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Data Related Uncertainty in Near-Surface Vulnerability Assessments for Agrochemicals in the San Joaquin Valley

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Precious groundwater resources across the United States have been contaminated due to decades-long nonpoint-source applications of agricultural chemicals. Assessing the impact of past, ongoing, and future chemical applications for large-scale agriculture operations is timely for designing best-management practices to prevent subsurface pollution. Presented here are the results from a series of regional-scale vulnerability assessments for the San Joaquin Valley (SJV). Two relatively simple indices, the retardation and attenuation factors, are used to estimate near-surface vulnerabilities based on the chemical properties of 32 pesticides and the variability of both soil characteristics and recharge rates across the SJV. The uncertainties inherit to these assessments, derived from the uncertainties within the chemical and soil data bases, are estimated using first-order analyses. The results are used to screen and rank the chemicals based on mobility and leaching potential, without and with consideration of data-related uncertainties. Chemicals of historic high visibility in the SJV (e.g., atrazine, DBCP [dibromochloropropane], ethylene dibromide, and simazine) are ranked in the top half of those considered. Vulnerability maps generated for atrazine and DBCP, featured for their legacy status in the study area, clearly illustrate variations within and across the assessments. For example, the leaching potential is greater for DBCP than for atrazine, the leaching potential for DBCP is greater for the spatially variable recharge values than for the average recharge rate, and the leaching potentials for both DBCP and atrazine are greater for the annual recharge estimates than for the monthly recharge estimates. The data-related uncertainties identified in this study can be significant, targeting opportunities for improving future vulnerability assessments.

Everything should be made as simple as possible, but not simpler.
—attributed to Albert Einstein

There's no such thing as a free lunch.
—Milton Friedman

The challenge of assessing groundwater vulnerability at the regional scale due to nonpoint-source agrochemical applications is not a trivial one (Corwin et al., 1997, 1999). The use of simple mobility–leaching indices to generate assessments of subsurface vulnerability provides information that can be useful within the decision-management arena. These assessments rest on chemical, soil, and recharge data that are sparse and, therefore, contain uncertainty.

The ~12,000-km^2 San Joaquin Valley (SJV) is the regional-scale system of interest in this study. Located at the southern end of California's Central Valley (Fig. 1), the SJV is a structural basin filled with thousands of meters of sedimentary material creating a nearly flat alluvial plain (Loague et al., 1998b). The soils on the more fertile eastern side of the SJV are derived from the granitic Sierra Nevada, with large areas of wind-deposited sands underlain by deep coarse-textured alluvial material (National Research Council, 1993). The sediments of the SJV tend to be interlayered gravel, sand, silt, and clay derived from the surrounding mountains and deposited in alluvial-fan, floodplain, flood basin, lacustrine, and marsh environments (Domagalski, 1998). As a direct result of large-scale irrigation infrastructure and agrochemical use, the SJV has produced a significant fraction of the food grown in North America for decades. The availability of precious groundwater resources in the SJV is well documented (e.g., Belitz, 1988; Faunt, 2009). The impacts from agrochemicals to groundwater quality within the SJV have received considerable attention (e.g., Holden, 1986; Smith, 1989; Domagalski and Dubrovsky, 1992; National Research Council, 1993; Barbash and Resek, 1996; Burow et al., 1998a,b, 1999, 2007).

There are now excellent regional-scale examples that illustrate the spatial variability of information and response for the near surface (e.g., Lemieux et al., 2008; Gleeson et al., 2011a,b). The

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Abbreviations: AF, attenuation factor; DBCP, dibromochloropropane; EDB, ethylene dibromide; RF, retardation factor; SJV, San Joaquin Valley; S_{AF}, standard deviation in AF; S_{RF}, standard deviation in RF.

methods used to assess the fate and transport of pesticides within the variably saturated subsurface, ranging from simple models to comprehensive physics-based models, are often reviewed (e.g., Jury and Fluher, 1992). An early approach to assess groundwater vulnerability across large areas was the standardized DRASTIC system (Aller et al., 1987). Past assessments of groundwater vulnerability for the SJV include the index-based approaches reported by Meeks and Dean (1990) and Zhang et al. (1995), the discriminate and logistic regression analyses reported by Teso et al. (1988, 1995, 1996), the transfer function approach reported by Stewart and Loague (2004), and physics-based simulations of flow and transport reported by Loague et al. (1998a,b) and Burow et al. (1999).

The two-fold objective of the work reported here is (i) to produce near-surface vulnerability maps for the entire SJV for a suite of targeted chemicals and (ii) to characterize the data-related uncertainties in these assessments. The significant difference between this study and past work is the quantitative characterization of the impact data-related uncertainties and alternative water-balance derived recharge estimates have on near-surface vulnerability assessments for 32 agrochemicals within the SJV. The vulnerability assessments reported here provide food for thought but should not be taken as a conclusive tool for ongoing management decisions.

Materials and Methods

Retardation and Attenuation Factors

The *attenuation factor* (AF) index, proposed by Rao et al. (1985) to screen and rank the likelihood of agrochemical leaching, has been used successfully for subsurface vulnerability assessments for the Hawaiian island of Oahu (Loague, 1991, 1994; Loague et al., 1989, 1990, 1996; Giambelluca et al., 1996) and the Canary island of Tenerife (Diaz-Diaz et al., 1998; Diaz-Diaz and Loague, 2000b). The AF index used here is as follows:

$$\mathrm{AF} = \exp\left(\frac{-0.639 d \mathrm{RF} \theta_{\mathrm{FC}}}{q t_{1/2}}\right) \qquad [1]$$

where d [m] is the distance from the surface to a compliance depth, RF [dimensionless] is the retardation factor, θ_{FC} [$\mathrm{m^3\ m^{-3}}$] is the soil-water content at field capacity, q [$\mathrm{m\ d^{-1}}$] is the net recharge, and $t_{1/2}$ [d] is the chemical half-life. The retardation factor in Eq. [1] is given by

$$\mathrm{RF} = 1 + \frac{\rho_b f_{\mathrm{OC}} K_{\mathrm{OC}}}{\theta_{\mathrm{FC}}} \qquad [2]$$

where ρ_b [$\mathrm{kg\ m^{-3}}$] is the soil bulk density, f_{oc} [mass fraction] is the soil organic carbon, and K_{oc} [$\mathrm{mL\ g^{-1}}$] is the chemical sorption coefficient. The RF and AF indices loosely represent the important processes of sorption and advection, decay, and sorption, respectively. Neither index provides concentration information. The range of possible values for AF is between zero and one; RF = 1.0 for nonsorbing chemicals. The simplifying assumptions and major limitations of the indices, relative to known processes of near-surface chemical transport and fate, were described by Kleveno et al. (1992). The scales used here to divide the AF and RF values into mobility–leaching ranges (after Khan et al., 1986) are given in Table 1. The assignment of numerical values to the various classes is subjective; therefore, the classes shown in Table 1 only indicate relative retardation and attenuation.

