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Spatial and temporal patterns of pesticide use on California almonds and associated risks to the surrounding environment

Yu Zhan, Minghua Zhang *

Department of Land, Air, and Water Resources, University of California, Davis, CA 95616, USA

HIGHLIGHTS

- Saptiotemporal patterns of pesticide use/risk in California almonds were studied.
- Use intensities of insecticides/fungicides/herbicides showed latitudinal gradients.
- Overall, herbicide use increased considerably, while fungicide use decreased.
- The risks to surface water, groundwater, and soil decreased in many areas.
- Risk patterns were mainly associated with use patterns of high-risk pesticides.

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ABSTRACT

Various stakeholders of California almonds have been investing efforts into mitigating pesticide impacts on human and ecosystem health. This study is the first comprehensive evaluation that examines the spatial and temporal patterns of pesticide use and associated environmental risks. The pesticide use data from 1996 to 2010 were obtained from the Pesticide Use Reporting database. The Pesticide Use Risk Evaluation indicator was employed to evaluate the pesticide environmental risks based on the pesticide properties and local environmental conditions. Analyses showed that the use intensities (*UI*) of insecticides (oils accounted for 86% of the total insecticide *UI*) and herbicides both increased from north to south; fungicides showed the opposite spatial pattern; and fumigants were used most intensively in the middle region. The *UI* of fungicides and herbicides significantly decreased and increased, respectively, throughout the study area. The insecticide *UI* significantly decreased in the north but increased in many areas in the south. In particular, the organophosphate *UI* significantly decreased across the study area, while the pyrethroid *UI* significantly increased in the south. The fumigant *UI* did not show a trend. The regional risk intensities of surface water (RI_W), soil (RI_S), and air (RI_A) all increased from north to south, while the groundwater regional risk intensity (RI_G) decreased from north to south. The main trends of RI_W, RI_G, and RI_S were decreasing, while the RI_A did not show a trend in any region. It's noticeable that although the herbicide *UI* significantly increased, the *UI* of high-leaching herbicides significantly decreased, which led to the significant decrease of RI_G. In summary, the temporal trends of the pesticide use and risks indicate that the California almond growers are making considerable progress towards sustainable pest management via integrated pest management, but still require more efforts to curb the fast increase of herbicide use.

1. Introduction

Almonds are one of the most important specialty crops in California, USA, which produced about 80% of the global almond supply and generated \$3.87 billion in revenue in 2012 (Almond Board of California, 2012). Almost all the California almond orchards (3080 km^2 in 2012) are located in the Central Valley (58,000 km^2), which has a mild climate, fertile soil, and abundant sunshine. The Central Valley is one of the most productive agricultural areas in the world. Key pests in almond are navel orangeworm (*Amyelois transitella*), San Jose scale (*Quadraspidiotus perniciosus*), peach twig borer (*Anarsia lineatella*), web-spinning spider mites, and ants (CEPA, 2011). In the dormant season, oil spray alone can control low to moderate populations of San Jose scale and mites. When populations of peach twig borer (also targeted during bloom) and San Jose scale are high, oils are likely sprayed with other insecticides. In the growing season, insecticide treatments (mainly in July and August) mostly control navel orangeworm. Diseases during winters and early springs, such as anthracnose (pathogen: *Colletotrichum acutatum*), brown rot blossom blight (pathogen: *Monilinia laxa*), and scab (pathogen: *Cladosporium carpophilum*) are controlled by various fungicides, e.g., captan, copper, or ziram (UC IPM, 2012). Weeds, such as bermudagrass (*Cynodon dactylon*), dallisgrass (*Paspalum dilatatum*), and

* Corresponding author at. Department of Land, Air, and Water Resources, 131 Veihmeyer Hall, University of California, Davis, CA 95616, USA. Tel.: +1 530 752 4953; fax: +1 530 752 1552.

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hairy fleabane (*Conyza bonariensis*), are treated with pre-emergence or post-emergence herbicides. To minimize the yield loss caused by pests, pathogens, and weeds, California almond growers apply a large amount of pesticides; 9.3 million tons of pesticide active ingredients were applied in 2010 (CEPA, 2012). However, the applied pesticides threaten the environment and human health, as evidenced by pesticide detections in groundwater (Kolpin et al., 2000) and surface water (Guo et al., 2007; Hladik et al., 2009).

Various stakeholders have made efforts to reduce or eliminate their uses of the pesticides that are known to harm human health or degrade environmental quality. The United States Environmental Protection Agency (USEPA) regulates pesticide use under the Federal Insecticide, Fungicide, and Rodenticide Act (FIFRA) and the Federal Food, Drug, and Cosmetic Act (FFDCA). Both of these acts were significantly amended by the Food Quality Protection Act of 1996 (FQPA), which set tougher safety standards, including mandatory pesticide reregistration (USEPA, 2012). In addition, integrated pest management (IPM) practices have been promoted to achieve the goal of sustainable pest management. Growers monitor pest pressure and apply pesticides only when necessary, and high-risk pesticides tend to be replaced with reduced-risk pesticides. For instance, organophosphates that were found to deteriorate surface water quality were partially replaced with oils or *Bacillus thuringiensis* (Bt), and hence the majority of insecticides (in terms of mass) applied on almonds in recent years were oils (Epstein et al., 2001; Zhang et al., 2005). To present an overall and more recent picture of the shift of pest management practices for California almonds, it is important to evaluate all the pesticides that are used, which has not been done in previous studies.

Analyzing the data for pesticide use alone is insufficient for evaluating environmental consequences of pest management practices (Barnard et al., 1997), thus numerous pesticide risk indicators considering pesticide effects and exposure have been developed around the world (Bockstaller et al., 2009), including PRoMPT (Whelan et al., 2007), SPIDER (Renaud et al., 2008; Renaud and Brown, 2008), EPRIP (Trevisan et al., 2009), and I-Phy (Lindahl and Bockstaller, 2012). These indicators vary in methodologies, input data requirements, indicator outputs, and applicable scales. Several indicator comparison studies have been carried out to identify ideal indicators for different purposes (Maud et al., 2001; Reus et al., 2002; Stenrod et al., 2008), but they have failed to reach clear agreements. In recent years, along with the advancement of the Geographic Information System (GIS) software techniques and accumulation of environmental data, pesticide risk indicators have become closely integrated with GIS for preparing site-specific environmental condition data and presenting risk maps (e.g., Centofanti et al., 2008; Sala et al., 2010; Schriever and Liess, 2007; Vaj et al., 2011).

Yet, two obstacles exist in applied pesticide risk evaluation: (1) the shortage of real pesticide application data; and (2) the lack of a suitable pesticide risk indicator equipped with extensive data of pesticide properties and environmental conditions. This study overcame these two obstacles with the Pesticide Use Reporting (PUR) database (CEPA, 2012) and the Pesticide Use Risk Evaluation (PURE) indicator (Zhan and Zhang, 2012). The PUR database has comprehensively recorded temporal and spatial data for agricultural pesticide use in California, USA since 1990. The PURE indicator was specifically developed for California agricultural pesticide use, and evaluates pesticide's risks to surface water, groundwater, soil, and air, by considering pesticide properties and on-site environmental conditions. The PURE indicator was validated against surface water monitoring data (Zhan and Zhang, 2012) and was evaluated with a sensitivity analysis (Zhan and Zhang, 2013).

This study provides the first comprehensive analysis of overall pesticide use for a crop, along with risk evaluation by a pesticide risk indicator. The goal is to evaluate the past performance of pest management in California almonds. The specific objectives are: (1) to characterize the spatial and temporal patterns of pesticide use; and (2) to analyze the spatial and temporal patterns of pesticide environmental risks. The results and conclusions are expected to reflect the outcome of California almond stakeholders' efforts towards sustainable pest management and to provide suggestions for prioritizing pest management practices.

