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Comparison of Regulatory Method Estimated Drinking Water Exposure Concentrations with Monitoring Results from Surface **Drinking Water Supplies**

SCOTT JACKSON,*,[†] PAUL HENDLEY,[‡] RUSSELL JONES,[§] NICK POLETIKA,[#] AND Mark Russell^{\perp}

BASF Corporation, 26 Davis Drive, Research Triangle Park, North Carolina 27709; Syngenta Crop Protection, 410 South Swing Road, Greensboro, North Carolina 27419; Bayer CropScience, 17745 South Metcalf Avenue, Stilwell, Kansas 66085; Dow AgroSciences, Dow AgroSciences LLC, 9330 Zionsville Road, Indianapolis, Indiana 46268; and DuPont Crop Protection, DuPont Stine Haskell Research Center, 1090 Elkton Road, Newark, Delaware 19714-0030

Crop-protection compounds are useful tools that enhance the quality of the food we enjoy. However, crop-protection products can enter aquatic systems either by direct or by indirect application. To better understand the possible frequency and magnitude of exposure to water resources, the regulatory community has developed a set of relatively straightforward models for estimating exposure to these water systems. The focus of this research was to compare how well the estimates of exposure to drinking water based on model calculations relate to actual monitoring data. Physical/chemical property data were entered in the EPA's exposure model FIRST and into PRZM/EXAMS. The predictions from FIRST and PRZM/EXAMS were then compared to actual monitoring data from a USGS/EPA cooperative program, which monitored for pesticides in vulnerable surface drinking water supplies during 1999 and 2000. Results from this examination indicate the exposure from the models can overpredict concentrations found in water by several orders of magnitude. An overprediction factor is presented that corrects model predictions to more closely approximate concentrations found in reservoirs (p = 0.05).

KEYWORDS: Crop protection; drinking water exposure; monitoring; PRZM/EXAMS; Index Reservoir

INTRODUCTION

Many synthetic compounds have been developed over the years in an effort to improve people's lives. However, it is society's obligation to ensure that these compounds do not deleteriously affect the environment. Therefore, to evaluate the possible impact of compounds on the environment, methods needed to be developed to allow assessments to be conducted. One of the goals of the scientific and regulatory community has been to develop a standard set of exposure models to predict expected environmental exposure based on proper use of cropprotection products. Exposure models have been developed to help predict and extrapolate compound behavior in groundwater, surface water, and soil systems. Exposure models have become popular in the regulatory community and with entities being regulated because relatively few measurements can be used to extrapolate to a wide variety of environmental conditions. However, the effectiveness of a model to predict behavior can

only be as good as its representation of the environment it is designed to predict. When the Office of Pesticide Programs (OPP) at the U.S. Environmental Protection Agency (EPA) assesses the risk of a pesticide, it considers the toxicity of the pesticide as well as exposure to the pesticide. In assessing risk to human health and the environment, the OPP routinely estimates concentrations of pesticides in natural water bodies, such as lakes or ponds, when it develops aquatic exposure assessments. The pesticide program also estimates concentrations of pesticides in water bodies when it considers the effect of pesticides on the value of water resources in its water quality assessments. OPP typically relies on mathematical models to generate the exposure estimates for drinking water and aquatic exposure assessments and water quality assessments. Models calculate estimated environmental concentrations using laboratory data that describe how quickly the pesticide degrades and how it may move in the environment.

FIRST is a program used to calculate both acute and chronic generic expected environmental concentration values in drinking water (DWECs). FIRST estimates runoff from a watershed into a reservoir that would be used for drinking water. FIRST is the EPA's first tier or coarse screen, which estimates expected water concentrations from a few basic chemical parameters and label information. FIRST is designed to mimic a PRZM/EXAMS

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^{*} Corresponding author [telephone (919) 547-2349; e-mail jacksosh; basf-corp.com].

BASF Corp.

[‡] Syngenta Crop Protection. § Bayer CropScience.

[#] Dow AgroSciences.

[⊥] DuPont Crop Protection.