Fig. 1. Location of the San Joaquin Valley study area within the central valley of California.

First-Order Uncertainty Analysis

In regional-scale assessments of subsurface vulnerability with, for example, the AF and RF indices, there will of course be uncertainty associated with error propagation (Heuvelink, 1998). First-order analysis is a simple approach for estimating the uncertainties in a deterministic model due to parameter uncertainty (Cornell, 1972). With first-order uncertainty analysis, the total uncertainty is determined from the component uncertainty contributed by each variable, C_i. The uncertainty in the ith parameter (i.e., data-related error) for RF and AF are given, respectively, by the following:

$$C_i =^{1} \left|\frac{\partial \mathrm{RF}}{\partial P_i}\right| S_{P_i} \qquad [3]$$

$$C_i =^{1} \left|\frac{\partial \mathrm{AF}}{\partial P_i}\right| S_{P_i} \qquad [4]$$

Table 1. Mobility–leaching scales for the retardation factor (RF) and attenuation factor (AF) indices (after Khan et al., 1986).

Value	Classification
RF	
=1.0	Very mobile (VM)
>1.0 and <2.0	Mobile (M)
≥2.0 and <3.0	Moderately mobile (MM)
≥3.0 and <10.0	Moderately immobile (MI)
≥10.0	Very immobile (VI)
AF	
≥0.0 and <0.001	Very unlikely (VU)
≥0.001 and <0.01	Unlikely (U)
≥0.01 and <0.1	Moderately likely (ML)
≥0.1 and <0.25	Likely (L)
≥0.25 and ≤1.0	Very likely (VL)

where $=^1$ means equal in the first-order sense and S_{P_i} is the standard deviation of the parameter P_i (i.e., ρ_b, f_{oc}, K_{oc}, and θ_{FC} for RF; d, RF, θ_{FC}, q, and $t_{1/2}$ for AF). The standard deviations (uncertainties) in RF and AF estimates are calculated, respectively, from the component uncertainties as

$$S_{\mathrm{RF}} =^1 \left(\sum_{i=1}^{n} C_i^2 \right)^{1/2} \qquad [5]$$

$$S_{\mathrm{AF}} =^1 \left(\sum_{i=1}^{n} C_i^2 \right)^{1/2} \qquad [6]$$

The equations for the AF and RF component uncertainties are given by Loague et al. (1990).

A risk-averse approach is taken for estimating the data-related uncertainties in the RF and AF vulnerability assessments for the SJV. While one can easily produce RF ± S_{RF} and AF ± S_{AF} estimates, here only the RF − S_{RF} and AF + S_{AF} scenarios are considered. For this study if RF − S_{RF} or AF + S_{AF} is less than or greater than one, respectively, then the values are (subjectively) set to one. For this study, the standard deviations for RF and AF have been subtracted and added, respectively, from the mean values without regard to the normality and degrees of confidence that can be associated with one, two, or three standard deviations. It is useful to think of the frequency distributions for these calculations as uniform and truncated based on scale limits (Loague et al., 1990). The reader is reminded that the various classes for RF and AF in Table 1 are arbitrary.

Data

The chemical, soil, and recharge information needed to parameterize Eq. [1–6] for the SJV is gleaned, respectively, from Hornsby et al. (1996), Blanke (1999), and Mills (2004). The mean and standard deviation values for K_{oc} and $t_{1/2}$ for the 32 agrochemicals examined in this study are given in Table 2. The 32 pesticides are (or have been) used in association with agriculture. The selection of these pesticides for this study was

Table 2. Characteristics of the 32 agrochemicals included in this study (after Hornsby et al., 1996; Diaz-Diaz and Loague, 2000a).

Chemical	K_{oc}†	S_{Koc}	$t_{1/2}$	$S_{t1/2}$
	mL g^{-1}		d	
Alachlor [2-chloro-N-(2,6-diethylphenyl)-N-(methoxymethyl)acetamide]‡	170	205	15	10
Aldicarb [2-methyl-2-(methylthio)-propionaldehyde O-(methylcarbamoyl)oxime]‡	30	21	30	22
Atrazine (2-chloro-4-ethylamino-6-isopropylamino-S-triazine)‡§	100	39	60	29
Bromacil [5-bromo-3-(sec-butyl)-6-methyluracil]‡	32	14	60	87
Carbaryl (1-naphthyl-N-methylcarbamate)‡	300	297	10	5
Carbofuran (2,3-dihydro-2,2-dimethyl-7-benzofuranyl-n-methylcarbamate)‡	22	23	50	23
Carboxin (5,6-dihydro-2-methyl-1,4-oxathiin-3-carboxanilide)‡	260	73	3	2
Chlorothalonil (tetrachloroisophthalonitrile)‡	1380	1276	30	25
Cyanazine (2[[4-chloro-6(ethylamino)-S-triazin-2-yl]amino]-2-methylpropionitrile)‡	190	111	14	28
2,4-D (2,4-dichlorophenoxy acetic acid)‡	20	12	10	4
2,4-DP [butoxyethyl ester of (±) 2-(2,4-dichlorophenoxy)propanoic acid]§	1000	285	10	1
Dalapon (2,2 dichloropropionic acid) (sodium salt)‡	1	1	30	4
DBCP (1,2-dibromo-3-chloropropane)§	70	26	180	44
DCPA (dimethyl 2,3,5,6-tetrachloro-1,4-benzenedicarboxylate)‡	5000	693	100	25
Diazinon [O, O-diethyl-O-(2-isopropyl-4-methyl-6-pyrimidinyl)phosphorothiote]‡§	1000	507	40	8
Dicamba (2-methoxy-3,6-dichlorobenzoic acid)‡§	2	2	14	9
1,2-Dichloropropane (1,2-Dichloropropane)‡§	50	1	700	321
Dinoseb (2-sec-butyl-4,6-dinitrophenol)‡	30	133	30	9
Diphenamid (N,N-dimethyl-a-phenylbenzeneacetamida)‡	210	42	30	9
Disulfoton (O,O-diethyl S-[2-(ethylthio)ethyl]phosphorodithioate)‡	600	1008	30	25
Diuron [3-(3,4-dichlorophenyl)-1,1-dimethylurea]‡§	480	252	90	86
EDB (1,2-dibromoethane)§	34	43	100	53
Methomyl [S-methyl-N-((methylcarbamoyl)oxy)-thioacetamidate]‡	72	38	30	12
Metolachlor [2-chloro-N-(2-ethyl-6-methylphenyl)-N-(2-methoxy-1-methylethyl)acetamide]‡	200	60	90	49
Metribuzin [4-amino-6-(1,1-dimethylethyl)-3-(methylthio)-1,2,4-triazin-5(4H)-one]‡	60	25	40	28
Oxamyl [methyl 2-(dimethylamino)-N-[[(methylamino)carbonyl]oxy]-2-oxoethanimimdothioate]‡	25	6	4	5
Prometon [6-methoxy-N,N′-bis(1-methylethyl)-1,3,5-triazine-2,4-diamine]§	150	157	500	560
Prometryn [N,N′-bis(1-methylethyl)-6-(methylthio)-1,3,5-triazine-2,4-diamine]§	400	644	60	102
Propazine [6-chloro-N,N′-bis(1-methylethyl)-1,3,5-triazine-2,4-diamine]§	154	32	135	93
Simazine [2-chloro-4,6-bis(ethylamino)-s-triazine]‡§	130	34	60	40
Tebuthiuron [N-[5-(1,1-dimethyl)-1,3,4-thiadizol-2-yl]-N,N′-dimethylurea]‡	80	178	360	282
Trifluralin [2,6-dinitro-N,N-dipropyl-4-(trifluoromethyl)benzenemamine]‡	8000	3608	60	21