2. Materials and methods

2.1. Study area

The Central Valley, where almost all of the almonds in California were cultivated, was selected as the study area (Fig. 1a). The study

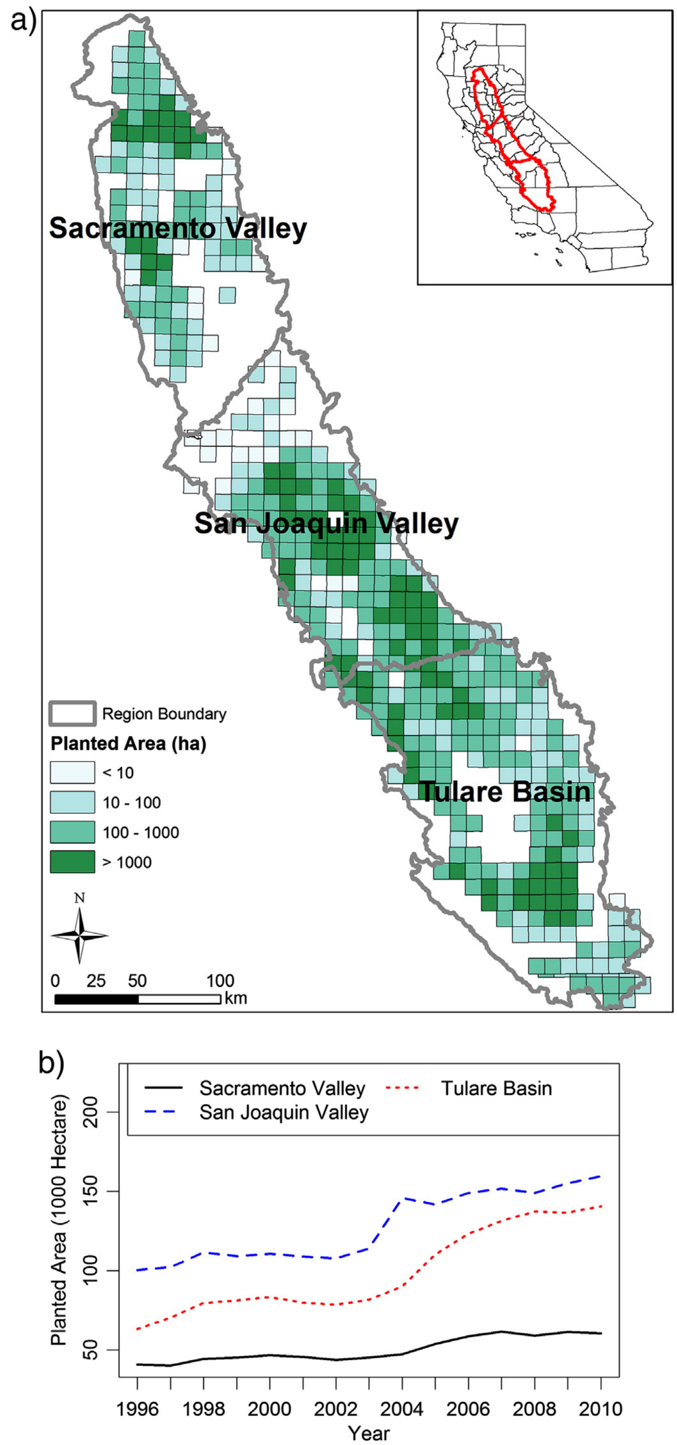


Fig. 1. Spatial and temporal patterns of the almond planted areas from 1996 to 2010 in the Central Valley, California, USA. (a) The average annual planted areas at township (~9.7 × 9.7 km^2) level, and (b) the annual planted areas for each region.

area was divided according to convention into three regions from north to south in the state: the Sacramento Valley (SAC), the San Joaquin Valley (SJQ), and the Tulare Basin (TUL). The average annual planted area of almonds in California from 1996 to 2010 was 277,000 ha, which was composed of 50,000 ha in SAC, 128,000 ha in SJQ, and 99,000 ha in TUL. Almonds are the most densely cultivated in central SJQ, south TUL, and north SAC (Fig. 1a). The planted areas increased in all three regions from 1996 to 2010, with a sharp increase from 2003 to 2007 (Fig. 1b). The three regions have somewhat different climatic conditions. From north to south, temperature increases while humidity decreases, resulting in different pest patterns and pest management practices.

The environmental conditions, including the climatic conditions, soil properties, and other data for environmental factors were compiled from various public data sources. The climatic conditions were obtained from the California Irrigation Management Information System (CIMIS) (CDWR, 2010). The soil properties were extracted from the Soil Survey Geographic (SSURGO) and the State Soil Geographic (STATSGO) databases (NRCS, 2008). The ground slope was calculated from a digital elevation model (DEM) database (NRCS, 2008). The groundwater depth was interpolated from the USGS groundwater monitoring data (USGS, 2010), and the farmland distance to surface water was calculated from the Cal-Atlas stream map (Cal-Atlas, 2008).

2.2. Pesticide use data and pesticide properties

The pesticide use data for California almonds from 1996 to 2010 were queried from the PUR database maintained by CDPR (CEPA, 2012). Nearly 2 million pesticide application records were retrieved, each including the application date and spatial section (~1.6×1.6 km^2) (USDI, 2009). This study took all possible pesticides into account, with a focus on the main pesticide categories of insecticides, fungicides, herbicides, and fumigants, which represented 139, 76, 76, and 8 pesticide AIs (active ingredients) (Table A1), respectively. Eleven AIs (e.g., sulfur) were classified as insecticides as well as fungicides. Furthermore, two highly concerned chemical groups of insecticides – organophosphate and pyrethroids (Table A2) – were also analyzed as pesticide categories. The annual field-level pesticide use intensity ($UI = \Sigma$(pesticide use amount) / field area; unit: kg/ha) was summarized by individual AIs, pesticide categories, and all pesticides. Then the field-level *UI*s were aggregated to township (~9.66×9.66 km^2) (USDI, 2009), region, and state levels using the area-weighted-mean approach.

The product- and AI-level pesticide properties were obtained from several data sources. The product-level properties, including the emission potential (*EP*) and percentage of AI, were from the pesticide product/label database maintained by CDPR (CEPA, 2010). The AI-level properties include chemical, physical, and toxicity properties. Specifically, the sorption coefficient (K_{OC}), the Henry's law constant (K_H), the aerobic (DT_{SO}) and anaerobic (DT_{SA}) half-life in soil, the half-life in water (DT_W), the maximum acute (LEC_A) and chronic ($NOEC_A$) toxicity to aquatic organisms (including fish, *Daphnia*, and algae), the acute (LC_W) and chronic ($NOEC_W$) toxicity to earthworms, and the acceptable daily intake (*ADI*) were obtained from the ChemPest database (CEPA, 2009), the Pesticide Properties Database (PPDB) (PPDB, 2012), and the Pesticide Action Network (PAN) (Kegley et al., 2011) in order of preference.

2.3. Pesticide risk indicator

On the basis of the pesticide properties and local environmental conditions, the PURE indicator (Zhan and Zhang, 2012) was used to evaluate the risk values of each pesticide application to surface water (R_W), groundwater (R_G), soil (R_S), and air (R_A).

Firstly, R_W was the maximum of the short-term and long-term risk values for surface water, which were the ratios of the predicted short-term (PEC_{WS}) and long-term (PEC_{WL}) pesticide concentrations loaded to surface water to the maximum acute and chronic pesticide toxicities, respectively, to the aquatic organisms (including fish, *Daphnia*, and algae). PEC_{WS} was determined by the pesticide drift process modeled with the Drift Calculator (FOCUS, 2001) and the pesticide runoff process using the SCS curve number method (SCS, 1972). PEC_{WL} was the average concentration during the 21 days (the typical period for measuring the chronic toxicity) after application.

Secondly, R_G was the ratio of the predicted pesticide concentration leaching to groundwater (PEC_G) to *ADI*. The adapted attenuation factor method originally proposed by Rao et al. (1985) was used to calculate PEC_G, where pesticide degradation, convection, and dispersion were taken into account.

Thirdly, R_S, similar to R_W, was the maximum of the short-term and long-term risk scores for soil, which were the ratios of the predicted short-term (PEC_{SS}) and long-term (PEC_{SL}) pesticide concentrations in topsoil to the acute and chronic pesticide toxicities to earthworms, respectively. PEC_{SS} was determined by the amount of pesticide reaching the ground right after the pesticide application, and hence PEC_{SL} was the average concentration in topsoil during the 21 days after application.

Finally, R_A was the product of the pesticide application rate (*RATE*), the *EP*, and the application method adjustment factor (*AMAF*). For a pesticide product containing multiple AIs, the product-level R_A was assigned to each AI in proportion to their mass percentages in that product.

As the four types of risk values were calculated for different environmental compartments, they cannot be compared with each other. Similar to *UI*, the annual field-level pesticide risk intensities (*RI*; unit: R/ha) were also summarized by AI, pesticide categories, and all pesticides. $RI_i = \Sigma$(pesticide risk values) / field area, where $i = W, G, S$, or A, which represent surface water, groundwater, soil, and air, respectively. Then the field-level *RI*s were aggregated to township (~9.66×9.66 km^2) (USDI, 2009), region, and state levels.

