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Investigating Potential Water Quality Impacts of Fungicides Used to Combat Soybean Rust in Indiana

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Abstract Asian soybean rust (ASR) is a foliar plant disease caused by the fungus *Phakopsora pachyrhizi* that is potentially devastating for US soybean production. It was first detected in soybean fields in the Midwestern US in October 2006 but did not cause any damage to soybean production then because most of that year's crop had been harvested by the time it appeared. In coming years, it is possible that ASR might enter soybean fields in the Midwest during the growing season and cause significant damage. The only current option for managing soybean rust is to use fungicides, many of which have been approved for use on soybeans by the US Environmental Protection Agency under emergency conditions. Since soybean fields traditionally have not received widespread applications of fungicides, it is important to understand the potential environmental impacts of using large quantities of fungicides to combat a potential ASR outbreak. Currently, the impacts of the fungicides used to combat soybean rust on surface and groundwater resources and on "off target" species are not fully known. In this study the National Agricultural Pesticide Risk Analysis hydrologic/water quality model was used to predict fungicide concentrations at edge of field and soil water concentrations at bottom of the root zone as a result of fungicide applications to control soybean rust in Indiana. It was also used to evaluate the likelihood of exceeding threshold chronic exposure concentrations of concern for human and aquatic organism health and identify areas of Indiana that are most vulnerable to contamination by fungicides. The model outputs for the different fungicides show spatial variations of fungicide losses in edge of field runoff and to bottom of root zone soil water or shallow groundwater at 5%, 10%, 25%, and 50% probability of exceedence, indicating that some fungicides may be present in concentrations above threshold values of concern for fish and humans. This provides a basis for developing approaches to minimize potential environmental

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impacts of fungicides, such as prioritizing implementation of best management practices in the most vulnerable areas.

Keywords Soybean rust · Fungicide · Risk assessment · Water quality · NAPRA WWW

1 Introduction

Asian soybean rust (ASR) is a foliar plant disease caused by the fungus *Phakopsora pachyrhizi*. It has been widespread across Asia, Australia, Africa, and South America causing significant soybean losses and increased production costs (Yorinori et al. 2003; Ogle et al. 1979). Soybean rust was first spotted in the continental US in late 2004 in Louisiana (Schneider et al. 2005), and since then, it has been classified as a potentially devastating disease for US soybean production (Sinclair and Hartman 1999) as it spread across the southern states. In 2008, soybean rust was found in 392 counties in 16 states, including Alabama, Arkansas, Florida, Georgia, Illinois, Kentucky, Louisiana, Maryland, Mississippi, Missouri, North Carolina, Oklahoma, South Carolina, Tennessee, Texas, and Virginia according to the US Department of Agriculture (USDA) soybean rust web site that was set up to track the movement of soybean rust in the USA (USDA 2008).

The spread of the disease is controlled significantly by climatic factors; hence, its effects vary considerably by region (Del Ponte et al. 2006). For example, the southern US is more susceptible than other regions to rust invasion because of warmer temperatures, higher relative humidity, and rainfall during the growing season, whereas in the middle part of the country where most US soybean production occurs, the climate is less supportive of infestation (Livingston et al. 2004). Future climate variability and change may thus play a significant role in the spread of the disease.

In 2008, the area under soybean production in the USA was estimated to be 30.3 million hectares, producing approximately 83,300 million kilograms of soybeans (USDA-ERS 2008). The USDA's Economic Research Service (ERS) has estimated that the expected value of net economic losses for the first year of significant soybean rust infestation in the USA would range from $640 million to $1.3 billion, and losses in the following 3–5 years could average $240 million to $2 billion per year, depending on the severity and extent of outbreaks (Daberkow 2004).

Because soybean rust spreads through windborne spores, it can reach Midwest US soybean fields through a south to north aerial dispersal of spores from infected plants (Krupa et al. 2006). Soybean rust was confirmed for the first time in Indiana on Oct 18, 2006 (Purdue University 2006), but there was no major damage as the crops were already mature and most had been harvested. However, if the fungus reaches Indiana and other Midwestern states during the growing season, it can cause significant damage. Indiana soybean production of 7,819 million kilograms in 2006 from 2.3 million hectares ranked fourth in the USA, behind Iowa, Illinois, and Minnesota (USDA-NASS 2006).

Cultural practices such as planting date, row width, and crop rotation sequences have little or no effect on soybean rust, so fungicides are the only option for managing soybean rust until disease-resistant varieties are developed (Shaner et al. 2005). At present, only three fungicides are federally registered for use on soybeans in Indiana: chlorothalonil, azoxystrobin, and pyraclostrobin (Shaner et al. 2005). Eleven additional fungicide active ingredients have been proposed for use by Indiana authorities to protect soybeans against soybean rust through an exemption request under the quarantine provisions of Section 18 of the Federal Insecticide, Fungicide and Rodenticide Act of the Environmental Protection Agency Office of Pesticide Programs (EPA 2005).

Fungicides have been widely used in agriculture to battle diseases in crops like onion (Sutton et al. 1986), potato (Hamm and Clough 1999), peanut (Smith and Littrell 1980), tomato (Chapin et al. 2006; Dillard and Cobb 1997), sugarbeet (Kiewnick et al. 2001), and small grains (Hrivna 2003), as well as in turfgrass (Nelson et al. 2003). However, if used incorrectly they are potentially serious environmental pollutants and can degrade surface water quality in runoff and contaminate groundwater through leaching. Fungicide concentrations in both surface and groundwater can be determined through laboratory experiments and field data collection. Stromqvist and Jarvis (2005) reported that excessive use of the fungicide Iprodione to prevent snow mold in golf course turf areas can result in detectable concentrations of the fungicide in the adjacent aquatic ecosystem through leaching and runoff. Potter et al. (2001) showed that the fungicide chlorothalonil presented a risk to aquatic life as a result of runoff from treated fields in southeastern states. Wu et al. (2005) observed that propiconazole was toxic to aquatic bryophytes at low concentrations.

Computer-based simulation models can also be used to provide useful environmental risk information for some fungicides. Vincelli (2004) conducted computer simulations of fungicide loading in surface water runoff and concluded that the amount of surface water runoff loadings of the fungicides chlorothalonil, propiconazole, and azoxystrobin from golf courses and lawns in Kentucky was over the LC_{50} value of at least one indicator species. Wauchopel et al. (2003) modeled chlorothalonil dissipation after multiple applications in peanuts using the Root Zone Water Quality Model, where model results indicate that under severe rainfall conditions during the application period, significant quantities of the parent compound may be transported by runoff into water sources.