† K_{oc}, soil organic carbon/water partition coefficient; S_{Koc}, standard deviation for K_{oc}; $t_{1/2}$, half-life in soil; $S_{t1/2}$, standard deviation for $t_{1/2}$.

‡ Identified by USEPA (1988) as high risk relative to groundwater contamination.

§ Detected in California groundwater (Domagalski and Dubrovsky, 1992).

based on identification by the USEPA (1988) as high risk relative to groundwater contamination and/or detection in California groundwater as reported by Domagalski and Dubrovsky (1992). It is worth noting that Holden (1986) reported the detection of 52 pesticides in California groundwater. Inspection of Table 2 reveals that for several chemicals, considerable uncertainty is associated with the chemical information (e.g., coefficient of variation values greater than one for K_{oc} and/or $t_{1/2}$). The chemicals featured for the vulnerability assessment illustrations (i.e., maps) in the Results section, due to their high profiles within the SJV (e.g., Loague et al. (1998a,b); Stewart and Loague, 2004), are dibromochloropropane (1,2-dibromo-3-chloropropane), also known as DBCP, and atrazine (2-chloro-4-ethylamino-6-isopropylamino-S-triazine).

The spatial distribution of soils within the SJV is illustrated in Fig. 2, which was generated by first digitizing and then combining (within a GIS framework) USDA Soil Conservation Service soil survey information for the study area (see Arkley, 1962, 1964; Arroues and Anderson, 1986; Huntington, 1971; Ulrich and Stromberg, 1962) into a comprehensive map at the order taxonomic level using the scaling approach described by Loague et al. (1989). The map in Fig. 2 is made up of 41,192 individual soil-mapping units. The mean and standard deviation values for θ_{FC}, ρ_b, and f_{oc} for the different soil orders (and soil order combinations) across the study area are given in Table 3. The soil characteristics in Table 3 are based on more than 13,000 values gleaned from the USDA Map Unit Interpretation Record (MUIR) database and the five soil surveys covering the study area. Inspection of Table 3 shows that there is some uncertainty within the soils information (e.g., coefficient of variation values less than one for θ_{FC}, ρ_b, and/or f_{oc}), albeit considerably less than for the chemical data. The individual steps used in the preparation of Fig. 2 and Table 3 were described by Blanke (1999). Maps found in Blanke (1999) illustrate the spatial variability (and uncertainty) of θ_{FC}, ρ_b, and f_{oc} across the study area. The d in Eq. [1] was conservatively set at 1.0 m for this study, with no uncertainty.

Three different annual recharge estimates for the study area were used. The first estimate is an average value of 0.87 mm d^{-1} (with a standard deviation of 0.59 mm d^{-1}), gleaned from the Fresno case study simulations reported by Loague et al. (1998a,b). The Fresno recharge estimates are for an area of 1174 km^2 over a 35-yr period (Loague et al., 1998a,b); see Fig. 1 for the location of the Fresno case study. The second and third estimates are for spatially variable recharge across the study area, based on a simple water-balance approach given by the following:

$$q = P + I - \mathrm{ET} \qquad [7]$$

where P is precipitation [m d^{-1}], I is irrigation [m d^{-1}], and ET is evapotranspiration [m d^{-1}]. Both annual and monthly water balances are generated to estimate annual recharge values. Potential evapotranspiration (PET) was estimated from temperature using the Thornthwaite and Mather (1955) method as:

$$\mathrm{PET} = 1.6\left(10T/I'\right)^{a} \qquad [8]$$

where T is temperature (°C), and a and I' are empirical weights (see Dunne and Leopold, 1978). The relationship between PET and ET used here is (Dunne and Leopold, 1978):

$$\mathrm{ET} = \mathrm{PET} \cdot f\left(\frac{\theta_{\mathrm{AW}}}{\theta_{\mathrm{AWC}}}\right) \qquad [9]$$

where f is an empirical weight (taken here as 1.0), θ_{AW} is the available soil-water content [m^3 m^{-3}], and θ_{AWC} is the available soil-water capacity [m^3 m^{-3}]. The θ_{AW} and θ_{AWC} are estimated as

$$\theta_{\mathrm{AW}} = (\theta - \theta_{\mathrm{WP}}) \qquad [10]$$

$$\theta_{\mathrm{AWC}} = (\theta_{\mathrm{FC}} - \theta_{\mathrm{WP}}) \qquad [11]$$

where θ is the soil-water content [m^3 m^{-3}], θ_{WP} is the soil-water content at the permanent wilting point [m^3 m^{-3}], and θ_{FC} is the soil-water content at field capacity. An average value of 90% of θ_{FC} was used for θ in this study for the entire SJV, based on the conservative (risk-averse) assumption that intensive irrigation maintains the soil-water content close to field capacity. The θ_{WP} was estimated for each soil order in the SJV on the basis of soil