2.4. Trend analysis and spatial mapping

The trend analysis and spatial mapping of *UI* and *RI* were performed in R (R Development Core Team, 2013), which is a free and versatile computation platform. Trends were detected with the Mann–Kendall trend test (Mann, 1945) implemented in package Kendall (McLeod, 2011), and slopes were calculated by the Theil–Sen estimator (Sen, 1968) implemented in package zyp (Bronaugh and Werner, 2013). The combination of the Mann–Kendall method and the Theil–Sen estimator is robust and widely used for analyzing environmental time-series data (Helsel and Hirsch, 2002). Significance was considered as $p < 0.1$. In addition, the *UI* and *RI* were mapped at township level (~9.7×9.7 km^2) (USDI, 2009) by using packages rgdal (Bivand et al., 2013) and sp (Pebesma and Bivand, 2005).

3. Results

3.1. Pesticide use intensity (UI)

Between 1996 and 2010, the state average annual *UI*s of insecticides, fungicides, herbicides, and fumigants were 17.00 kg/ha, 4.05 kg/ha, 3.21 kg/ha, and 1.09 kg/ha, respectively (Table 1); and the average annual *UI*s of organophosphates and pyrethroids were 0.98 kg/ha and 0.06 kg/ha, respectively (Table A3). At the regional level, the average annual *UI* of insecticides and herbicides both increased from north to south, the fungicide *UI* decreased from north to south, and the fumigant *UI* was the highest in the middle region. The same latitudinal patterns were observed on the township scale, with smooth spatial transition (Fig. 2). Furthermore, the regional average annual *UI* of organophosphates in TUL was about three times as that in SAC or SJQ, while the regional average annual *UI* of pyrethroids in SAC was less than a half of that in SJQ or TUL (Table A3). The spatial maps (Fig. A1a and A1b) confirm the regional *UI* patterns of organophosphates and pyrethroids.

Table 1
Use intensities (*UI*) by pesticide use category for California almonds from 1996 to 2010, with statewide top-five pesticides in each use category.

Use category/pesticide	State		SAC		SJQ		TUL	
	Mean	Slope	Mean	Slope	Mean	Slope	Mean	Slope
Insecticides	17.00	−0.22	7.63	−0.83**	13.28	−0.28·	26.54	0.26
Petroleum oil, unclassified	10.03	−0.22*	2.81	−0.43**	6.79	−0.27*	17.98	−0.01
Mineral oil	4.59	0.20*	2.60	−0.37**	4.70	0.13	5.28	0.43**
Sulfur	0.47	0.00	0.96	0.11*	0.44	−0.04**	0.25	0.01
Propargite	0.46	−0.06**	0.24	−0.01·	0.37	−0.05**	0.73	−0.12**
Chlorpyrifos	0.45	−0.01	0.23	0.01*	0.37	−0.02**	0.67	−0.01
Fungicides	4.05	−0.28**	5.34	−0.05	4.46	−0.41**	2.83	−0.22**
Ziram	0.95	−0.08**	2.06	−0.04	0.69	−0.09**	0.73	−0.08**
Copper hydroxide	0.81	−0.08**	0.29	−0.02**	1.15	−0.11**	0.63	−0.04*
Captan	0.48	−0.08**	0.64	−0.05*	0.58	−0.10**	0.25	−0.05**
Sulfur	0.47	0.00	0.96	0.11*	0.44	−0.04**	0.25	0.01
Maneb	0.35	−0.06**	0.54	−0.05*	0.37	−0.07**	0.22	−0.04**
Herbicides	3.21	0.17**	2.94	0.13**	3.10	0.14**	3.45	0.22**
Glyphosate, isopropylamine salt	1.24	0.00	1.40	−0.01	1.21	0.02	1.21	−0.01
Paraquat dichloride	0.46	0.03*	0.31	0.05**	0.37	0.02·	0.65	0.04*
Glyphosate, potassium salt	0.26	0.04**	0.16	0.02*	0.20	0.04**	0.36	0.05**
Oryzalin	0.23	−0.00	0.34	0.01	0.24	−0.01	0.18	−0.01
Oxyfluorfen	0.22	0.01*	0.15	0.01**	0.21	0.01*	0.26	0.01*
Fumigants	1.09	−0.02	0.11	2E-04	1.51	−0.01	1.06	−0.02
1,3-Dichloropropene	0.77	0.07	0.05	0.00	1.06	0.09·	0.76	0.06*
Methyl bromide	0.26	−0.04**	0.04	−0.01**	0.33	−0.06**	0.30	−0.04**
Sodium tetrathiocarbonate	0.03	0.00	3E-3	0.00	0.06	0.00	7E-5	0.00
Metam-sodium	0.02	−1E-3**	7E-6	0.00	0.03	−3E-3**	0.01	0.00
Chloropicrin	0.01	−0.00	0.01	3E-4	0.01	−0.00	4E-3	−0.00

SAC: the Sacramento Valley; SJQ: the San Joaquin Valley; TUL: the Tulare Basin. Mean: average annual use intensity (kg/ha). Slope: the Theil–Sen slope (kg/ha/year) with significance level calculated by the Mann–Kendall trend test. ** $p < 0.01$; * $p < 0.05$; · $p < 0.1$.

The statewide *UI* of fungicides and herbicides significantly decreased and increased, respectively, while the statewide *UI* of insecticides and fumigants showed no trends (Table 1). All the *UI* trends at regional level were consistent with the trends at state level, except for the increased trends of the insecticide *UI* in SAC and SJQ, and the lack of trends observed for the fungicide *UI* in SAC. Among insecticides, the organophosphate *UI* significantly decreased in all regions, while the pyrethroid *UI* significantly increased only in TUL (Table A3). Figs. 3 and A1 show specific areas with significant *UI* changes. Increase of herbicide *UI* occurred across the whole Central Valley, while decrease of fungicide *UI* spread over SJQ and TUL. Decrease of organophosphate *UI* occurred over the whole Central Valley, while increase of pyrethroid *UI* mainly took place in TUL. Figs. 4 and A2 show the yearly *UI* by pesticide use categories at regional level. The decrease of insecticide *UI* in SAC and SJQ mainly happened from 1996 to 2000; and the organophosphate *UI* continuously decreased in all regions, while the pyrethroid *UI* kept steady in most years but increased a lot in SJQ and TUL in 2010. In SJQ and TUL the fungicide *UI* decreased consistently, while in SAC it decreased initially until 2001, followed by a period of rapid increase. The herbicide *UI* in all three regions slightly decreased from 1996 to 2001 and then increased dramatically. The fumigant *UI* kept steady in SAC but varied widely in SJQ and TUL.

The statewide top-five-*UI* pesticides by use category accounted for 94%, 76%, 75%, and 99.6% of the *UI* of insecticides, fungicides, herbicides, and fumigants, respectively (Table 1). For insecticides, "petroleum oil, unclassified" and mineral oil accounted for the majority of the insecticide *UI* and the total pesticide *UI*. Most of the top-five insecticides either significantly decreased or showed no trend in their *UI*. For fungicides, SAC heavily relied on ziram and sulfur, SJQ used copper hydroxide the most intensively, and TUL tended to have even applications of ziram and copper hydroxide. Most of the *UI* of the top-five fungicides decreased significantly. For herbicides, "glyphosate, isopropylamine salt" was the dominant herbicide in all regions and showed no trends. The *UI* of the other top herbicides except oryzalin increased significantly in all regions. For fumigants, 1,3-dichloropropene and methyl bromide accounted for 94% of statewide fumigant uses. The former increased significantly in SJQ and TUL, while the latter significantly decreased in all regions.

3.2. Pesticide risk intensity (RI)

Between 1996 and 2010, the state average annual RI_W, RI_G, RI_S, and RI_A were 81 R/ha, 98 R/ha, 182 R/ha, and 90 R/ha, respectively (Table 2). Organophosphates contributed 45%, 9%, 17%, and 16% of the total RI_W, RI_G, RI_S, and RI_A, respectively, while pyrethroids contributed 11%, 0%, 5%, and 2%, respectively (Table A3). At regional level, the average annual RI_W, RI_S, and RI_A increased from north to south, while RI_G decreased from north to south. The spatial gradients of *RI* on the township level were less clear than those of *UI*, and the high-*RI* areas were more clustered (Fig. 5). Northern SAC and southern TUL had clustered areas of both high RI_W and high RI_G, with a few high-RI_W areas scattered in middle SJQ. In contrast, high-RI_S and high-RI_A areas were located in middle SJQ and northeastern TUL. Moreover, the risk maps of organophosphates (Fig. A3) are similar to those of all pesticides (Fig. 5) to a certain extent. For pyrethroids, RI_W and RI_S were the only concerns: the high-RI_W areas scattered across the whole Central Valley, while the high-RI_S areas clustered in TUL (Fig. A6).