The potential adverse impacts of fungicides are directly related to their persistence and mobility in soil and the aqueous environment. Beigel et al. (1997) demonstrated that the diffusion and sorption coefficients of the fungicide triticonazole in soil was time dependent, which enabled them to predict the temporal variation of triticonazole concentration in soils. Jamet and Eudeline (1992) compared the mobility of 17 triazole fungicides in a silt loam soil and concluded that their mobility was inversely related to their octanol/water partition coefficient. Wu et al. (2003) investigated the sorption and desorption characteristics of propiconazole to different particle/aggregate-size fractions of agricultural runoff material and showed that sorption increased with decreasing particle size.

Environmental factors, such as local hydrology, soil characteristics, and climate, also contribute to the ultimate fate and potential of fungicides to reach non-target biota. Kim et al. (2003) suggested that the formation of bound residues of propiconazole was higher in silty clay loam soils than in sandy loam soils. However, they also found that the rates of degradation and mineralization of propiconazole were lower in silty clay loam soils than in sandy loam soil, and the half-life value in sandy loam soils was lower than that in the silty clay loam soils. Riise et al. (2004) studied the loss of bentazone and propiconazole from agricultural fields in southeast Norway and found that the loss of pesticide was more from the silty clay loam soils than silt loam soils. Kreuger (1998) studied the loss of some fungicides such as propiconazole and a few other pesticides to stream water in a small agricultural catchment in Sweden and concluded that the occurrence of these compounds in surface water is influenced by soil, weather conditions, and chemical properties.

Anthropogenic factors such as management practices including fungicide application rate and application timing also affect fungicide transport. Potter et al. (2005) indicated that repeated application of the fungicide tebuconazole increased its rate of dissipation in soil and reduced its persistence, thus influencing the risk of contamination through leaching. Beigel et al. (1999) demonstrated that the persistence of the fungicide triticonazole increased with application rate. Similar studies with pesticides have revealed that application rate is a dominant factor in determining pesticide concentration in surface waters (Sorensen et al. 2003).

Soybean fields normally do not receive widespread application of fungicides; therefore, it is expected that the new use of large quantities of fungicides to control soybean rust may have a negative impact on the environment. There have been no studies in Indiana to assess the impact of fungicides used to control soybean rust on the environment, including impacts on surface and groundwater resources and “off target” species. It can be assumed that soybean rust fungicides will behave in a similar way as other fungicides used in agricultural cropping activities. Therefore, residues of the fungicides used to control soybean rust may reach water sources through surface runoff or by subsurface leaching, as demonstrated by Battaglin and Sandstrom (2007), potentially compromising water sources for human and other uses and degrading downstream ecosystems. As a proactive step in environmental protection, it is thus essential to assess potential impacts and to identify areas that are particularly vulnerable to contamination by fungicides. This will provide a basis for developing approaches to minimize potential environmental impacts of fungicides, such as prioritizing implementation of best management practices or alternative management practices to the most vulnerable areas.

This project made use of the National Agricultural Pesticide Risk Analysis (NAPRA) model to predict average annual fungicide concentrations at edge of field and soil water concentrations at bottom of the root zone as a result of their application to control soybean rust and to evaluate the probability of exceeding threshold chronic exposure concentrations of concern for drinking water for humans and ambient water quality for aquatic organisms through the off-site movement of these products. Similar studies by Lim et al (2006) showed that the NAPRA model can be used to reasonably predict pesticide (atrazine) and nutrient losses to shallow groundwater. Adeuya et al. (2005) used Groundwater Loading Effects of Agricultural Management Systems (GLEAMS)-NAPRA to model the average annual nutrient losses of two watersheds in Indiana, and the simulated water quality data resulting from the model were very similar to observed data.

The objectives of this study were to examine the potential magnitude of fungicides reaching ground and surface water within Indiana as a result of their use in controlling ASR and to create NAPRA-predicted fungicide loss probability occurrence maps for Indiana from fungicide application scenarios. This provided a predictive modeling method using the probabilistic worst case estimates that is very similar to the second tier of aquatic exposure assessment using the Pesticide Root Zone Model followed by USEPA (USEPA 1998).

2 Materials and Methods

Parameters and assumptions in a risk assessment study tend to be associated with high degrees of uncertainty. As a result, worst case estimates of potential exposure are thought to be more protective of the environment or public health (Asante-Duah 1993). Therefore, the field-scale NAPRA model was run in a worst-case scenario mode, applying each of 14 soybean rust fungicides three times during the growing season at their maximum rates. The NAPRA model computed fungicide loss probability values to investigate water quality impacts of soybean rust fungicides. Fungicide loss probability maps were then created from the model outputs to provide a spatial image of the risk of surface water and shallow groundwater contamination.

2.1 Model Description

Lim and Engel (2003) developed the NAPRA WWW decision support system (http://cobweb.ecn.purdue.edu/~napra) to provide simple general access to a complex hydrologic/water quality model for a wide range of users. The NAPRA WWW system uses the GLEAMS (Leonard et al. 1987) model to simulate hydrology, pesticide, and nutrient losses in runoff, sediment, and to shallow groundwater (Lim and Engel 2003). The NAPRA WWW decision support system requires four major input types: (1) field characteristics, (2) management practices, (3) pesticides, and (4) nutrients. It can also simulate crop rotations and multiple pesticide and nutrient applications for each crop. The GLEAMS model within the NAPRA WWW system requires soil properties [obtained from either State Soil Geographic (STATSGO) database (NRCS 1994) or the National Soil Information System (NASIS) database (NRCS 1992)], user-defined crop management information, long-term daily temperature and precipitation data, user-defined tillage practice data, pesticide properties, and nutrient properties to create input files (Lim and Engel 2003). The GLEAMS model is run with these input files and generates the hydrology, pesticide and nutrient loss probability of exceedence (POE) and graphs of the simulated results (Lim and Engel 2003). The NAPRA WWW system can be used to predict spatial variations of pesticide and nutrient losses in surface and shallow groundwater.