Table 3. Characteristics of the soils within the San Joaquin Valley study area.†

Soil order/combination	θ_{FC}‡	$S\theta_{FC}$	ρ_b	$S\rho_b$	f_{oc}	Sf_{oc}
	m^3 m^{-3}		kg m^{-3}		mass fraction	
Alfisols	0.251	0.052	1519	68.3	0.006	0.003
Aridisols	0.288	0.071	1438	184.8	0.008	0.005
Entisols	0.213	0.063	1566	67.6	0.006	0.003
Inceptisols	0.244	0.065	1510	57.8	0.006	0.002
Mollisols	0.276	0.87	1485	86.0	0.013	0.005
Vertisols	0.390	0.063	1362	62.6	0.011	0.003
Alfisols/Entisols	0.232	0.058	1542	67.9	0.006	0.003
Alifisols/Inceptisols	0.248	0.059	1514	63.1	0.006	0.002
Alfisols/Inceptisols/Mollisols	0.257	0.068	1504	70.7	0.008	0.003
Alfisols/Mollisols	0.264	0.069	1502	77.1	0.009	0.004
Aridisols/Entisols	0.250	0.067	1502	126.2	0.007	0.004
Entisols/Mollisols	0.245	0.075	1525	76.8	0.009	0.004

† Information originally from the USDA Map Unit Interpretation Record (MUIR) Website, which has been shut down by Natural Resources Conservation Service (NRCS); the information is now available from the NRCS Soil Data Mart Website at http://soildatamart.nrcs.usda.gov/ (accessed 25 Mar. 2012).

‡ θ_{FC}, volumetric fraction of soil-water content at field capacity; $S\theta_{FC}$, standard deviation for θ_{FC}; ρ_b, soil bulk density; $S\rho_b$, standard deviation for ρ_b; f_{oc}, soil organic carbon; Sf_{oc}, standard deviation for f_{oc}.

texture (Brady, 1984; Stewart and Loague, 2004). Estimates of recharge were made within a GIS framework, using spatially variable temperature, rainfall, and land use information. The two spatially variable estimates of annual recharge for the SJV are illustrated in Fig. 3. Inspection of Fig. 3 shows that the recharge rates are significantly greater based on the annual water balance than for the summed monthly water balances. Both maps in Fig. 3 show considerable spatial variations in the estimated recharge. Estimating component uncertainties for the spatially variable recharge rates in Fig. 3 was beyond the scope of this study. Mills (2004) provides maps showing the spatial variability of P, T, θ_{WP}, PET, and PET across the SJV as used in the water-balance estimates.

Results

Retardation Factor

Table 4 summarizes RF estimates of near-surface vulnerability for 32 chemicals developed for the six soil orders in the SJV. The results in Table 4 are ranked, classified, and reclassified on the basis of mobility and data-related uncertainty. The uncertainty results are presented only for the risk-averse case (i.e., RF – S_{RF}). Inspection of Table 4 reveals that relative to soil taxonomy, the rankings are the same, as the values of $S\theta_{\mathrm{FC},}$ $S\rho_{\mathrm{b}}$, and Sf_{oc} are relatively small across the soil orders. The differences between the RF and RF – S_{RF} estimates are explained by large $S_{K\mathrm{oc}}$ values. With consideration for the data-related uncertainties, the changes in the vulnerability estimates are greatest for carbaryl, prometryn, disulfoton, and chlorothalonil. (The RF and S_{RF} estimates, on which the results in Table 4 are based, are provided in Supplemental Table S1.)

Figures 4a and 4b are near-surface vulnerability maps of DBCP mobility across the SJV, based on, respectively, the RF and RF – S_{RF} values in Table 4. Inspection of Fig. 4a and 4b shows that without and with consideration for data-related uncertainties, the DBCP vulnerability estimates generally fall, respectively, in the *moderately immobile* and *mobile* classifications. In terms of subsurface vulnerability, the maps in Fig. 4a and 4b are neither the best nor the worst relative to spatially variable estimates. (Subsurface vulnerability maps for the SJV, based on RF and RF – S_{RF} estimates, for each chemical listed in Table 2 are provided in Supplemental Fig. S1–S32.)

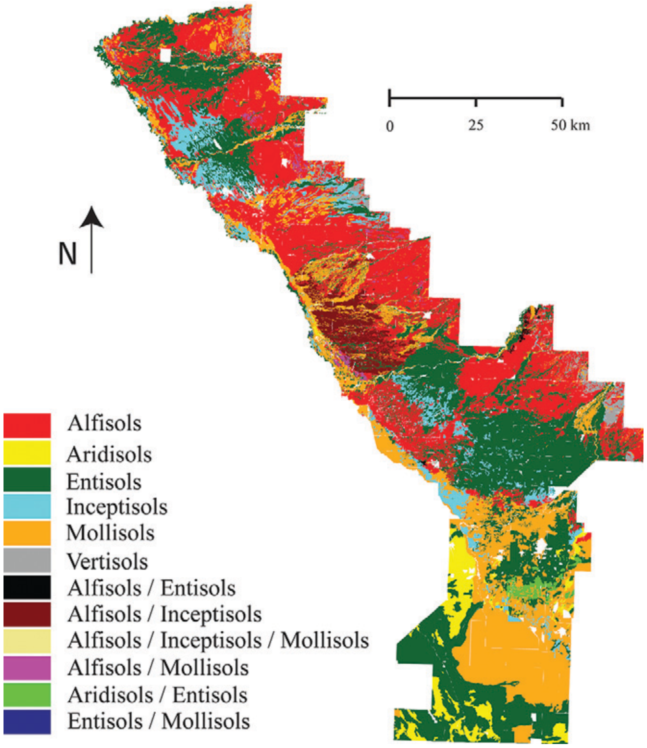

Fig. 2. Locations of the six soil orders (and their combinations) within the San Joaquin Valley study area.