The statewide RI_G and RI_S significantly decreased, while the statewide RI_W and RI_A didn't show trends (Table 2). Regionally in SAC, none of the risk types showed any trends. In SJQ, all risk types significantly decreased except RI_A, which did not have trends in any region. In TUL, both RI_G and RI_S significantly decreased. Fig. 6 shows specific areas with significant *RI* changes. In SAC, the areas where *RI* significantly increased/decreased were scattered. In SJQ RI_W, RI_G and RI_S significantly decreased across large areas. In TUL, both RI_W and RI_S significantly decreased in a clustered area located in the south, and RI_A significantly increased at the west edge. More temporally specific, the RI_W of TUL largely decreased from 1996 to 2002 and then bounced back in 2006, while the RI_W of SAC and SJQ were relatively steady (Fig. 7a). The RI_G of SJQ and TUL showed two stages, separated in 2004 and 2000, respectively, while the RI_G of SAC did not show a clear trend (Fig. 7b). The RI_S of all regions decreased sharply from 1996 to 2000 or 2001, and then rose slowly till 2006, followed by a short decreasing period (Fig. 7c). The RI_A of all three regions showed similar temporal patterns as RI_S (i.e., decrease–increase–decrease) (Fig. 7d). The difference is that RI_A recovered to the initial level at the end of the increase stage while RI_S only partially recovered.

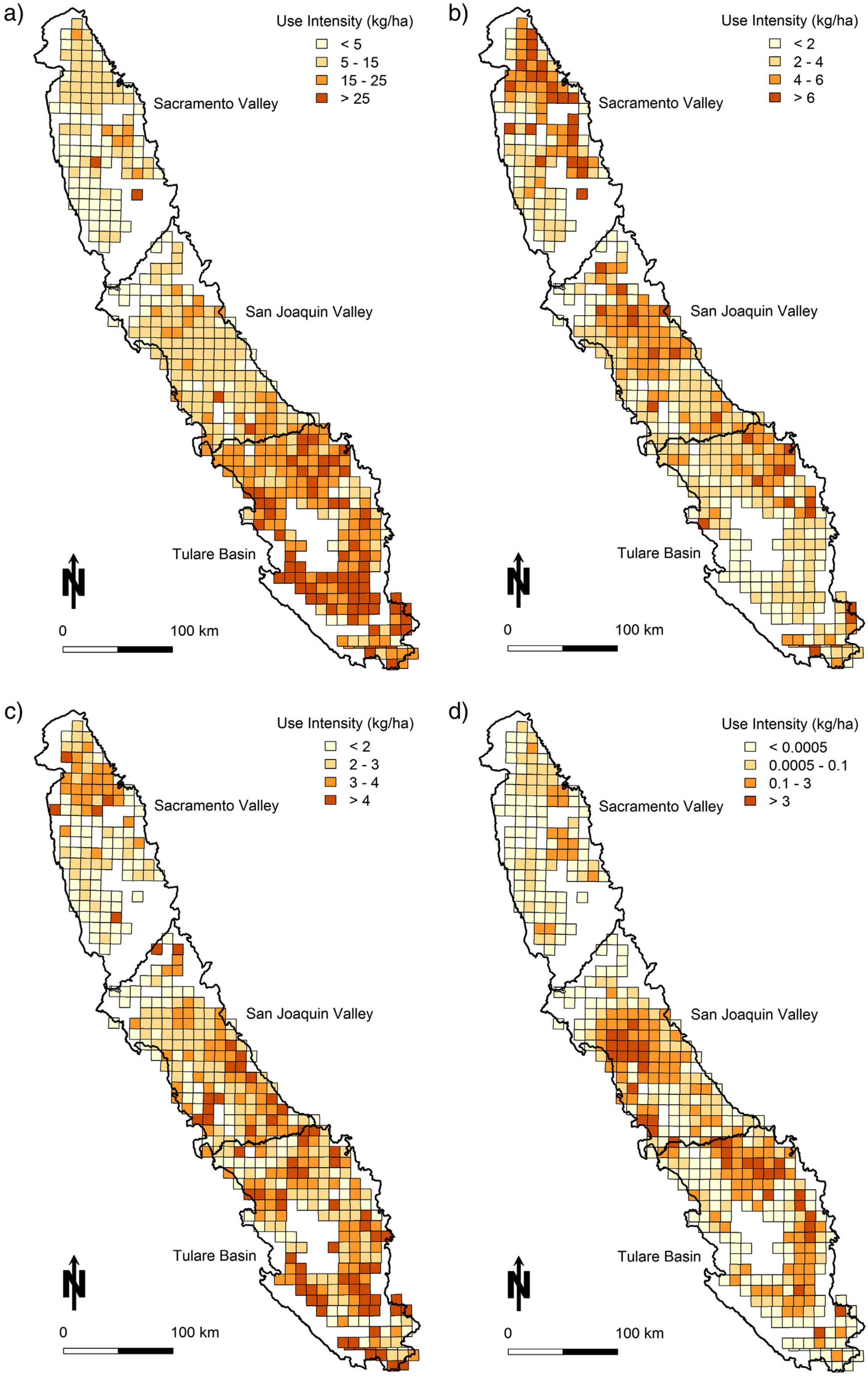


Fig. 2. Spatial patterns of the average annual use intensities (*UI*; kg/ha) of (a) insecticides, (b) fungicides, (c) herbicides, and (d) fumigants for California almonds from 1996 to 2010.

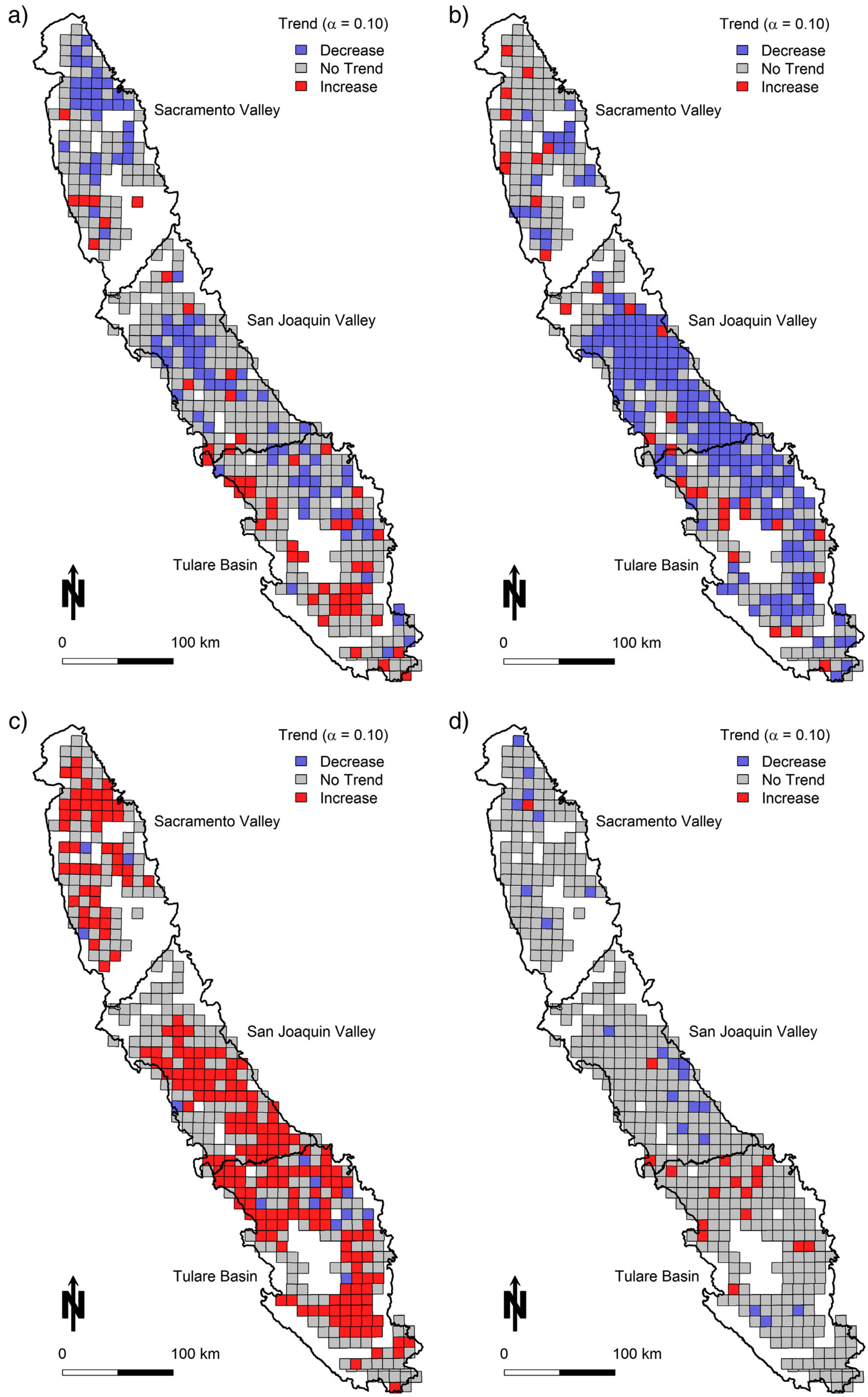


Fig. 3. Temporal trends of the annual use intensities (*UI*; kg/ha) of (a) insecticides, (b) fungicides, (c) herbicides, and (d) fumigants for California almonds from 1996 to 2010.