2.2 Fungicide Loss Modeling

A statewide environmental risk assessment for 14 fungicides to be used in fighting soybean rust was conducted. Table 1 lists the physicochemical properties of the fungicides used. The NAPRA WWW system was run for cropped areas of Indiana [based on the Indiana National Agricultural Statistics Service (NASS) 2000 land use map] with the14 fungicides. The model runs were performed using STATSGO soil data that provided the soil properties, a single weather station and assuming continuous soybeans. STATSGO soil types are uniquely distinguished by a Map Unit ID (MUID) field or identifier. Each MUID in the STATSGO soil database is composed of 21 components, and each component has different soil properties. However, since no spatial information is associated with each component, the model outputs are the average fungicide concentration values for all components within one map unit (MUID) of STATSGO. The spatial distribution of the STATSGO

Table 1 Physico-chemical properties of soybean rust fungicides[1]

Fungicide Name	Foliar half life (days)	Solubility (mg/L)	Soil half life (days)	Koc
Chlorothalonil (ANSI)	10	0.6	30	1,380
Azoxystrobin (BSI, ISO)	3	6.7	65	1,590
Pyraclostrobin	3	2.3	83	11,000
Trifloxystrobin (ISO)	3	0.6	5	2,709
Propiconazole	30	110	110	1,000
Tebuconazole	30	25	610	1,000
Tetraconazole	30	156	117	1,226
Myclobutanil (ANSI)	10	142	66	500
Cyproconazole	10	140	144	900
Metconazole	10	15	73	1,000
Flusilazole	10	54	352	1,664
Famoxadone	10	0.1	20	3,665
Flutriafol	3	130	370	1,225
Prothioconazole	10	300	2.8	1,765

[1] From NRCS/UMass Extension/ARS. (2006). Pesticide properties database. USDA Natural Resources Conservation Service, Amherst MA; University of Massachusetts Extension, Amherst, MA; USDA Agricultural Research Service, Tifton, GA.

soil texture across Indiana is shown in Fig. 1. The other input parameters for the NAPRA model simulations used in this study are shown in Table 2, and they represent average conditions in Indiana. Foliar applications of the fungicides to the soybean canopy were the standard disease management practice, and one fungicide was modeled at a time with a worst case scenario of three applications during the

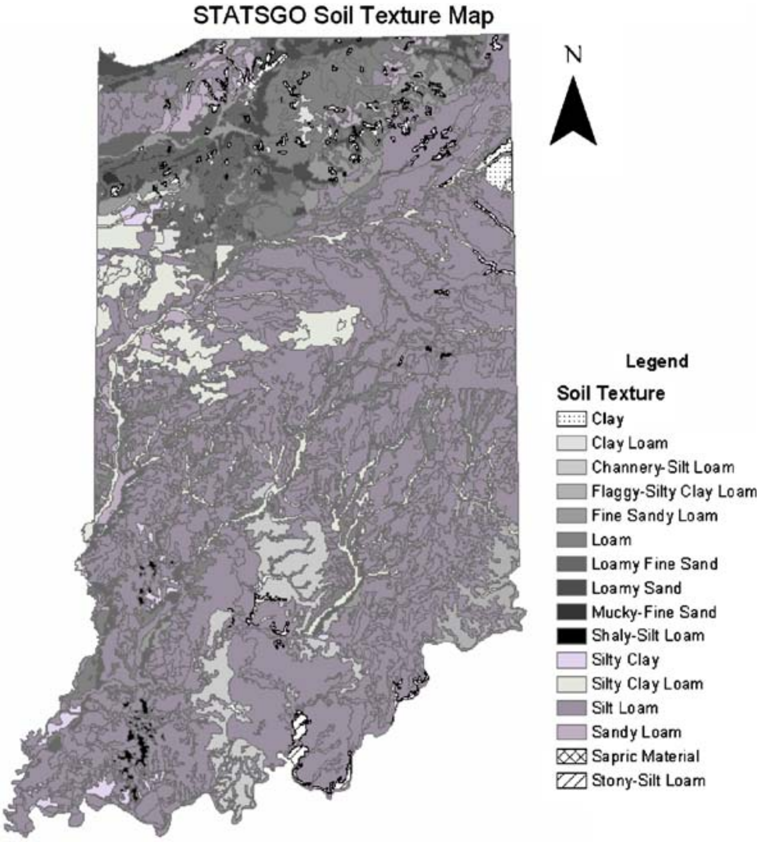

Fig. 1 STATSGO soil texture map of Indiana (NRCS 1994)

Table 2 Input parameters for the NAPRA model and their values

Input parameters	Input values
Landuse	Drilled soybean (Straight row + conservation tillage)
Hydrologic condition	Good
Crop Type	Continuous soybean
Tillage	No till
Effective rooting depth	0.75 m
Planting date	22nd April
Maturity date	15th September
Harvest date	1st October
Fungicide application method	Surface Application
Amount of soil surface covered by residue and vegetation	75%

growing season. The application dates for the 14 fungicides are listed in Table 3, and the application rates for the fungicides are summarized in Table 4. Different application timings were used for different fungicides because different fungicides are effective at different stages of soybean rust infection. The application rates are the maximum allowable rates permitted based on the labels for each of the 14 fungicides.

NAPRA-simulated fungicide loss (i.e., the fungicide concentrations in edge of field runoff and bottom of the root zone groundwater) probability results were compiled for each fungicide for each soil in each county for Indiana. This was done by plotting the fungicide concentrations in the edge of the field runoff/bottom of the root zone groundwater against the exceedence probabilities. The POE is obtained from the following equation (Cryer et al. 1998):

$$\text{Exceedence probability} = \frac{\text{rank}}{n+1}$$

where n = number of data points and rank = the integer value from 1 to n.

The annual runoff and shallow groundwater concentrations at 5% POE represent the worst case scenario of fungicide application, which may be expected to occur once in 20 years. In other words, the probability of such concentrations of fungicides occurring in edge of the field runoff or shallow groundwater is 5% in a given year. This is based on the USEPA Office of Pesticide Programs' guidance and methodology for predicting surface water exposure based upon numerical model results for single field chemical runoff (EPA 1998).