Table 1. Summary of the Compounds Modeled, the Scenario Chosen, Application Method with Application Rate and Number of Applications

		rate (lb/ appl no.		
compound (class)	EPA model scenario used	acre)	(interval)	appl method
aldicarb (I)	CA fruit (noncitrus)	10	1	ground
aldicarb (l)	MS cotton	5	1	ground
aldicarb (l)	FL citrus	5	1	ground
bromoxynil (H)	MS cotton	0.5	3 (10 days)	aerial
carbaryl (I)	FL citrus	5	4 (14 davs)	aerial
acifluorfen (H)	MS soybean	0.5	1 1	ground
bentazon (H)	MS soybean	1	2 (14 days)	ground
dicamba (H)	CA alfalfa	0.5	2 (14 davs)	around
imazaguin (H)	MS sovbean	0.13	1	around
imazethapyr (H)	OH corn	0.1	1	around
MCPA (H)	OR wheat	2	2 (90 days)	around
bentazon (H)	MS sovbean	1	2 (90 days)	around
dicamba (H)	CA alfalfa	0.5	2 (14 days)	around
2.4-D (H)	FL sugarcane	2.00	1	around
2.4-DB (H)	TX alfalfa	2.00	1	around
clopyralid (H)	OH corn	0.50	1	around
flumetsulam (H)	OH corn	0.07	1	around
orvzalin (H)	NC apples	6.00	1	around
picloram(H)	OR wheat	0.25	1	around
tebuthiuron (H)	FL sugarcane	20.00	1	around
triclopyr (H)	OR Christmas	9.00	1	aerial
benomyl (F)	GA onions	1.0	3 (14 days)	aerial
bensulfuron-methyl (H)		0.1	1	aerial
bromacil (H)	FL citrus	1.9	2 (60 days)	ground
chlorimuron-ethyl (H)	MS soybean	0.019	2 (14 days)	aerial
diuron (H)		12.0	1	ground
linuron (H)	PA vegetable	1.0	2 (14 days)	ground
methomyl (I)	PA vegetable	0.9	10 (2 days)	ground
metsulfuron-methyl (H)	C C	0.188	1	ground
nicosulfuron (H)	OH corn	0.063	1	aerial
oxamyl (I)	NC apples	1.0	8 (5 days)	airblast
sulfometuron-methyl (H)	OR Christmas	0.375	1	ground
terbacil (H)	LA sugarcane	1.2	2 (60 days)	aerial
tribenuron-methyl (H)	TX wheat	0.016	2 (14 days)	aerial
2-hydroxyatrazine (D)	LA sugarcane		4 (21 days)	aerial
atrazine(H)	LA sugarcane	2.5	4 (21 days)	aerial
deethylatrazine (D)	LA sugarcane	0.333	4 (21 days)	aerial
deisopropylatrazine (D)	LA sugarcane	0.07	4 (21 days)	aerial
fluometuron (H)	MS cotton	1.7	3 (14 days)	ground
metalaxyl (F)	GA peaches	4	3 (90 days)	aerial
norflurazon (H)	FL citrus	3.8	2 (180 days)	ground
propiconazole (F)	PA turf	1.78	4 (7 days)	ground

simulation. For tier 2 surface water screening assessments, the OPP uses the linked PRZM and EXAMS models (PRZM/ EXAMS), which better model the specific characteristics of the chemical. Standard scenarios used with PRZM/EXAMS include more site-specific information regarding the application method and the impact of daily weather on the treated field over a period of 36 years compared to tier I models. As a higher level screening tool, PRZM/EXAMS uses maximum application rates and frequencies for a vulnerable surface drinking water reservoir.

The focus of this work was to evaluate the ability of two exposure models, FIRST and PRZM/EXAMS, to predict pesticide concentrations found in actual reservoirs used for drinking water supply following standard EPA regulatory modeling practice.

MATERIALS AND METHODS

One of the primary requirements for the compounds selected in this comparison is that they were monitored for in the study we used for comparison. Additional factors that helped determine which molecules were selected included availability of compound physical/chemical property information and that the EPA had standard crop scenarios available for the uses we needed to model predictions for at the tier II level. Model input parameterization of physical-chemical properties was done following EPA standard operation procedures (1-7), which can be found on the EPA's OPP water model Web site. The physical-chemical properties themselves were provided by the registrants having compounds modeled in this research. The tier I modeling using the