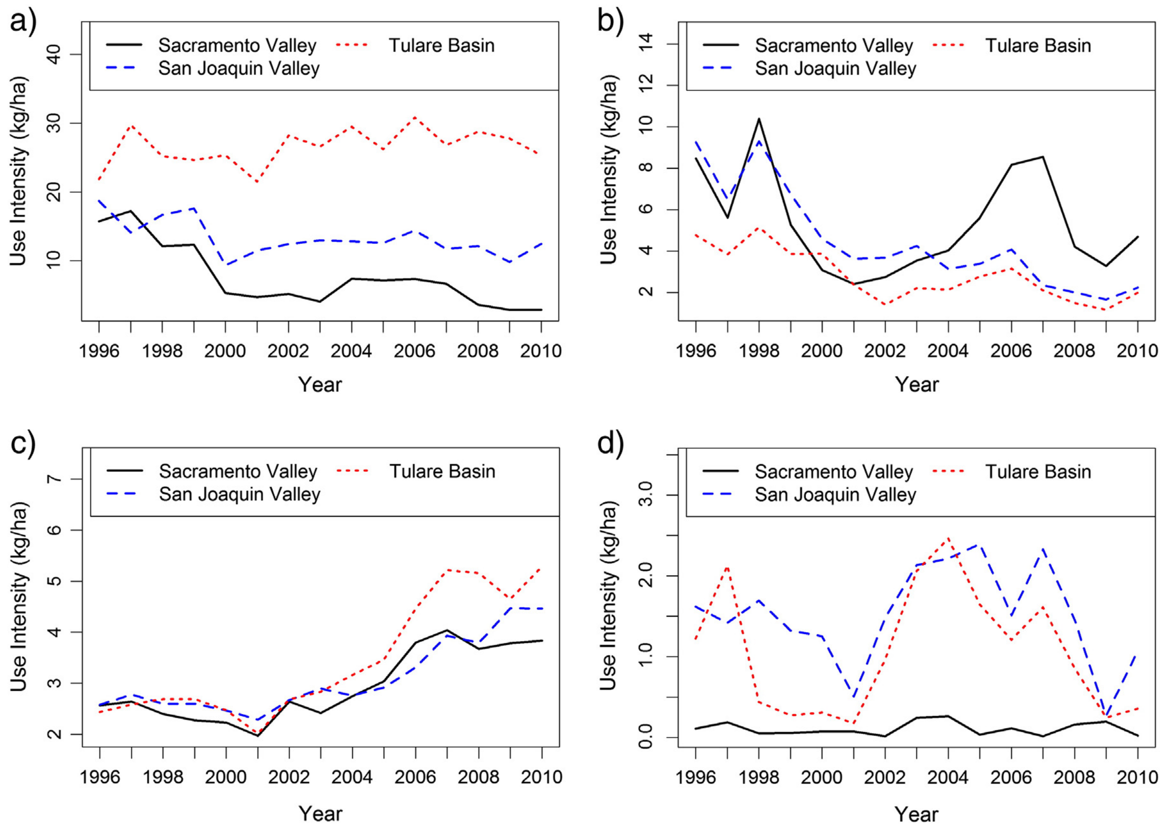


Fig. 4. Regional annual use intensities (*UI*; kg/ha) of (a) insecticides, (b) fungicides, (c) herbicides, and (d) fumigants for California almonds from 1996 to 2010.

The temporal patterns of *RI* were quite different between organophosphates and pyrethroids. For organophosphates, RI_W significantly decreased only in SJQ, RI_G and RI_S significantly decreased in all regions, and RI_A significantly decreased in SJQ and TUL (Table A3). Fig. A4 shows the *RI* trends for organophosphates at township level. RI_W significantly decreased in large areas of SJQ and in a small portion of SAC and TUL, which was reflected in the basin-level trend of RI_W. RI_A had a similar spatial pattern with RI_W, but significantly decreased in larger areas in TUL. The temporal trends of RI_G and RI_S showed a similar spatial pattern. Fig. A5 shows the yearly change of *RI* at regional level. The decrease of RI_S was apparent in all regions, one peak for RI_W and RI_A of TUL occurred in 2006, and one peak for RI_G of SAC appeared in 1999. For pyrethroids (RI_G and RI_A were negligible), RI_W significantly increased only in TUL, while RI_S significantly increased in SJQ and TUL. RI_W significantly decreased in small areas of northern SAC and middle SJQ, and significantly increased in large areas of TUL (Fig. A7a). RI_S significantly increased across the whole Central Valley (Fig. A7c). The RI_W of TUL increased continuously and rapidly from 2005 to 2010 (Fig. A8a). The RI_S of TUL increased slowly but steadily from the beginning of the study period, and increased much faster from 2005 (Fig. A8c).

The statewide top-five pesticides by risk type accounted for 80%, 86%, 44%, and 66% of RI_W, RI_G, RI_S, and RI_A, respectively (Table 2). For RI_W, ziram, copper hydroxide, and chlorpyrifos were the top contributors in SAC, SJQ, and TUL, respectively. The RI_W from ziram significantly decreased in SJQ only. The RI_W from copper hydroxide significantly decreased in all regions. In contrast, the RI_W from chlorpyrifos didn't show a trend in any region. For RI_G, oxyfluorfen and simazine were the main contributors in all regions. The RI_G of all the top-five pesticides significantly decreased in all regions, except for the RI_G of oxyfluorfen which significantly increased in SAC and showed no trend in the other two regions. For RI_S, most of the top-five contributors significantly decreased in all regions, except for the RI_S of 1,3-dichloropropene that significantly increased in SJQ and TUL, and the RI_S of mineral oil that significantly increased in TUL. For RI_A, 1,3-dichloroprone was the top contributor and significantly increased in SJQ and TUL, but only accounted for 3% of RI_A in SAC and did not show a trend.

4. Discussion

4.1. Spatial patterns of pesticide use and risk

4.1.1. Pesticide use intensity (UI)

The spatial patterns of *UI*, mainly caused by spatially different pest pressures, were highly associated with climate conditions and farming activities. In the study area (i.e., the Central Valley, California), the temperature increases from north to south while the humidity decreases from north to south. In southern areas, more insecticides (including more organophosphates, pyrethroids, and other insecticides) and herbicides were applied, indicating that higher temperatures with sufficient water supply via irrigation favored the growth of insects and weeds. In contrast, fungi prefer cool and moist environments, which resulted in more intensive fungicide use in northern areas. In addition, the spatial pattern of fumigant *UI* was mainly due to farming activities. Fumigants were mainly used to treat soil-borne diseases when almond fields were newly cultivated or replanted (CEPA, 2008), as well as for post-harvest pests. The rapid expansion of almond fields in SJQ and TUL resulted in higher average annual fumigant *UI* in these two regions (Fig. 2d).

In addition to the general spatial patterns of the *UI*s, the existence of clustered high-*UI* areas demonstrates location specificity, which was likely associated with local pest pressure (including insects, pathogens, and weeds). The areas with denser almond fields (Fig. 1a) seemed to suffer higher pest pressures reflected in higher pesticide *UI*. In northern SAC more intense fungicides and herbicides were applied. In central SJQ higher *UI*s of fungicides and fumigants were observed. In southern TUL insecticides and herbicides were used more intensively. The spatial correlation between the cultivation density and the pest pressure might be induced by pest dispersion ranges. That is, closer distances among almond fields facilitated the dispersion and subsequent burst

Table 2
Risk intensities (*RI*) for California almonds from 1996 to 2010, with statewide top-five pesticides for each risk type.