To compute 5%, 10%, 25%, and 50% fungicide loss probability values, simulated weather data for 60 years were used in the NAPRA/GLEAMS runs. The precipitation data were generated by climate generator (CLIGEN), a stochastic weather generator for climate inputs (Nicks et al. 1995), and the temperature data were generated by the generation of weather element for multiple applications (Johnson et al. 1996). ArcView GIS was used to process the results to create the fungicide loss probability maps

Table 3 Application timings for the soybean rust fungicides

Fungicide Name	Time of Application 1	Time of Application 2	Time of Application 3
Chlorothalonil (ANSI)	23-Jun	7-Jul	21-Jul
Azoxystrobin (BSI, ISO)	23-Jun	7-Jul	21-Jul
Pyraclostrobin	23-Jun	7-Jul	21-Jul
Trifloxystrobin (ISO)	23-Jun	7-Jul	21-Jul
Propiconazole	7-Jul	21-Jul	4-Aug
Tebuconazole	7-Jul	21-Jul	4-Aug
Tetraconazole	7-Jul	21-Jul	No application
Myclobutanil (ANSI)	7-Jul	21-Jul	4-Aug
Cyproconazole	28-May	11-Jun	25-Jun
Metconazole	28-May	11-Jun	25-Jun
Flusilazole	28-May	11-Jun	25-Jun
Famoxadone	28-May	11-Jun	25-Jun
Flutriafol	28-May	11-Jun	25-Jun
Prothioconazole	28-May	11-Jun	25-Jun

Table 4 Application rates for the soybean rust fungicides

Fungicide name	Application rate (kg/ha)
Chlorothalonil (ANSI)	0.51
Azoxystrobin (BSI, ISO)	0.56
Pyraclostrobin	0.29
Trifloxystrobin (ISO)	0.18
Propiconazole	0.53
Tebuconazole	0.25
Tetraconazole	0.20
Myclobutanil (ANSI)	0.34
Cyproconazole	0.03
Metconazole	0.06
Flusilazole	0.12
Famoxadone	0.06
Flutriafol	0.06
Prothioconazole	0.12

using the STATSGO soil layer. Annual mean concentrations for the 14 fungicides in edge of field runoff and bottom of the root zone soil water were also predicted with the model.

The chronic exposure of humans through drinking water impacted by fungicides and/or of aquatic species to a fungicide above the threshold concentration of concern may potentially impact directly or indirectly the health of the exposed individual or organism. The human drinking water chronic exposure toxicity and the fish chronic exposure toxicity threshold concentration values used within the NAPRA model are summarized in Table 5. The NAPRA results were also used to calculate the percentage of cropped areas in Indiana that were estimated to have fungicide concentrations in edge of field runoff or bottom of root zone soil water above the chronic exposure threshold concentration values for fish and humans at 5%, 10%, 25%, and 50% probabilities of exceedence.

3 Results and Discussion

The NAPRA WWW generated maps show spatial variations of fungicide losses in edge of field runoff and to bottom of root zone soil water or shallow groundwater at 5%, 10%, 25%, and 50% POE. The examples shown in Figs. 2 and 3 illustrate average annual tebuconazole concentrations in runoff and shallow groundwater, respectively, at a 5% POE. The estimated variations of tebuconazole concentrations in runoff and shallow groundwater across Indiana result from variations in soil across the state combined with interactions of the fungicide chemical and physical properties with different soil types. From Fig. 2, it is evident that the concentration of tebuconazole lost in runoff is greater in the central part of Indiana than in the northern and southern regions. Comparing this pattern with that of soil texture (Fig. 1), it is evident that the concentration loss with surface runoff is highest in clayey and silty

Table 5 Fungicide threshold concentrations of concern for chronic exposure of humans consuming drinking water and fish aquatic habitat

Fungicide name	Human HA/CHCL (ppb)[a]	Fish MATC (ppb)[b]
Chlorothalonil (ANSI)	45.7	4.42
Azoxystrobin (BSI, ISO)	1260	168.44
Pyraclostrobin	210	3.88
Trifloxystrobin (ISO)	350	5.75
Propiconazole	9.1	134.16
Tebuconazole	21	17.32
Tetraconazole	1.65	2,650
Myclobutanil (ANSI)	175	329.79
Cyproconazole	1.16	452
Metconazole	33.6	299.7
Flusilazole	0.49	153.9
Famoxadone	9.8	2.4
Flutriafol	70	11,584
Prothioconazole	350	244.7

[a] Plotkin et al. (2006a)

[b] Plotkin et al. (2006b)