AF with Average Recharge

Table 5 summarizes AF estimates of near-surface vulnerability for 32 chemicals developed for the six soil orders in the SJV. The results in Table 5 are ranked, classified, and reclassified on the basis of leaching potential and data-related uncertainty. The uncertainty results are presented only for

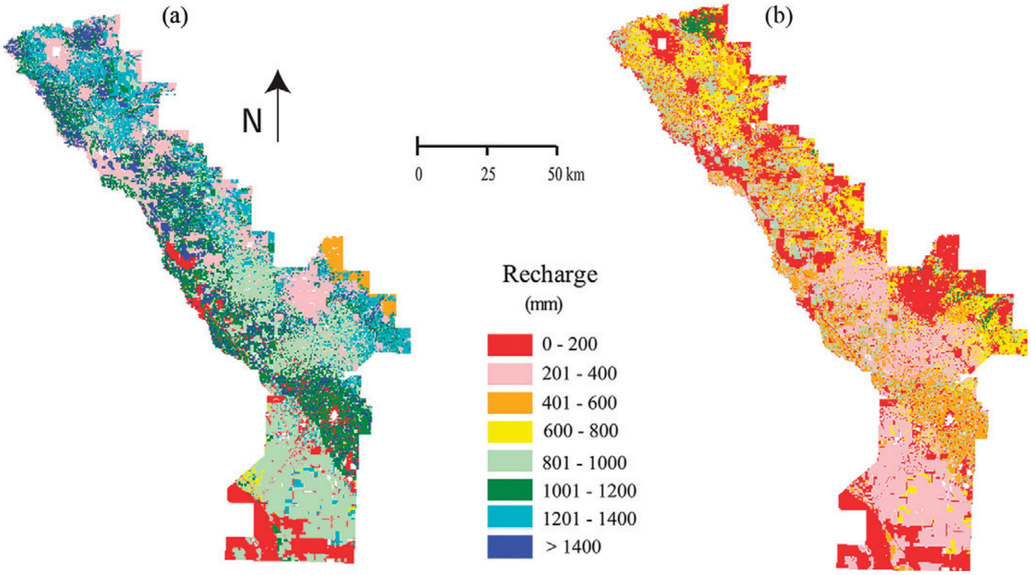

Fig. 3. (a) Annual recharge estimates based on an annual water balance for the San Joaquin Valley study area. (b) Annual recharge estimates based on monthly water balances for the San Joaquin Valley study area.

Table 4. Ranking, classification, and uncertainty of retardation factor (RF) estimates for the soils within the San Joaquin Valley study area for 32 chemicals. (See Table 2 for full chemical names.)

Chemical	Soil order					
	Alfisols	Aridisols	Entisols	Inceptisols	Mollisols	Vertisols
Dalapon	1,[†] M,[‡] VM[§]	1, M, VM	1, M, VM	1, M, VM	1, M, VM	1, M, VM
Dicamba	2, M, VM	2, M, VM	2, M, VM	2, M, VM	2, M, VM	2, M, VM
2,4-D	3, M, M	3, M, M	3, M, M	3, M, M	3, MM, M	3, M, M
Carbofuran	4, M, VM	4, M, VM	4, M, VM	4, M, VM	4, MM, VM	4, M, VM
Oxamyl	5, M, M	5, MM, M	5, MM, M	5, MM, M	5, MM, M	5, M, M
Aldicarb	6, MM, M	6, MM, M	6, MM, M	6, MM, M	6, MI, M	6, MM, M
Dinoseb	7, MM, VM	7, MM, VM	7, MM, VM	7, MM, VM	7, MI, VM	7, MM, VM
Bromacil	8, MM, M	8, MM, M	8, MM, M	8, MM, M	8, MI, M	8, MM, M
EDB	9, MM, VM	9, MM, VM	9, MM, VM	9, MM, VM	9, MI, VM	9, MM, VM
1,2-Dichloropropane	10, MM, M	10, MI, M	10, MI, M	10, MI, MM	10, MI, MM	10, MM, MM
Metribuzin	11, MI, M	11, MI, M	11, MI, M	11, MI, M	11, MI, MM	11, MI, MM
DBCP	12, MI, M	12, MI, M	12, MI, M	12, MI, MM	12, MI, MM	12, MI, MM
Methomyl	13, MI, M	13, MI, M	13, MI, M	13, MI, M	13, MI, MM	13, MI, M
Tebuthiuron	14, MI, VM	14, MI, VM	14, MI, VM	14, MI, VM	14, MI, VM	14, MI, VM
Atrazine	15, MI, MM	15, MI, MM	15, MI, MM	15, MI, MM	15, MI, MI	15, MI, MM
Simazine	16, MI, MM	16, MI, MM	16, MI, MI	16, MI, MI	16, VI, MI	16, MI, MI
Prometon	17, MI, VM	17, MI, VM	17, MI, VM	17, MI, VM	17, VI, VM	17, MI, VM
Propazine	18, MI, MI	18, MI, MI	18, MI, MI	18, MI, MI	18, VI, MI	18, MI, MI
Alachlor	19, MI, VM	19, MI, VM	19, MI, VM	19, MI, VM	19, VI, VM	19, MI, VM
Cyanazine	20, MI, MM	20, MI, MM	20, MI, MM	20, MI, MM	20, VI, MI	20, MI, MI
Metolachlor	21, MI, MI	21, MI, MI	21, MI, MI	21, MI, MI	21, VI, MI	21, MI, MI
Diphenamid	22, MI, MI	22, MI, MI	22, MI, MI	22, MI, MI	22, VI, MI	22, MI, MI
Carboxin	23, VI, MI	23, VI, MI	23, VI, MI	23, VI, MI	23, VI, MI	23, VI, MI
Carbaryl	24, VI, VM	24, VI, VM	24, VI, VM	24, VI, VM	24, VI, VM	24, VI, VM
Prometryn	25, VI, VM	25, VI, VM	25, VI, VM	25, VI, VM	25, VI, VM	25, VI, VM
Diuron	26, VI, MI	26, VI, MI	26, VI, MI	26, VI, MI	26, VI, VI	26, VI, VI
Disulfoton	27, VI, VM	27, VI, VM	27, VI, VM	27, VI, VM	27, VI, VM	27, VI, VM
2,4-DP	28, VI, VI	28, VI, VI	28, VI, VI	28, VI, VI	28, VI, VI	28, VI, VI
Diazinon	29, VI, MI	29, VI, MI	29, VI, VI	29, VI, VI	29, VI, VI	29, VI, VI
Chlorothalonil	30, VI, VM	30, VI, VM	30, VI, VM	30, VI, VM	30, VI, VM	30, VI, M
DCPA	31, VI, VI	31, VI, VI	31, VI, VI	31, VI, VI	31, VI, VI	31, VI, VI
Trifluralin	32, VI, VI	32, VI, VI	32, VI, VI	32, VI, VI	32, VI, VI	32, VI, VI

[†] Ranking of RF estimates, from most to least vulnerable.