Risk/pesticide	State		SAC		SJQ		TUL	
	Mean	Slope	Mean	Slope	Mean	Slope	Mean	Slope
Surface water	81	−0.8	57	−2.0	67	−2.4·	111	−1.1
Chlorpyrifos	31	1.2	9	−0.03	21	−0.7	54	2.3
Copper hydroxide	18	−1.7**	10	−0.4**	22	−1.6*	18	−1.5
Ziram	7	−0.7*	19	−0.8	5	−0.7**	4	−0.3
Permethrin	4	−0.3·	2	−0.01	4	−0.3*	6	−0.2
Chloropicrin	4	−0.2	9	0.2*	5	−0.6**	1	−0.04
Groundwater	98	−4.1**	185	−2.5	90	−4.5**	64	−3.0·
Oxyfluorfen	37	1.0	83	6.8**	18	0.3	39	−0.5
Simazine	29	−1.9**	26	−1.6**	48	−2.8*	6	−0.5**
Diazinon	7	−0.7**	32	−3.3**	1	−0.1*	1	−0.02**
Norflurazon	6	−0.9**	7	−0.8**	8	−0.9**	3	−0.3**
Propargite	5	−0.7**	12	−0.7	3	−0.4**	5	−0.9**
Soil	182	−9.8*	145	−7.1	183	−9.4*	199	−9.7*
Copper hydroxide	22	−2.3**	8	−0.6**	31	−3.1**	17	−1.1*
1,3-Dichloropropene	19	1.4*	1	0.03	25	2.0·	19	1.7*
Ziram	17	−1.5**	39	−1.5	12	−1.6**	12	−1.6**
Methidathion	15	−2.8**	11	−0.9**	9	−1.4**	25	−5.6**
Mineral oil	9	0.1	6	−0.9**	9	0.03	10	0.7**
Air	90	1.7	42	0.2	92	0.6	112	3.0
1,3-Dichloropropene	20	1.8	1	0.1	27	2.2	20	1.7*
Oxyfluorfen	12	0.5	9	0.7*	12	0.3	15	0.5
Chlorpyrifos	12	−0.5	6	0.4	10	−0.6*	17	−0.7
Petroleum oil, unclassified	9	1.0**	1	−0.1*	6	0.6**	17	1.6**
Methyl bromide	7	−1.0**	1	−0.1**	8	−1.4**	7	−0.9**

SAC: the Sacramento Valley; SJQ: the San Joaquin Valley; TUL: the Tulare Basin. Mean: average annual risk intensity (R/ha). Slope: the Theil–Sen slope (R/ha/year) with significance level calculated by the Mann–Kendall trend test. ** $p < 0.01$; * $p < 0.05$; · $p < 0.1$.

of pests. On the other hand, in northeastern TUL where the almond fields were relatively sparser, the *UI*s of insecticides, fungicides, and fumigants were also high. It indicated that some other factors (e.g., farm management practices) influenced the local pesticide *UI* or pest pressure, which requires more investigation in the future.

4.1.2. Pesticide risk intensity (RI)

Compared with *UI*, the spatial patterns of *RI* were affected by more factors, including environmental conditions (e.g., surface water distance and groundwater depth) and pesticide properties. Firstly, the high-RI_W areas were all close to surface water and used pesticides highly toxic to aquatic organisms (e.g., chlorpyrifos). The risk to surface water was the main environmental concern of treating insects with organophosphates and pyrethroids, which are generally highly toxic to aquatic organisms, moderately persistent, and soluble in water for organophosphates or bound to sediment for pyrethroids. The high-RI_W areas in south TUL were near the Kern River and the Poso Creek, with intensive applications of organophosphates and pyrethroids. Similarly, the high-RI_W areas in north SAC were close to the Sacramento River, and fungicides were applied intensively in these areas. Additionally, the high-RI_W areas scattered in central SJQ were near the San Joaquin River, with intensive use of insecticides and fungicides.

Secondly, high-RI_G areas were mainly caused by the combined effects of high herbicide use and shallow groundwater level. Herbicides are usually mobile in soil as indicated by low soil sorption coefficients (K_{OC}). In a national groundwater survey, most of the detected pesticides were herbicides in areas with shallow groundwater level (Kolpin et al., 2000). Both north SAC and southwestern TUL had high-RI_G areas. As expected, in these areas the groundwater level was shallow and herbicide *UI* was high. Contrarily, in the areas near the boundary between SJQ and TUL where the groundwater level was deep, although the herbicide *UI* was also high, the RI_G was not as high as that in north SAC and southwestern TUL.

Finally, the spatial patterns of RI_S and RI_A both were largely affected by total pesticide *UI*, while the pesticide toxicities to earthworms played an important role in RI_S and the emission potentials were important to RI_A. The findings were consistent with the sensitivity analysis on the PURE indicator (Zhan and Zhang, 2013). There existed high-RI_S and high-RI_A areas in central SJQ and northeastern TUL, which was mainly caused by high fumigant *UI* in those areas. In addition, the high-RI_S areas in northern SAC were largely due to the intense use of fungicides.

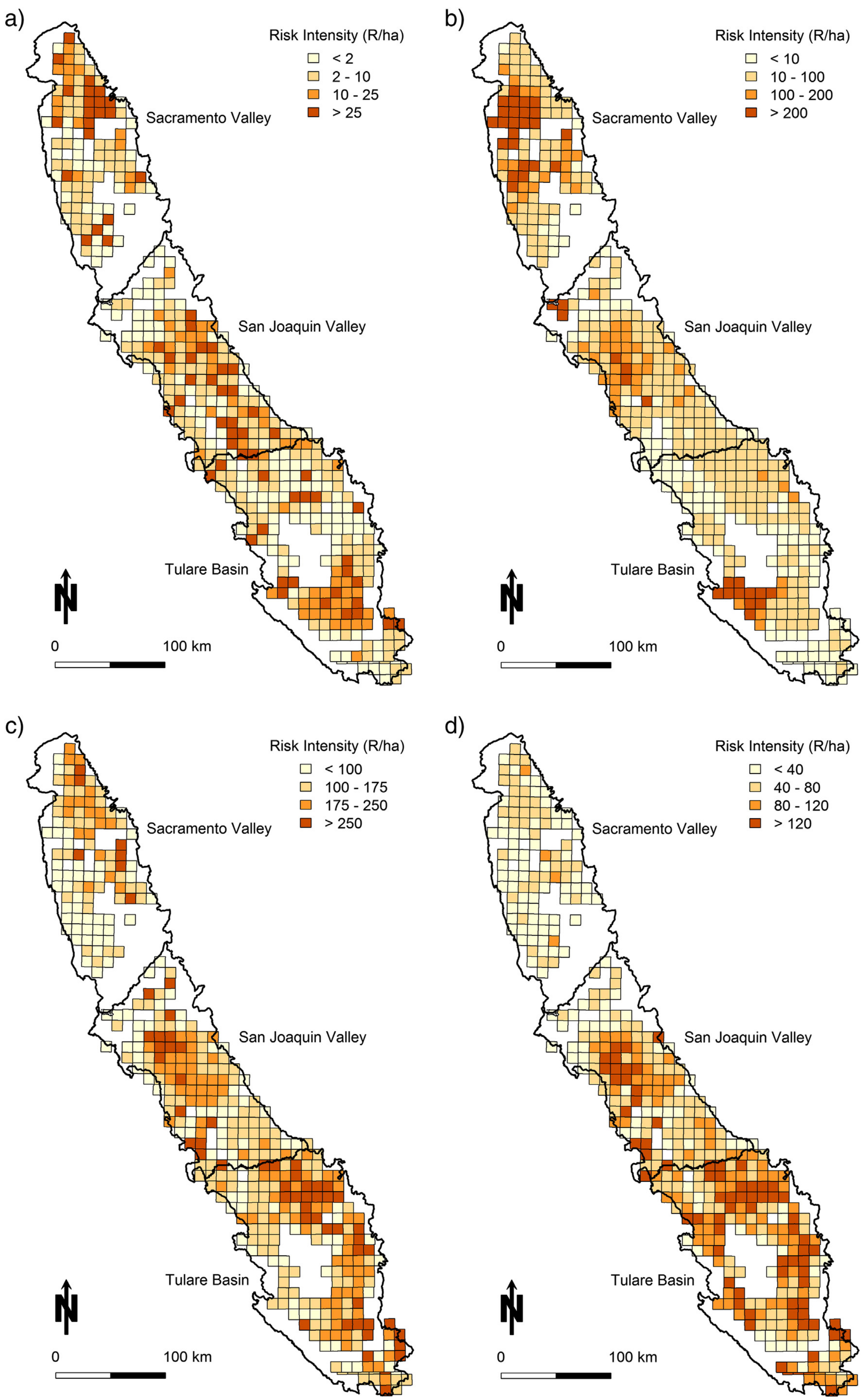
4.2. Temporal patterns of pesticide use and risk