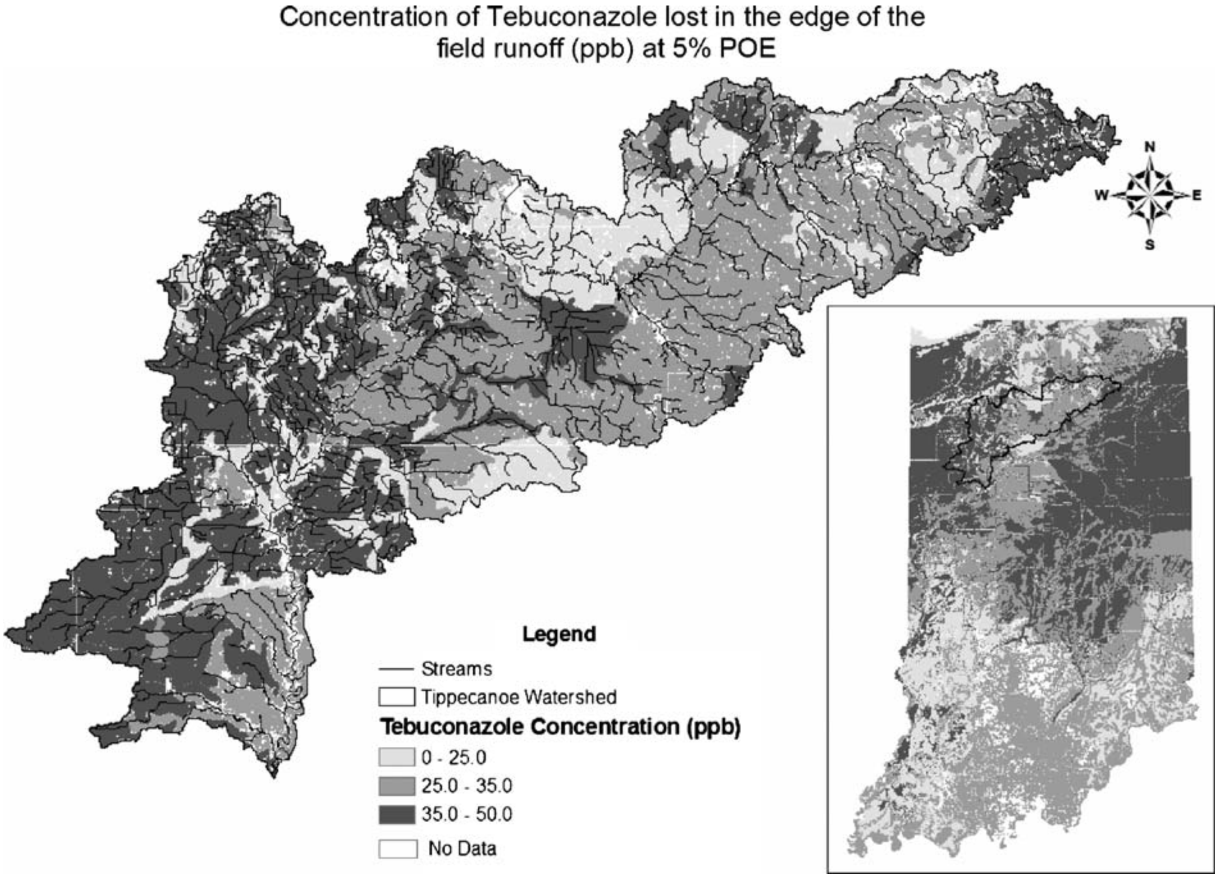

Fig. 2 Simulated spatial distribution of tebuconazole loss in edge of field runoff across Indiana at a 5% probability of exceedence

clay loam soils. The concentration of tebuconazole lost is also high in the silt loams of central Indiana, whereas concentration losses with surface runoff are low in the loamy and sandy loam soils of northern Indiana. Figure 3 shows that the average annual concentration of tebuconazole lost to shallow groundwater is more in the southern part of Indiana and lower in the center of the state. Comparing Figs. 2 and 3, it is evident that the concentration loss to bottom of the root zone soil water or shallow groundwater is higher in areas where the concentration loss in surface water runoff is lower, and vice versa. This pattern can be attributed to soil properties, as soils that aid in runoff, e.g., clayey soils, retard the leaching process, while soils that increase leaching will reduce losses through runoff.

The average annual maximum and minimum concentrations of the fungicides in edge of field runoff and bottom of the root zone soil water or shallow groundwater at 5% POE are compiled in Table 6. From Table 6, it is clear that some fungicides have higher concentrations lost in runoff than to soil percolation and vice versa. This is due to the physicochemical properties of the fungicides and their interactions with different soil types as would be for other pesticides/chemicals modeled with NAPRA. The higher concentrations in runoff may also be due to a large rainfall event near the time of fungicide application. For example, the fungicides chlorothalonil and azoxystrobin may have higher concentration losses in runoff because of their persistence in soil (30- and 60-day half-life, respectively) in combination with the loose sorption of the pesticides to soil particles (1,380 and 1,590 Koc, respectively), whereas the fungicides tebuconazole, flusilazole, and flutriafol have higher concentration losses to bottom of the root zone soil water because of their longer soil half lives (610, 352, and 370 days, respectively) allowing them to be persistent enough to pass into shallow groundwater, if present. Pyraclostrobin, trifloxystrobin, met-

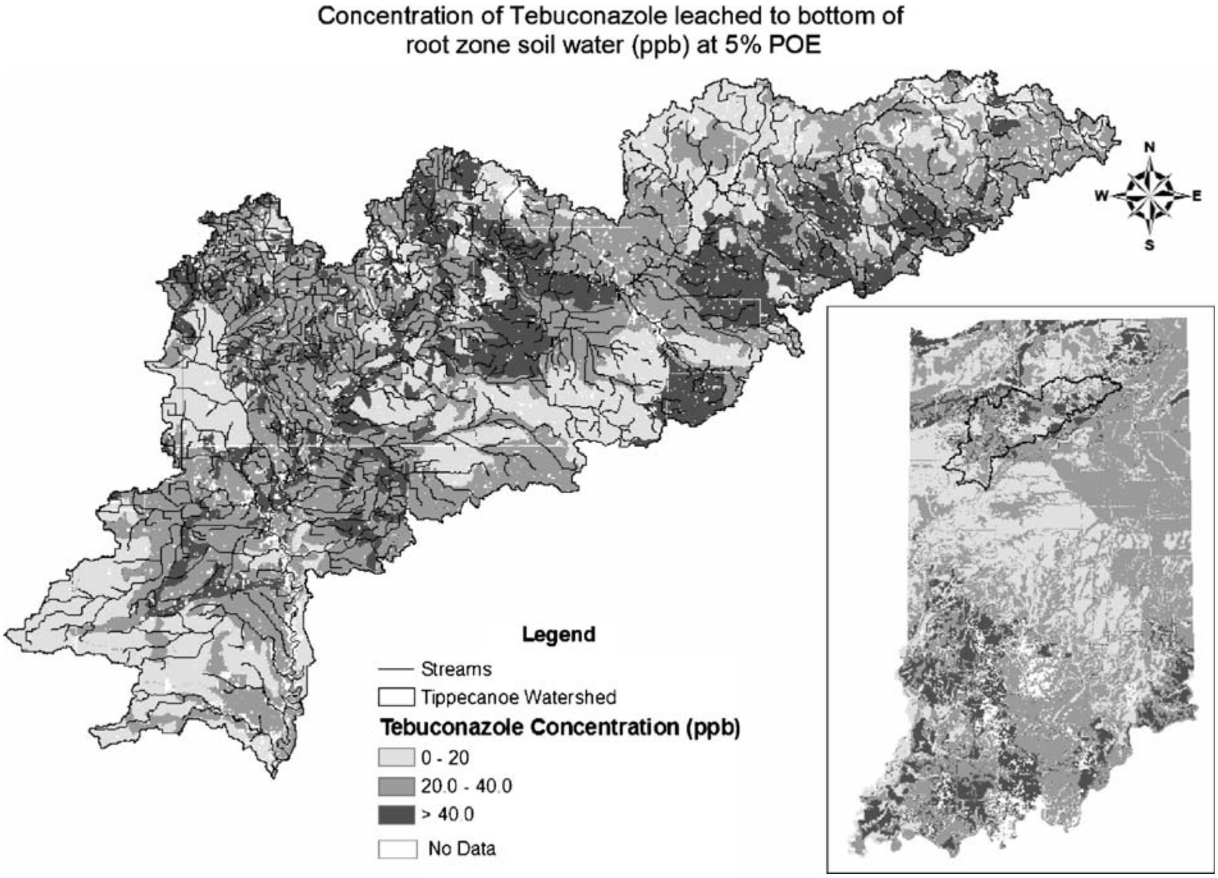

Fig. 3 Simulated spatial distribution of tebuconazole loss leached to bottom of root zone soil water across Indiana at a 5% probability of exceedence