[‡] Classification of RF estimates: very mobile (VM), mobile (M), moderately mobile (MM), moderately immobile (MI), very immobile (VI); the leaching scale is given in Table 1.

[§] Classification of RF – S_{RF} estimates: very mobile (VM), mobile (M), moderately mobile (MM), moderately immobile (MI), very immobile (VI); the leaching scale is given in Table 1.

the risk-averse case (i.e., AF + S_{AF}). The rankings in Tables 5 differ significantly from those in Table 4 (i.e., advection, decay, and sorption [AF] versus sorption [RF]). Inspection of Table 5 reveals that relative to soil taxonomy, the rankings are nearly the same without and with consideration of data related uncertainties, as the values of $S\theta_{FC,}$ $S\rho_{b}$, and Sf_{oc} are relatively small across the soil orders. The differences between the AF and AF + S_{AF} estimates are explained by large values of $S_{K_{oc}}$ and $S_{t1/2}$. With consideration for the data-related uncertainties, the changes in the vulnerability estimates are greatest for tebuthiuron, prometon, and bromacil. (The AF and S_{AF} estimates, on which the results in Table 5 are based, are provided in Supplemental Table S2.)

Figures 4c and 4d are near-surface vulnerability maps of DBCP leaching potentials across the SJV, based on, respectively, the AF and AF + S_{AF} values in Table 5. Inspection of Fig. 4c and 4d shows that without and with consideration for data related uncertainties, the DBCP vulnerability estimates generally fall, respectively, in the *very unlikely* to *moderately unlikely* and *unlikely* to *likely* classifications. In terms of subsurface vulnerability, the maps in Fig. 4c and 4d are neither the best nor the worst relative to spatially variable estimates. (Subsurface vulnerability maps for the SJV, based on AF and AF + S_{AF} estimates with an average recharge rate, for each chemical listed in Table 2 are provided in Supplemental Fig. S1–S32.)

AF with Spatially Variable Recharge

Figures 5 and 6 are near-surface vulnerability maps of DBCP and atrazine leaching potentials across the SJV, respectively, based on AF and AF + S_{AF} estimates. The vulnerability estimates in Fig. 5 and 6 were developed with two spatially variable recharge scenarios. The uncertainty results are presented only for the risk-averse case (i.e., AF + S_{AF}). Comparing the three different recharge scenarios for DBCP clearly indicates that the vulnerability estimates are greater for the spatially variable rates (see Fig. 5aI and 5aII) than for the average rate (see Fig. 4c). Inspection of Fig. 5 and 6 shows that the vulnerability for DBCP is greater than for atrazine, driven by the chemical characteristics (see Table 2). As should be expected, the leaching potential for DBCP and atrazine is greater for the higher recharge rates (see Fig. 3).

Inspection of Fig. 5 shows that without and with consideration for data-related uncertainties, respectively, the DBCP vulnerability estimates generally fall within the *moderately likely* to the *very likely* and *likely* to *very likely* classifications. Inspection of Fig. 6 shows that without and with consideration for data-related uncertainties the atrazine vulnerability estimates generally fall within the *very unlikely* to *moderately likely* classifications. In terms of subsurface vulnerability, the maps in Fig. 5 and 6 are neither the best nor the worst for the spatially variable estimates. (Subsurface vulnerability maps for the SJV, based on AF and AF + S_{AF} estimates for spatially variable recharge estimates, for each chemical listed in Table 2 are provided in Supplemental Fig. S33–S64.)

Discussion

The simplicity of using mobility–leaching indices for regional-scale nonpoint-source vulnerability assessments as demonstrated here is extremely seductive relative to exhaustive numerical simulations driven by the coupled nonlinear partial-differential equations of variably saturated fluid flow and solute transport (e.g., Stewart and Loague, 2003, 2004). The regional-scale assessments presented here clearly show that there are differences in the estimated vulnerabilities due to variability in the chemical and soil properties, variability in the recharge estimates, and the uncertainty within the databases. This study provides a quantitative characterization of data-related uncertainty impacts for applications of the RF and AF indices. Note that Ugalde (2000) investigated the impact of correlation between soil variables (i.e., ρ_b, f_{oc}, θ_{FC}) on the uncertainty in RF estimates for aldicarb and carboxin for the SJV, finding the differences (between the no cross and cross correlation) to be <10%. There is no question that the indices used here are laced with model error by the simplification and omission of important processes. McGrath et al. (2009), by comparison, described a leaching index that includes consideration for rainfall variability and preferential flow. There are always tradeoffs between model complexity, model error, and data requirements (Loague and Corwin, 1996; Loague et al., 1996).

As demonstrated here, uncertainties within the chemical and soil databases can play a key role in clarifying the usefulness of regional-scale nonpoint-source vulnerability assessments. Improvements to the chemical and soil databases would reduce uncertainty in the vulnerability assessments for the SJV. This study also makes clear that the amount and spatial distribution of recharge greatly affects estimates of leaching potential. The differences between average recharge rates and spatially variable recharge estimates can be significant. Future efforts designed to characterize the vulnerability associated with nonpoint sources of agrochemical contamination should focus first on securing the best possible estimate of spatially variable recharge and the underlying component uncertainties.