FIRST model was conducted by the registrants, whereas the tier II PRZM/EXAMS modeling was done by a third-party company. The FIRST model provides an acute and a chronic value, whereas the PRZM/EXAMS models were run for multiple years using the specific crop and specific weather scenarios used by the EPA. For tier II modeling, results from the multiple-year runs were sorted from high to low, and the 90th percentile result was the value used for comparison for both acute and chronic endpoints (following standard agency procedures). Results from the modeling runs were then compared to the maximum monitoring values found in the USGS reservoir study, Pesticides in Selected Water Supply Reservoirs and Finished Drinking Water 1999-2000 Summary of Results from a Pilot Monitoring Program (8). A brief historical background on the reservoir study design is provided. The International Life Sciences Institute (ILSI) held a workshop to address the question of water sampling frequency and what various sampling schemes represent concerning accuracy and precision (9). The ILSI group comprised academics, regulators, and industry scientists. Conclusions from the workshop were that collection of 33 samples per season produced 50th, 95th, and 99th percentile estimates, which were well within the 90% confidence interval, and between 1 and 25% of the true estimates. Furthermore, sampling weekly was determined to be adequate for estimating the 50th, 90th, and 95th percentile concentrations for acute assessments. Site number could also be substituted for the required number of sampling years to accurately describe concentrations. The conclusions from the workshop were independently verified by a group of statisticians at the Research Triangle Institute, Research Triangle Park, NC (9). The findings of the ILSI group were used to design the USGS reservoir study we used in this comparison.



Figure 1. Comparison of tier I acute model predictions versus monitoring results.

The stated objectives of the USGS reservoir study were to provide estimates of annual-mean pesticide concentrations and high concentrations that occur for short periods of time, such as the annual 95th and 99th percentile concentrations. The study collected and analyzed water samples for two years from 12 reservoirs distributed throughout the country. Monitoring frequencies varied from site to site and ranged from a bimonthly baseline with 11 samples in a year to enhanced sampling (every 3 days) at some sites during the use season for a maximum of 37 samples per year. Sample collection was focused on the primary application times for the products, May–September. Periodic samples were collected in the winter months, normally targeting January. The authors of the reservoir study state that the systems selected for the monitoring program were reservoirs considered to be highly vulnerable to pesticide contamination.

Table 1 is a summary of the compounds used for comparison in this study along with the model scenario used, application rate, and method of application. For tier I comparisons of modeling to monitoring data, the monitoring value used was the maximum observed for both acute and chronic model result comparisons. For tier II comparisons of modeling to monitoring data, the acute modeled result was compared to the maximum observed monitoring value obtained. For tier II chronic comparison of modeling to monitoring data, the modeled result was compared to the 95% monitoring value obtained.

RESULTS AND DISCUSSION

Tier I Modeling versus Monitoring. Once model runs were completed using FIRST, it was then possible to compare the predicted exposure estimates to actual monitoring data collected. Figure 1 is the comparison of tier I predicted reservoir concentrations with actual measured values. Modeled versus monitored data are sorted from greatest overprediction to the least as paired comparisons. Results presented in Figure 2 indicate that modeling resulted in several orders of magnitude overprediction compared to actual water concentrations. To determine if there was any relationship between overprediction and model input, input factors were analyzed using a backward stepwise regression. The backward stepwise analysis was used to confirm and check that the authors had not overlooked any process that might contribute to predictions. It was determined that the total compound application rate (application rate \times number of applications) was the best indicator of model overprediction. Figure 3 is a plot of the log of total active ingredient applied to the log of overprediction (modeling/ monitoring). A regression coefficient of 0.64 (r^2) was determined



Figure 2. Tier I acute overprediction (modeling/monitoring) by compound.



Figure 3. Regression of total pounds applied to tier I acute model overprediction.

from this relationship based on a simple regression. The next step in our analysis was to make the same comparison done with the acute values using the model-predicted chronic concentrations. Figure 4 is the comparison of tier I chronic predicted reservoir concentrations with actual measured values. Modeled versus monitored data are sorted from greatest overprediction to the least as paired comparisons. Results presented in Figure 5 indicate that chronic modeling values resulted in several orders of magnitude overprediction compared to actual water concentrations. As done similarly with the acute analysis, a plot of the log of total active ingredient applied to the log of overprediction (modeling/monitoring) was created. A plot of the relationship can be found in Figure 6. A regression coefficient of 0.39 (r^2) was determined from the relationship, which was not as predictive as the acute relationship. It was not anticipated that a chronic relationship would be as predictive as the acute relationship was. A simple explanation for the differences in acute and chronic model prediction relationship to total applied compound would be that acute (instantaneous) model prediction does not take into consideration any degrada-



Figure 4. Comparison of tier I chronic model predictions versus monitoring results.