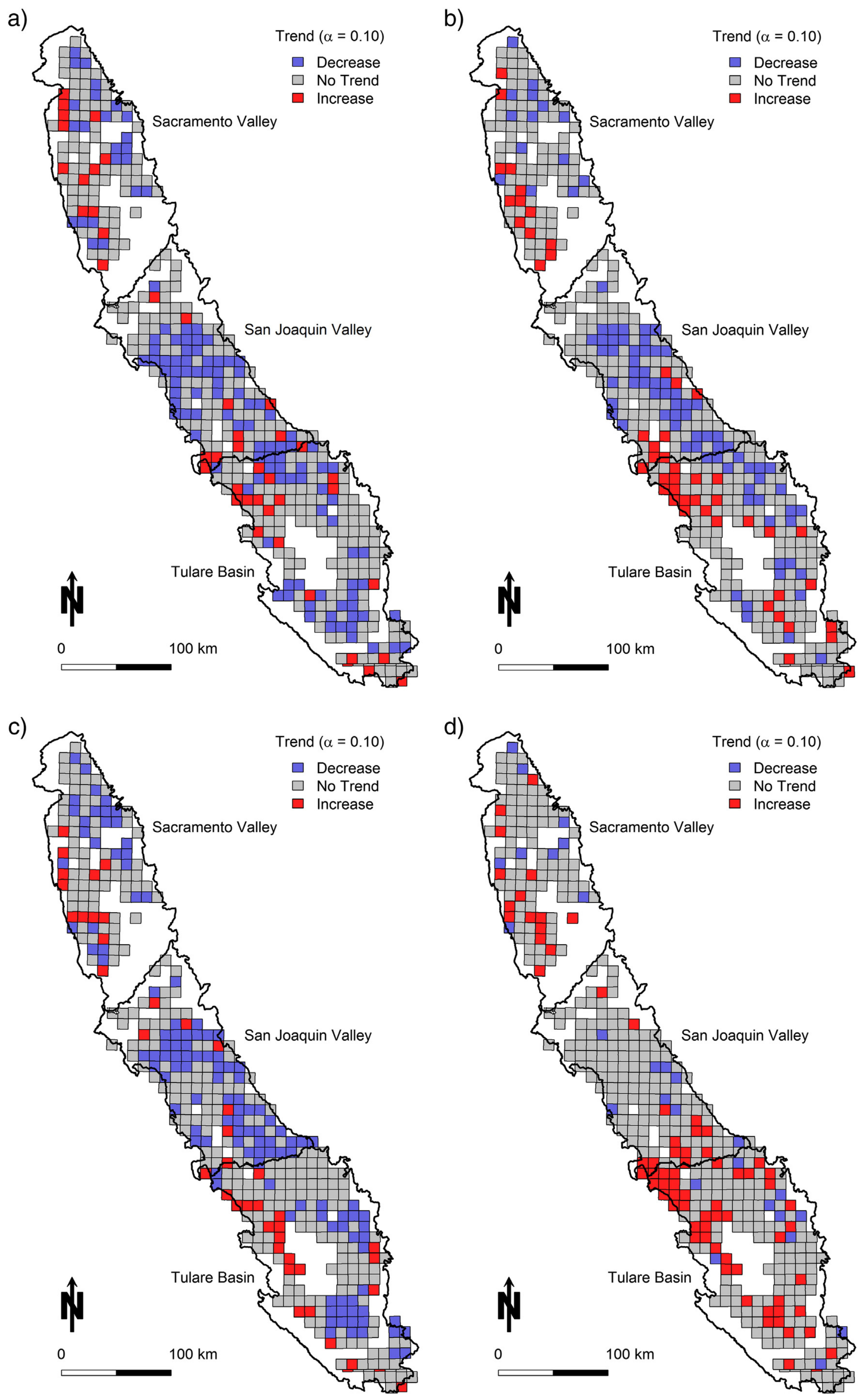
The temporal patterns of *UI* and *RI* were the results of the shift of pest management practices led by governmental regulations, the integrated pest management (IPM) promotion, availabilities of new pesticides, and phasing-out of pesticides known to pose highly adverse impacts on human health and environment.

4.2.1. Insecticides

Insecticide use was under stringent regulation, which largely affected the temporal patterns of insecticide *UI* and the associated *RI*. Although propargite and chlorpyrifos were used much less than the top-three-*UI* insecticides (i.e., "petroleum oil, unclassified", mineral oil, and sulfur), their risks to human and ecosystem health were much higher. Propargite is known to cause human health problems as a carcinogen and reproductive toxicant (e.g., Mills and Yang, 2007), as well as environmental problems (e.g., Bradford et al., 2010). Therefore, the use of propargite has been restricted by regulations, resulting in the significant decrease in its use in all three regions. In addition, chlorpyrifos has been frequently detected in surface water in California (CEPA, 2007) and is highly toxic to aquatic organisms. It has been on the Clean Water Act 303 (d) list of impaired waterways since 1998 in the Total Maximum Daily Load (TMDL) program (CEPA, 2013). In this risk evaluation, chlorpyrifos was the top RI_W contributor and one of the main RI_A contributors (Table 2). In TUL the elevated RI_W after 2005 was caused by the increased use of chlorpyrifos (Fig. 7a). An important concern is the significantly increased use of chlorpyrifos in SAC (though still lower than the other two regions), which might be due to elevated

Fig. 5. Spatial patterns of the annual risk intensities (R/ha) of (a) surface water risk (RI_W), (b) groundwater risk (RI_G), (c) soil risk (RI_S), and (d) air risk (RI_A) for California almonds from @ 1996 to 2010.


a)
Risk Intensity (R/ha)
< 2
2 - 10
10 - 25
> 25
Sacramento Valley
San Joaquin Valley
Tulare Basin
N
0
100 km
b)
Risk Intensity (R/ha)
< 10
10 - 100
100 - 200
> 200
Sacramento Valley
San Joaquin Valley
Tulare Basin
N
0
100 km
c)
Risk Intensity (R/ha)
< 100
100 - 175
175 - 250
> 250
Sacramento Valley
San Joaquin Valley
Tulare Basin
N
0
100 km
d)
Risk Intensity (R/ha)
< 40
40 - 80
80 - 120
> 120
Sacramento Valley
San Joaquin Valley
Tulare Basin
N
0
100 km


a)
Trend (α = 0.10)
Decrease
No Trend
Increase
Sacramento Valley
San Joaquin Valley
Tulare Basin
N
0
100 km
b)
Trend (α = 0.10)
Decrease
No Trend
Increase
Sacramento Valley
San Joaquin Valley
Tulare Basin
N
0
100 km
c)
Trend (α = 0.10)
Decrease
No Trend
Increase
Sacramento Valley
San Joaquin Valley
Tulare Basin
N
0
100 km
d)
Trend (α = 0.10)
Decrease
No Trend
Increase
Sacramento Valley
San Joaquin Valley
Tulare Basin
N
0
100 km

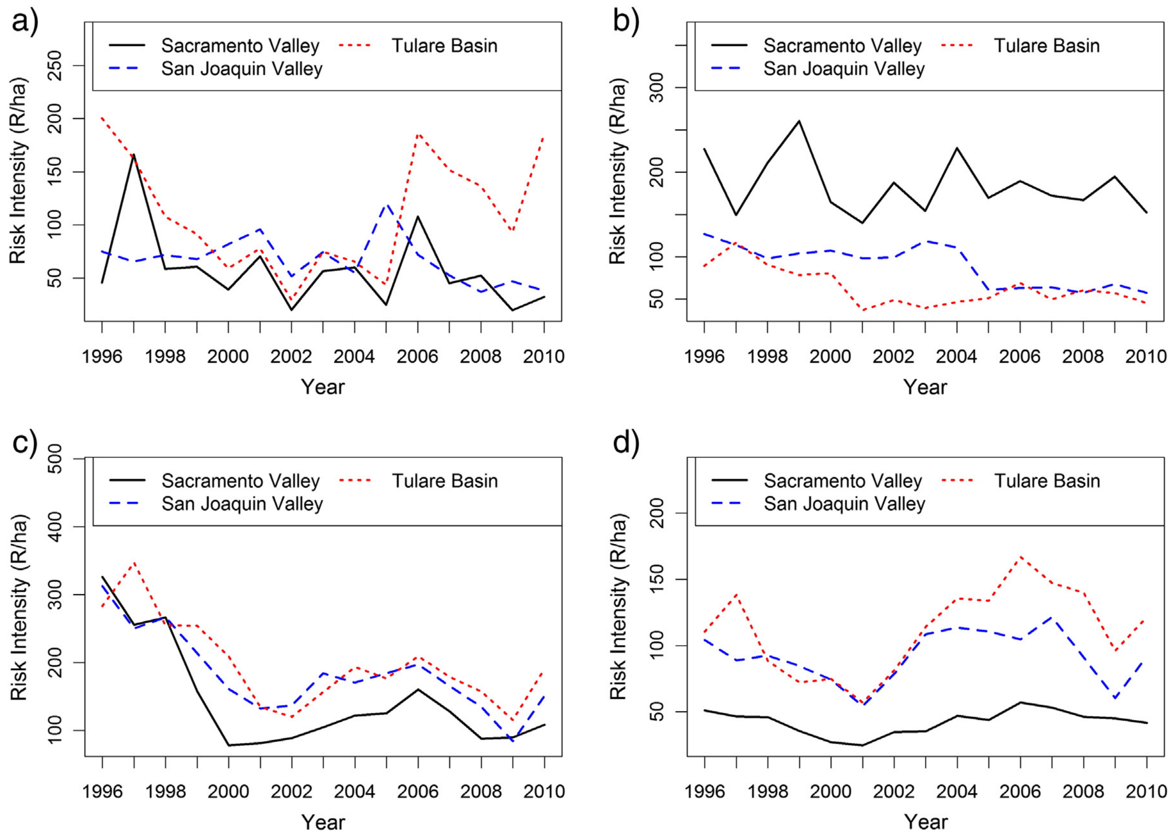


Fig. 7. Regional annual risk intensities (R/ha) of (a) surface water risk (RI_W), (b) groundwater risk (RI_G), (c) soil risk (RI_S), and (d) air risk (RI_A) for California almonds from 1996 to 2010.