conazole, famoxadone, and prothioconazole have negligible concentrations lost through runoff and soil percolation. Negligible concentrations of trifloxystrobin and famoxadone may have occurred because of the combination of their low solubility values (0.61 and 0.052 mg/l, respectively) and short half-lives (5 and 20 days). In contrast, prothioconazole demonstrated similar results but is highly water soluble (300 mg/l) with a short half-life of 2.8 days. Negligible runoff and percolation concentration results also occurred with pyraclostrobin, which is characterized as tightly sorbed to soil particles (11,000 Koc) combined with low solubility in water (2.4 mg/l). However, the lower toxicity value of pyraclostrobin for fishes, coupled with its persistence in the soil resulting from higher Koc and lower solubility makes this negligible concentration potentially toxic to fishes. Finally, metconazole has similar properties to cholorthalonil and azoxystrobin but is applied in small quantities. The low dose amount appears to have contributed to the negligible concentrations in runoff and soil percolation results. The other four fungicides (propiconazole, myclobutanil, tetraconazole, and cyproconazole) have nearly equal concentration losses in runoff and soil percolation, which may be due to their high water solubility, persistent soil half-life, and low to intermediate soil sorption values.

The NAPRA-generated results were also used to determine the percentage of the cropped area in Indiana that has fungicide edge of field runoff and bottom of the root zone soil water concentrations above chronic exposure threshold values of concern for drinking water for humans and aquatic habitat for fish at 5%, 10%, 25%, and 50% probabilities of exceedence (Tables 7, 8, 9, and 10). From Table 7, it is evident that the fungicides propiconazole, tetraconazole, and flusilazole potentially impact surface water quality, as there is 50% chance for the edge of field concentrations of these fungicides to exceed the

Table 6 Average annual maximum and minimum concentrations of the 14 ASR fungicides in edge of field runoff and bottom of root zone soil water in continuous soybean planting scenario

Fungicide	5% POE			
	Maximum concentration (ppb) in runoff	Minimum concentration (ppb) in Runoff	Maximum concentration (ppb) in leaching	Minimum concentration (ppb) in leaching
Chlorothalonil (ANSI)	76.0	9.8	3.0	0.0
Azoxystrobin (BSI, ISO)	27.4	5.7	0.0	0.0
Pyraclostrobin	13.7	1.5	0.3	0.0
Trifloxystrobin (ISO)	2.1	0.1	0.0	0.0
Propiconazole	42.3	13.2	46.7	0.0
Tebuconazole	45.7	14.2	224.5	2.5
Tetraconazole	10.3	4.0	10.8	0.0
Myclobutanil (ANSI)	16.0	3.3	18.4	0.0
Cyproconazole	2.3	0.7	5.5	0.2
Metconazole	0.0	0.0	0.0	0.0
Flusilazole	16.1	5.7	53.9	1.2
Famoxadone	1.5	0.4	0.0	0.0
Flutriafol	9.0	2.8	38.3	1.4
Prothioconazole	1.5	0.2	0.0	0.0

chronic exposure concentration of concern for drinking water in a given year for nearly 98% of the cropped area of Indiana It is interesting to note that the percentage of the cropped area that is predicted to have concentrations of propiconazole, tetraconazole, and flusilazole above the human health advisory concentration value is the same at 5%, 10%, 25%, and 50% POE. This is because, for each of these

Table 7 Percentage of cropped area where average annual fungicide concentration in edge of field runoff is above the chronic exposure health advisory value for human drinking water consumption at 5%, 10%, 25%, and 50% probability of exceedence

Fungicide name	Percent of the cropped area where fungicide concentration in edge of field runoff is above the chronic exposure health advisory value for humans at			
	5% POE	10% POE	25% POE	50% POE
Chlorothalonil (ANSI)	53.6	2.6	1.3	0.0
Azoxystrobin (BSI, ISO)	0.0	0.0	0.0	0.0
Pyraclostrobin	0.0	0.0	0.0	0.0
Trifloxystrobin (ISO)	0.0	0.0	0.0	0.0
Propiconazole	98.2	98.2	98.2	98.2
Tebuconazole	95.0	93.3	90.8	69.7
Tetraconazole	98.2	98.2	98.2	98.2
Myclobutanil (ANSI)	0.0	0.0	0.0	0.0
Cyproconazole	87.3	80.2	54.5	47.1
Metconazole	0.0	0.0	0.0	0.0
Flusilazole	98.2	98.2	98.2	98.2
Famoxadone	0.0	0.0	0.0	0.0
Flutriafol	0.0	0.0	0.0	0.0
Prothioconazole	0.0	0.0	0.0	0.0

Table 8 Percentage of the cropped area where average annual fungicide concentration in bottom of root zone soil water is above the chronic exposure health advisory value for humans at 5%, 10%, 25%, and 50% POE