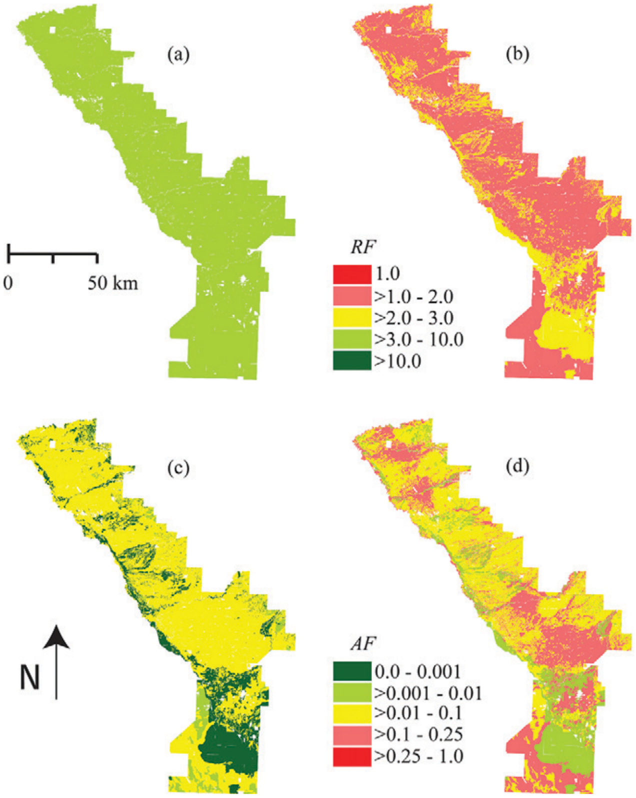

Fig. 4. DBCP vulnerability maps, in the form of (a) RF, (b) RF − S_{RF}, (c) AF, and (d) AF + S_{AF} estimates, for the San Joaquin Valley study area. For AF, the estimates are based on average annual recharge values. RF, retardation factor; S_{RF}, standard deviation in RF; AF, attenuation factor; S_{AF}, standard deviation in AF.

The near-surface vulnerability assessments reported here for a 1-m depth should not be used to assess the vulnerability of groundwater resources at much greater depths (e.g., ~ 25 m in parts of the study area). As it was not possible to rigorously evaluate the assessments reported here against actual chemical occurrences, they are offered from a "what-if" perspective. It is interesting to note, however, that four of the first five ranked chemicals in Table 5 (1,2-dichloropropane, prometon, DBCP, and ethylene dibromide [EDB]) have all been detected in the groundwater within the SJV (Bennett et al., 2006; Burton and Belitz, 2008; Landon and Belitz, 2008), which lends some qualitative support the assessments. There are problems making comparisons of this type in judging model performance. For example, simazine, often detected in groundwater within the SJV is ranked 16th in Table 5, whereas tebuthiuron (ranked second) is rarely detected.

An unavoidable limitation of the assessments presented here is the subjectivity of the classification schemes in Table 2. In general, the relative ranking of chemical leaching potentials is more useful than the classifications. It is possible, due to large applications rates (which are not considered in the assessments reported here), that some compounds are capable of leaching past the root zone despite being strongly sorbed within the near surface (e.g., diuron and simazine). An interesting question to consider relative to the observed versus predicted comparisons is whether the dominate underlying processes

Table 5. Ranking, classification, and uncertainty of attenuation factor (AF) estimates for the soils within the San Joaquin Valley study area for 32 chemicals. (See Table 2 for full chemical names.)

Chemical	Soil order					
	Alfisols	Aridisols	Entisols	Inceptisols	Mollisols	Vertisols
1,2-Dichloropropane	1,[†] VL,[‡] VL[§]	1, VL, VL	1, VL, VL	1, VL, VL	1, L, VL	1, VL, VL
Tebuthiuron	2, L, VL	2, ML, VL	2, L, VL	2, L, VL	2, ML, L	2, ML, VL
Prometon	3, ML, VL	3, ML, L	3, ML, VL	3, ML, VL	3, U, ML	3, ML, L
DBCP	4, ML, ML	4, U, ML	4, ML, L	4, ML, ML	4, VU, U	4, U, ML
EDB	5, ML, ML	5, U, ML	5, ML, ML	5, ML, ML	5, VU, U	5, VU, U
Dalapon	6, VU, U	6, VU, U	6, U, ML	6, U, U	6, VU, U	6, VU, U
Bromacil	7, VU, ML	8, VU, U	8, U, ML	8, VU, ML	8, VU, VU	8, VU, U
Carbofuran	8, VU, U	7, VU, U	7, U, ML	7, VU, U	7, VU, VU	7, VU, U
Propazine	9, VU, U	9, VU, VU	9, VU, U	9, VU, U	10, VU, VU	9, VU, VU
Aldicarb	10, VU, VU	10, VU, VU	10, VU, VU	10, VU, VU	11, VU, VU	10, VU, VU
Dinoseb	11, VU, VU	11, VU, VU	11, VU, VU	11, VU, VU	12, VU, VU	11, VU, VU
Atrazine	12, VU, VU	13, VU, VU	13, VU, VU	13, VU, VU	14, VU, VU	13, VU, VU
Dicamba	13, VU, VU	12, VU, VU	12, VU, VU	12, VU, VU	9, VU, VU	12, VU, VU
Metribuzin	14, VU, VU	14, VU, VU	14, VU, VU	14, VU, VU	13, VU, VU	14, VU, VU
Metolachlor	15, VU, VU	15, VU, VU	15, VU, VU	15, VU, VU	15, VU, VU	15, VU, VU
Simazine	16, VU, VU	16, VU, VU	16, VU, VU	16, VU, VU	16, VU, VU	16, VU, VU
Methomyl	17, VU, VU	17, VU, VU	17, VU, VU	17, VU, VU	17, VU, VU	17, VU, VU
2,4-D	18, VU, VU	18, VU, VU	18, VU, VU	18, VU, VU	18, VU, VU	18, VU, VU
Diuron	19, VU, VU	19, VU, VU	19, VU, VU	19, VU, VU	19, VU, VU	19, VU, VU
Prometryn	20, VU, VU	20, VU, VU	20, VU, VU	20, VU, VU	20, VU, VU	20, VU, VU
Diphenamid	21, VU, VU	21, VU, VU	21, VU, VU	21, VU, VU	21, VU, VU	21, VU, VU
Alachlor	22, VU, VU	23, VU, VU	23, VU, VU	23, VU, VU	23, VU, VU	23, VU, VU
Oxamyl	23, VU, VU	22, VU, VU	22, VU, VU	22, VU, VU	22, VU, VU	22, VU, VU
Cyanazine	24, VU, VU	24, VU, VU	24, VU, VU	24, VU, VU	24, VU, VU	24, VU, VU
Disulfoton	25, VU, VU	25, VU, VU	25, VU, VU	25, VU, VU	25, VU, VU	25, VU, VU
Diazinon	26, VU, VU	26, VU, VU	26, VU, VU	26, VU, VU	26, VU, VU	26, VU, VU
Carbaryl	27, VU, VU	27, VU, VU	27, VU, VU	27, VU, VU	27, VU, VU	27, VU, VU
Chlorothalonil	28, VU, VU	28, VU, VU	28, VU, VU	28, VU, VU	28, VU, VU	28, VU, VU
DCPA	29, VU, VU	29, VU, VU	29, VU, VU	29, VU, VU	28, VU, VU	29, VU, VU
Carboxin	30, VU, VU	30, VU, VU	30, VU, VU	30, VU, VU	28, VU, VU	30, VU, VU
Trifluralin	31, VU, VU	30, VU, VU	31, VU, VU	31, VU, VU	28, VU, VU	30, VU, VU
2,4-DP	31, VU, VU	30, VU, VU	31, VU, VU	31, VU, VU	28, VU, VU	30, VU, VU

[†] Ranking of AF estimates, from most to least vulnerable.