insect pressure. Best Management Practices (BMP) (Reichenberger et al., 2007; Zhang and Zhang, 2011) or IPM practices should be promoted in SAC to alleviate the environmental pressure from the chlorpyrifos use.

Besides chlorpyrifos, other organophosphates as well as pyrethroids were also regulation focuses mainly regarding water body impairment. Although the *UI* of chlorpyrifos only significantly decreased in SJQ and even significantly increased in SAC, the *UI* of all organophosphates significantly decreased in all three regions. In other words, a majority of organophosphates significantly decreased statewide, such as the phasing-out of diazinon, naled, and malathion, which resulted in the significant decrease of RI_G, RI_S, and RI_A. Biological control or organically acceptable methods were recommended to replace organophosphate treatments. For instance, *B. thuringiensis* and spinosad were promoted to control peach twig borer (UC IPM, 2012). However, as the *UI* of chlorpyrifos (one of the main RI_W contributor) didn't show a trend in SAC or TUL, the RI_W of organophosphates didn't change significantly in the two regions. In addition, pyrethroids were considered as effective and environmentally-friendly alternatives to organophosphates until they were found to occur in sediment at a high volume, posing risk to water-column and sediment-dwelling creatures (Weston and Lydy, 2010). Considering the presence in sediment, the *UI* of pyrethroids kept steady in SAC and SJQ, but significantly increased in TUL likely due to more intensive insect pressure.

4.2.2. Fungicides

As required by FQPA, all the top-five fungicides except sulfur went through the reregistration process, which might be the main cause of the significant decrease of these fungicides, e.g., the maximum seasonal rate of maneb was reduced from 22.84 to 17.13 kg/ha (USEPA, 2005). Besides governmental regulations, the introduction of new fungicides, such as chlorothalonil and boscalid, also led to the decrease of the main fungicides. The fungicide *UI* increase in SAC from 2001 to 2005 was mainly due to the increased use of sulfur. As ziram and maneb were found to be associated with Parkinson's disease (Wang et al., 2011), stricter regulation on the uses of ziram and maneb is expected in the future. Copper hydroxide was the main RI_W and RI_S contributor. Copper hydroxide is persistent in the field and adversely affects aquatic organisms in the form of soluble copper, which is acutely and chronically toxic to aquatic organisms at low levels (Rice et al., 2006). The quick decreases of the *UI* of copper hydroxide led to the quick decrease of RI_W in TUL from 1996 to 2002 (Fig. 7a) and the quick decrease of RI_S in all regions from 1996 to 2001 (Fig. 7c). The decrease of RI_S was a side-effect of pesticide regulations where soil health was not an important concern. Earthworms, as nontarget beneficial soil organisms, play an important role in soil ecosystems (Das Gupta et al., 2011), but the pesticide risk to them has often been overlooked (Reinecke and Reinecke, 2007). Greater attention should be paid to soil health in the future.

4.2.3. Herbicides

Although the *UI* of herbicides (the main RI_G contributing use category) significantly increased, the RI_G significantly decreased. The steep increase of herbicide *UI* from 2001 to 2010 (Fig. 4c) was possibly due to the shift of weed management practices, the growing problem of weed resistance to glyphosate, or the impacts of climate change (Bloomfield et al., 2006). The resistance to glyphosate was mainly caused by the heavy use of glyphosate for strip spray, which largely replaced pre-emergence herbicides (CEPA, 2005). Rotating herbicides of different modes of action (MoA) is important to mitigate the resistance problem, though growers tend to use the product(s) that are the most economical and/or are

Fig. 6. Temporal trends of the annual risk intensities (R/ha) of (a) surface water risk (RI_W), (b) groundwater risk (RI_G), (c) soil risk (RI_S), and (d) air risk (RI_A) for California almonds from 1996 to 2010.

perceived to be the most effective. On the other hand, the decrease of RI_G was because herbicides that were prone to leach to groundwater (e.g., simazine and norflurazon) were replaced with other herbicides that were less mobile in soil even at larger *UI*. Nevertheless, the phase-out of these herbicides left fewer choices for growers in dealing with the weed resistance problem. Finally, the highest RI_G contributor, oxyfluorfen, was also the second highest RI_A contributor. The RI_A decreasing period from 1996 to 2001 and increasing period from 2001 to 2006 (Fig. 7d) were highly associated with the decreasing and then increasing use of oxyfluorfen during that period. The significantly increasing RI_G for oxyfluorfen in SAC deserves more attention.

4.2.4. Fumigants

The temporal pattern of fumigant *UI* was mainly the mixed result of the annual *UI* of 1,3-dichloropropene and methyl bromide, which both were under strict regulations. Required by the Montreal Protocol in 1993 and regulated under the US Clean Air Act, methyl bromide was phased out due to its effect on ozone depletion (Messenger and Braun, 2000), which resulted in the decreases of fumigant *UI* from 1996 to 2001 in SJQ and TUL. The increases of fumigant *UI* from 2001 to 2004 in SJQ and TUL were due to the increased *UI* of 1,3-dichloropropene, which was heavily used when planting or replanting almonds. In the risk evaluation, the increase of 1,3-dichlorpropene *UI* played an important role in the increase of RI_A from 2001 to 2006. In addition, 1,3-dichloropropone was also a main RI_S contributor, causing the increase of RI_S from 2001 to 2006. Fumigants cause the volatile organic compounds (VOC) problem, which adversely impact human health and environment (Gao et al., 2008). A township cap of 1,3-dichlorpropene (i.e., the total application amount in a township must be below a certain threshold) was implemented to restrict its use (Carpenter et al., 2001). Researchers have been looking for alternatives to methyl bromide while considering both economic costs and effectiveness (Qin et al., 2013; Zasada et al., 2010). It is expected that methyl bromide will be banned completely in the near future, 1,3-dichloropropene will be used more efficiently, and more new fumigants will appear.

4.3. Risk evaluation uncertainties

The uncertainties of this risk evaluation study emerged from the input data and the indicator algorithms. In particular, pesticide property data were compiled from different databases, which might be measured under different conditions. Also, some pesticide properties are sensitive to environmental conditions, but only the measured value under a certain condition was used as the indicator input, such as the soil sorption coefficient (K_{OC}) that is sensitive to soil properties (Weber et al., 2004). In addition, environmental condition data were of uncertainties as well. For instance, the local precipitation data were interpolated from the measured data at meteorological sites using the kriging technique, where prediction uncertainties emerged. Another example is the irrigation data, which were missing and therefore estimated using a water balance model (Snyder et al., 2007). Moreover, in the PURE indicator, the worst-case scenarios and the empirical equations, e.g., the SCS curve number method (SCS, 1972), brought uncertainties to the risk results as well. In the future, uncertainties may be partially quantified under the framework proposed by Refsgaard et al. (2007).

4.4. Implications for past performance and future work

In summary, as the almond yield per area remained stable from 1996 to 2010 (Almond Board of California, 2012), the temporal trends of the pesticide use and risks indicate that the California almond growers have made considerable progress towards sustainable pest management in general. In the future, a grower-level analysis on pesticide use and risk is recommended to identify both effective and environmentally-friendly pest management practices, which should be outreached to more growers of almond and other crops. Meanwhile, more attention should be focused on the intensified use of herbicides and emerging problems of herbicide resistance in California. Also, areas identified on the spatial maps with high or increasing pesticide use/risks need to be investigated in greater detail and validated with monitoring data in the future. Finally, the spatial and temporal analysis methods used here should also be applied to other crops in California or other regions.

Conflict of interest

The authors declare no conflict of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2013.11.022.

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