Fungicide name	Percent of the cropped area where fungicide concentration in bottom of root zone soil water is above the chronic exposure health advisory value for humans at			
	5% POE	10% POE	25% POE	50% POE
Chlorothalonil (ANSI)	0.0	0.0	0.0	0.0
Azoxystrobin (BSI, ISO)	0.0	0.0	0.0	0.0
Pyraclostrobin	0.0	0.0	0.0	0.0
Trifloxystrobin (ISO)	0.0	0.0	0.0	0.0
Propiconazole	0.0	0.0	0.0	0.0
Tebuconazole	54.1	9.6	8.2	8.2
Tetraconazole	0.0	0.0	0.0	0.0
Myclobutanil (ANSI)	0.0	0.0	0.0	0.0
Cyproconazole	51.8	0.0	0.0	0.0
Metconazole	0.0	0.0	0.0	0.0
Flusilazole	98.2	89.3	17.5	15.9
Famoxadone	0.0	0.0	0.0	0.0
Flutriafol	0.0	0.0	0.0	0.0
Prothioconazole	0.0	0.0	0.0	0.0

fungicides, the lowest concentration in runoff at any location at any of the above probabilities exceeded the calculated human health advisory values. As a result, there is a 50% chance for most of the cropped areas to have edge of field runoff concentrations above chronic exposure threshold values in any year. Tebuconazole and cyproconazole follow these three fungicides closely with concentrations in edge of field runoff exceeding their calculated health advisory threshold values for human beings in 95% and 87% of the cropped area of the state in a worst case scenario at 5% POE. The percentage area for these two fungicides decreases with increasing probability of occurrence. Among other fungicides, there is a 5%

Table 9 Percentage of the cropped area where average annual fungicide concentration in edge of field runoff is above the chronic toxicity value for fish at 5%, 10%, 25% and 50% POE

Fungicide name	Percent of the cropped area where fungicide concentration in edge of field runoff is above the chronic toxicity value for fish at			
	5% POE	10% POE	25% POE	50% POE
Chlorothalonil (ANSI)	98.2	98.2	98.2	97.0
Azoxystrobin (BSI, ISO)	0.0	0.0	0.0	0.0
Pyraclostrobin	97.0	97.0	96.4	96.4
Trifloxystrobin (ISO)	0.0	0.0	0.0	0.0
Propiconazole	0.0	0.0	0.0	0.0
Tebuconazole	98.2	98.1	98.1	85.7
Tetraconazole	0.0	0.0	0.0	0.0
Myclobutanil (ANSI)	0.0	0.0	0.0	0.0
Cyproconazole	0.0	0.0	0.0	0.0
Metconazole	0.0	0.0	0.0	0.0
Flusilazole	0.0	0.0	0.0	0.0
Famoxadone	0.0	0.0	0.0	0.0
Flutriafol	0.0	0.0	0.0	0.0
Prothioconazole	0.0	0.0	0.0	0.0

Table 10 Percentage of the cropped area where average annual fungicide concentration in bottom of root zone soil water is above the chronic toxicity value for fish at 5%, 10%, 25%, and 50% POE

Fungicide name	Percent of the cropped area where fungicide concentration in bottom of root soil water is above the chronic toxicity value for fish at			
	5% POE	10% POE	25% POE	50% POE
Chlorothalonil (ANSI)	0.0	0.0	0.0	0.0
Azoxystrobin (BSI, ISO)	0.0	0.0	0.0	0.0
Pyraclostrobin	0.0	0.0	0.0	0.0
Trifloxystrobin (ISO)	0.0	0.0	0.0	0.0
Propiconazole	0.0	0.0	0.0	0.0
Tebuconazole	74.7	17.5	9.6	9.6
Tetraconazole	0.0	0.0	0.0	0.0
Myclobutanil (ANSI)	0.0	0.0	0.0	0.0
Cyproconazole	0.0	0.0	0.0	0.0
Metconazole	0.0	0.0	0.0	0.0
Flusilazole	0.0	0.0	0.0	0.0
Famoxadone	0.0	0.0	0.0	0.0
Flutriafol	0.0	0.0	0.0	0.0
Prothioconazole	0.0	0.0	0.0	0.0

chance that chlorothalonil concentrations in runoff will exceed calculated health advisory values for humans in 53% of the cropped area of the state in a given year. Other fungicides, such as azoxystrobin, pyraclostrobin, trifloxystrobin, myclobutanil, metconazole, famoxadone, flutriafol, and prothioconazole, are predicted not to pose a threat to source water used for drinking, as no area in Indiana is predicted to have concentrations exceeding the human health threshold concentrations of concern for these fungicides.

The percentage of the cropped area in the state predicted to have average annual fungicide concentrations in the bottom of the root zone soil water or shallow groundwater, which are above calculated health advisory concentration values for humans for all 14 fungicides for the range of probabilities considered, is compiled in Table 8. For humans in Indiana, there is a 5% chance in a given year that the fungicide flusilazole will impact water from nearly 98% of the cropped area of the state, with concentrations in the bottom of the root zone soil water or shallow groundwater above the values at which the fungicide may pose a chronic exposure concern for humans. The percentage area with concentrations in shallow groundwater above the health advisory value decreases with increasing probability of occurrence, with approximately 15% of the cropped area exceeding concentrations above chronic exposure threshold levels of concern at 50% POE. For tebuconazole and cyproconazole, there is a 5% chance that their concentrations in shallow groundwater will exceed the calculated human health advisory value in nearly 50% of the cropped area in the state in a given year. Others such as azoxystrobin, chlorothalonil, pyraclostrobin, trifloxystrobin, myclobutanil, metconazole, famoxadone, prothioconazole, propiconazole, and tetraconazole are not estimated to pose a chronic exposure concern to humans, as no area in Indiana is predicted to have fungicide concentrations in shallow groundwater exceeding the calculated human health advisory values.

The percentage of the cropped area in the state predicted to have average annual fungicide concentrations in edge of field runoff exceeding the chronic exposure toxicity levels for fish for the 14 fungicides at a range of probabilities is compiled in Table 9. From Table 9, it can be inferred that the fungicides chlorothalonil, pyraclostrobin, and tebuconazole are potentially harmful for fish in Indiana because more than 96% of the cropped area of the state is predicted to have concentrations of these fungicides in edge of field runoff above their chronic toxicity threshold of concern, and there is a 50% chance for these percentages of affected area to occur in a given year. For all of these fungicides, the lowest concentration in runoff at any location is greater than the chronic toxicity values at all probability ranges. Others such as azoxystrobin, propiconazole, tetraconazole, myclobutanil, cyproconazole, metconazole, flusilazole, flutriafol, prothioconazole, trifloxystrobin, propiconazole, and famoxadone do not seem to pose as a great concern to fish, as no cropped area in Indiana is predicted to have fungicide concentrations in runoff exceeding the chronic toxicity values.

