first few minutes of contact. Desorption was also rapid, with most desorption occurring within minutes after the film samples were removed to fresh air. Sorption/desorption may be important in reducing emissions and determining worker exposure, and should be considered in measurements involving agricultural films.

Management Practices

The W-82 group has focused attentions on testing best management practices under field conditions and have been directly involved in technology transfer. Several strategies were found to be effective in controlling off-site losses or decreasing the persistence of pesticides and other chemicals that are applied to the land surface water (CA-Riverside-USDA/ARS; Nevada-USDA-ARS; WA; FL).

The performance of several drip and furrow irrigation systems used to apply the systemic low-risk aphicide, imidacloprid was investigated under filed conditions. Imidacloprid was found to remain in the top three feet of the soil profile in two commercial furrow-irrigated hop yards with different water scheduling practices. The infrequent watering common to furrow irrigation seemed to allay translocation, but surface erosion was enhanced. In contrast to furrow irrigation, drip irrigation eliminated soil erosion. In two commercial hop yards, we have noted that a surface drip system and a shallowly buried drip tape system operated on a daily watering schedule adequately slowed imidacloprid movement below three feet. A more deeply buried drip system using polyethylene tubing and labyrinth emitters was not effective at preventing imidacloprid movement below three feet when irrigation occurred daily for four hours. When irrigation was turned on only in response to soil sensing of a predetermined optimal matrix potential, imidacloprid stayed in the injection zone and did not translocate. A water management plan was implemented involving no irrigation for one week following subsurface injection of imidacloprid. Imidacloprid leaching was reduced compared to treatments with routine daily irrigation following application. However, leaching was still greater than when irrigation was controlled by feedback from moisture sensors. Our preliminary results suggest that drip chemigation may be effectively employed without significant leaching if during the early season irrigation is delayed for at least one week following aphicide application.

Methyl isothiocyanate (MITC) residues in air measured during and immediately after application by drip irrigation injection were 1-2 orders of magnitude lower than those previously published for applications involving surface irrigation. MITC is a potential replacement for methyl bromide. Application of organic wastes as a means of reducing the emission of both methyl bromide and MITC by enhancing their degradation in surface soil was investigated. The degradation of both compounds was significantly accelerated in composted manure or biosolid-manure amended soils, and the enhancement was much greater for MITC than for methyl bromide. As certain organic amendments can suppress soil pathogens on their own, integrating fumigation with organic waste application may potentially provide complementary pest control activity. Addition to the soil surface of the fertilizer ammonium thiosulfate (ATS) was also found to reduce methyl bromide emissions. As ATS is an inexpensive fertilizer, the reported approach is promising for field application.

Model Development & Evaluation

Model development or validation was performed by several groups (MN; FL; WA; Riverside-USDA). A model-based prescription for best irrigation and fertilizer management practices for irrigated potato production in the glacial outwash soil of North Central Minnesota was developed (MN). A decision support system, "Pesticide Economic and Environmental Tradeoffs (PEET)", for improving pesticide product selections which minimize surface and groundwater contamination, while maintaining economic profitability was developed (FL). Software to aid in management decision, "Soil, environmental and agricultural management systems (SEAMS)", was developed (FL). Simulation models were developed for forward and inverse modeling of the in-situ remediation process, and partitioning, interfacial, and bioreactive tracer data used to assess the volume, interfacial area, and degradation rates of a non-aqueous liquid contamination in the subsurface (FL). A modeling approach for approximating the effect of sorption on degradation in soils was developed, which can be applied when site specific degradation data is not available (WA). Approaches for modifying estimates of sorption input parameters that are typically measured on the <2 mm size fraction were developed to include the effect of larger particle sizes which can improve transport modeling (WA). A simulation model for fate and transport of methyl bromide in vegetable plastic-mulched beds was developed (FL). The impact of uncertainty in model input data on predicted pesticide leaching, and sorption and transport of acidic pesticides in variable-charged soils was evaluated (FL).

SUMMARY IF CRITICAL REVIEW

In summary, W-82 produced results of practical utility in diverse areas ranging from soil remediation to minimizing worker exposure and off-site transport of fumigants to the atmosphere. Work completed under Objectives 1 and 2 were used directly to improve management practices and stewardship of our natural resources (Objective 3). Much of the work of W-82 is used by agency personnel, growers, and educators. Most of the work is carried out by students and postdoctoral fellows who become the research and development workforce of the future.

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APPENDICES D from Participants