[‡] Classification of AF estimates: very likely (VL), likely (L), moderately likely (ML), unlikely (U), very unlikely (VU); the leaching scale is given in Table 1.

[§] Classification of AF + S_{AF} estimates: very likely (VL), likely (L), moderately likely (ML), unlikely (U), very unlikely (VU); the leaching scale is given in Table 1.

match. For example, relative to regional-scale groundwater quality data, one must determine if the contamination is from a point or nonpoint source (e.g., spills or dumping versus label-recommended applications). Loague and Abrams (1999) discussed this problem for EDB hotspots. In an ideal world, the sampling scheme would be designed a priori to properly test model performance. Clearly, comprehensive ground truth information, from within the near surface for this study, is invaluable for judging model performance (see Loague and Green, 1990, 1991). With information on near-surface occurrences, one could design and establish more rigorous (site specific) classification schemes to characterize vulnerability assessments of the type reported here.

It is always better to regulate than it is to remediate. Assuming that shortfalls in the chemical and soil databases can be satisfied, and improvements made for the water-balance derived recharge estimates, revised assessments of subsurface vulnerability, in the spirit of those reported here, may be useful to those charged with regulating the future use of agriculture chemicals in the SJV. Furthermore, the areas and chemicals shown to have the greatest potential for mobility and leaching, on the basis of a simple screen–rank approach, could be targeted for future measure and model investigations, driven by risk-adverse motives, with detailed field observations and comprehensive physics-based simulation (forensic or predictive) focused on worst-case scenarios and optimal remediation strategies (see Diaz-Diaz and Loague, 2001).

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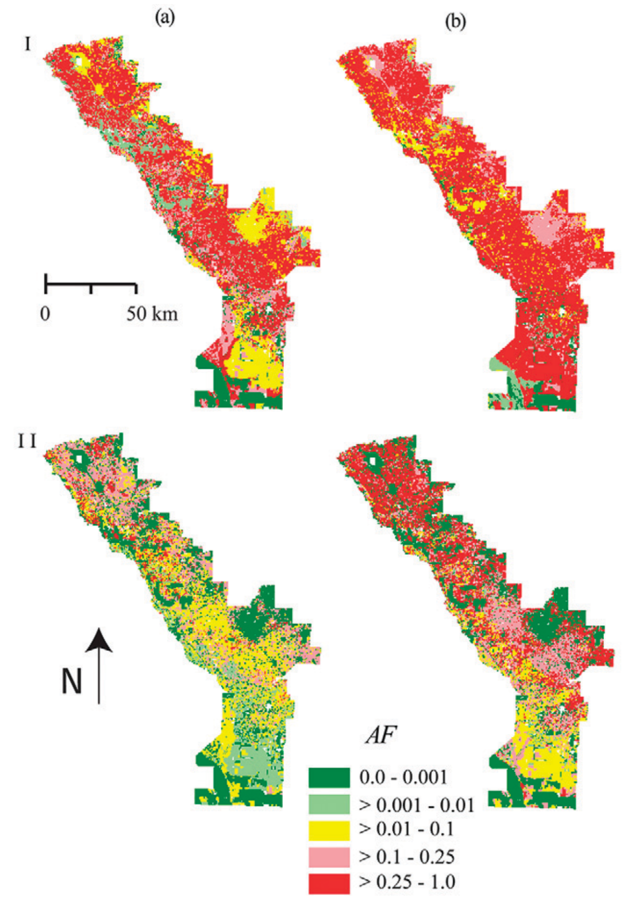

Fig. 5. DBCP vulnerability estimates for the San Joaquin Valley study area based on the annual (I) and monthly (II) recharge estimates. (a) AF; (b) AF + S_{AF}. RF, retardation factor; S_{RF}, standard deviation in RF; AF, attenuation factor; S_{AF}, standard deviation in AF.

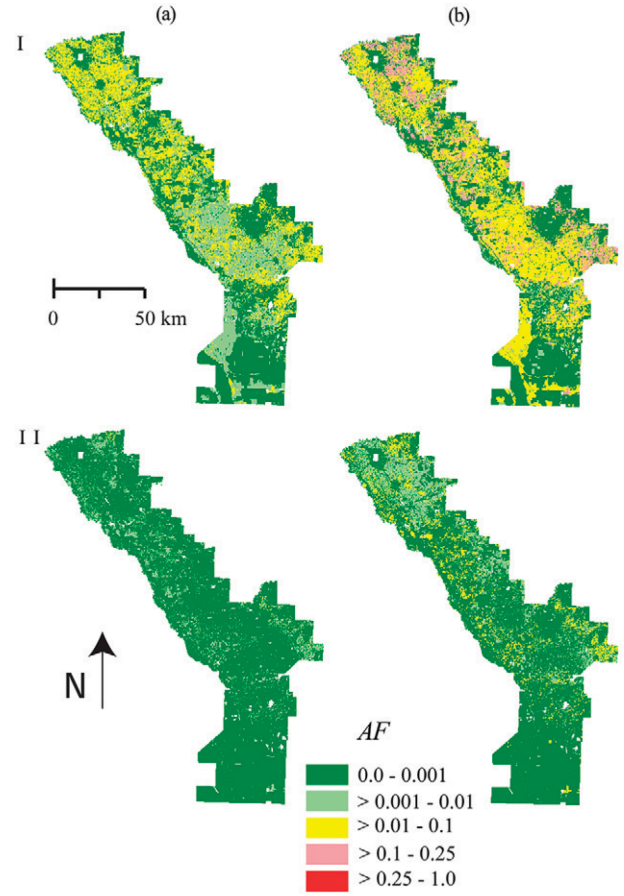

Fig. 6. Atrazine vulnerability estimates for the San Joaquin Valley study area based on the annual (I) and monthly (II) recharge estimates. (a) AF; (b) AF + S_{AF}. RF, retardation factor; S_{RF}, standard deviation in RF; AF, attenuation factor; S_{AF}, standard deviation in AF.

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