The percentage of the cropped area in the state predicted to have average annual fungicide concentrations in the bottom of the root zone soil water or shallow groundwater exceeding the chronic toxicity levels for fish for the 14 fungicides at a range of probabilities is compiled in Table 10. Within much of Indiana, water that is leached below the root zone is intercepted by subsurface drains and is discharged in surface drainage channels. Thus, fungicides leaching toward shallow groundwater are likely to quickly reach surface waters. The concentration of tebuconazole in the root zone soil water is predicted to exceed the chronic exposure toxicity value of the fungicide for fish in nearly 75% of the cropped area of the state once every 20 years. The concentration of the other fungicides in the root zone soil water is not predicted to exceed fish threshold toxicity values of concern in Indiana.

As shown in this study, the NAPRA model predicted that the average annual concentration of some of the fungicides such as azoxystrobin, trifloxystrobin, famoxadone, prothioconazole, myclobutanil, metconazole, and flutriafol are below the concentrations at which there may be chronic exposure concerns to humans and fish, indicating that they may impact the environment less than other compounds considered. If they are effective in combating ASR, their use should be prioritized over the ones that pose greater concern for sensitive non-target areas, such as drinking water source water watersheds and aquatic organism habitat. Note that this is based on worst case conditions for fungicide applications: All cropped areas were in continuous soybeans, all soybeans received the same fungicide, and no dilution of edge of field runoff or bottom of root zone soil water or shallow groundwater was considered. Fungicides that are potentially detrimental to the environment in certain areas of Indiana may require the use of best management practices such as setbacks and filter strips to reduce their environmental impacts if they are used to fight ASR.

The results presented are based on worst case conditions for fungicide applications, where all cropped areas were modeled as continuous soybeans. Balderacchi et al. (2008) has shown that crop rotation mitigates the risk from pesticide contamination by 48–74% depending upon the number of crops involved in the rotation. Therefore, for a corn–soybean rotation (currently the dominant crop rotation within Indiana), the fungicide concentrations in runoff and in the bottom of the root zone soil water from small agriculturally dominated watersheds would be approximately 50% of the modeled values if other model inputs remained the same. Modeled fungicide losses for a single application are about one-third of the amount modeled with three applications. This is because the rainfall patterns, temperatures, crop conditions, and other conditions that would result in fungicide degradation and movement of fungicides to water are similar for the period of expected application and the period following application. Therefore, for a small agriculturally dominated watershed with corn soybean rotation with one application of a soybean rust fungicide to soybeans, the annual fungicide concentrations in runoff and in the bottom of the root zone soil water would be about one-sixth of the values modeled.

The modeled fungicide losses from a small agricultural watershed with a corn–soybean rotation indicate that the maximum chlorothalonil concentration in runoff is higher than its chronic toxicity value for fish, whereas the maximum concentration of tebuconazole in the bottom of the root zone soil water exceeds its chronic toxicity values for both fish and humans. For tetraconazole in this scenario, the maximum concentrations in both runoff and shallow groundwater exceed its chronic toxicity value for humans. For flusilazole for this situation, the minimum concentration in runoff and the maximum concentration in shallow groundwater exceeded its chronic toxicity value for humans. The percentage of cropped areas in Indiana with concentrations of soybean rust fungicides exceeding the threshold concentrations of concern for both humans and fishes in a corn soybean rotation scenario will be about 50% of the cropped areas with concentrations of soybean rust fungicides exceeding the threshold concentrations of concern for both humans and fishes in continuous soybean planting scenario.

This modeling is based on expected worst case conditions for fungicide applications, where all cropped areas were in continuous soybeans, all soybeans received the same fungicide at label maximum rates and maximum number of applications, and no dilution of edge of field runoff or bottom of root zone soil water was considered. Since the loss concentrations of soybean rust fungicides modeled with NAPRA represent the concentrations in the edge of field runoff and in the bottom of the root zone soil water, concentrations will likely be diluted by the time runoff reaches surface water bodies. There is also a possibility of further degradation of the compounds during their transport and once reaching water.

It is important to calibrate a model and validate its results against field data to increase confidence in predicted results. In this study, fungicide concentrations in edge of field runoff and bottom of root zone soil water were needed to achieve this purpose. Since soybean rust fungicides have not been used extensively across Indiana, the results of this study could not be compared against field data to assess model performance. However, the model has been validated for other pesticides within Indiana with acceptable performance (Lim et al. 2006).

Moreover, uncertainties associated with the input parameters and assumptions used in this modeling may lead to overestimation of the modeled outputs, but these uncertainties do not invalidate the use of results in decision making.

4 Conclusions

An environmental risk evaluation for 14 fungicides that may be approved for use in fighting ASR in Indiana was conducted using the NAPRA hydrologic/water quality modeling system. NAPRA-simulated fungicide loss probability results representing the worst case scenarios (i.e., maximum application rates) were compiled for each fungicide for each soil in each county. The average annual fungicide loss concentrations at 5%, 10%, 25%, and 50% POE were compared to the chronic exposure threshold concentrations of concern in water at which the fungicides may impact the health of exposed fish and humans. These values were used to calculate the percentages of the cropped areas in Indiana identified as having fungicide field runoff and/or bottom of root zone soil water concentrations above threshold values of concern for fish and humans. The NAPRA results were also used to infer which fungicides among the 14 may potentially impact sources of water used by fish and humans.

The results indicate that chlorothalonil, propiconazole, cyproconazole, flusilazole, tebuconazole, and tetraconazole concentrations were present in runoff or in shallow groundwater, and thus, their off-site movement may affect source water drinking water, whereas chlorothalonil, pyraclostrobin, and tebuconazole are potentially above chronic exposure thresholds of concern for fish. Others such as azoxystrobin, trifloxystrobin, famoxadone, prothioconazole, myclobutanil, metconazole, and flutriafol do not runoff or leach to groundwater at concentrations above chronic exposure thresholds of concern for either humans or fish in Indiana. This analysis, along with the spatial representation of surface and subsurface contamination, will be helpful in identifying the potential magnitude of fungicide losses to the environment and the spatial patterns of these losses. When combined with data from toxicity tests, published information, and computational toxicology results, these results will help in evaluating and ranking fungicides according to their potential impact and will be helpful to policy makers as they work on balancing crop protection with environmental protection and in developing best management practice guidelines to mitigate potential risks from these fungicides.

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