Used Maximum Monitoring Value Found

Figure 5. Tier I chronic overprediction (modeling/monitoring) by compound.

tion mechanisms, whereas a chronic model prediction does. Because chronic model prediction includes degradation mechanism processes, this can lead to sources of variance as environmental conditions change.

Tier II Modeling versus. Monitoring. To make comparisons of modeling to monitoring at tier II, the coupled exposure models PRZM and EXAMS were used consistent with EPA methodology. Once modeling was completed using PRZM/ EXAMS, it was then possible to compare the predicted exposure estimates to actual monitoring data collected. Figure 7 is the comparison of tier II predicted reservoir concentrations with actual measured values. Modeled versus monitored data are sorted from greatest overprediction to the least as paired comparisons. Results presented in Figure 8 indicate that modeling resulted in several orders of magnitude overprediction compared to actual water concentrations. To determine if there was any relationship between overprediction and model input, factors were analyzed using a backward stepwise regression as was done for the tier I modeling. On the basis of this analysis, it was determined that total application (pounds/acre) was the best indicator of model overprediction, similarly to tier I



Figure 6. Regression of total pounds applied to tier I chronic model overprediction.





Figure 7. Comparison of tier II acute model predictions versus monitoring results.

analysis. Figure 9 is a plot of the log of total active ingredient applied to the log of overprediction (modeling/monitoring). A simple regression coefficient of 0.72 (r^2) was determined from this relationship. The next step in our analysis was to make the same comparison done with the acute values using the model predicted chronic concentrations. However, chronic comparisons were made to 95th percentile monitoring values, whereas all other comparisons were made to the maximum residue value found. Figure 10 is the comparison of tier II chronic predicted reservoir concentrations with actual measured values. Modeled versus monitored data are sorted from greatest overprediction to the least as paired comparisons. Results presented in Figure 11 indicate that chronic modeling values resulted in 2-4 orders of magnitude overprediction compared to actual water concentrations. As similarly done with the acute analysis, a plot of the log of total active ingredient applied to the log of overprediction (modeling/monitoring) was made. A plot of the relationship can be found in **Figure 12**. A regression coefficient of 0.41 (r^2) was determined from the relationship, which was not as predictive as the acute relationship was. Because acute model prediction does not take into consideration any degradation



Figure 8. Tier II acute overprediction (modeling/monitoring) by compound.



Figure 9. Regression of total pounds applied to tier II acute model overprediction.

mechanisms whereas a chronic model prediction does, it was not anticipated that a chronic relationship would be as predictive as the acute relationship was. Although the findings of tier II modeling were similar to those found in the tier I work, tier II modeling regression relationships were slightly more predictive than those developed in the tier I modeling. Tier II modeling could have been expected to provide a slightly better regression relationship because it requires many more inputs, which should help increase precision.

Comparison of Tier I to Tier II Modeling. The purpose of a tiered assessment process is that at tier I a simple and quick assessment procedure is followed with the compromise of reduced predictability. At tier II, a more involved assessment process is required with the hope that a better prediction of environment exposure will be achieved. Because parametrizing a tier II model requires more data and time to obtain a prediction, a question of interest is whether the time and data requirements are worthwhile. **Figure 13** is a comparison of tier I to tier II acute model predictions. On the basis of the comparison in **Figure 13**, it is apparent that although some







compound.

reduction in overprediction was obtained using tier II modeling methodology, the reductions were not consistent or a great improvement in accuracy. Figure 14 is a comparison of tier I to tier II chronic model predictions. From the comparison in Figure 14, it is apparent that some reduction in overprediction was obtained using tier II modeling methodology; however, the reductions were not consistent or a great improvement in accuracy, either. To gain some understanding of the modeling overpredictions observed in this work, it is necessary to examine the conceptual index reservoir configured to represent actual water bodies. The EPA's index reservoir is a 172.8 ha area watershed with a 5.3 ha surface area reservoir. The reservoir is placed in the center of the watershed. The modeling includes spray drift from either aerial or ground application methods, which directly enters the reservoir from all directions. The conceptual index reservoir design does not account for runoff from fields untreated with the compound of concern. The conceptual design has no base flow into the reservoir nor dilution of runoff water concentrations predicted to leave the field





Figure 12. Regression of total pounds applied to tier II chronic model overprediction.



Figure 13. Comparison of tier I with tier II acute exposure predictions.

traveling to the index reservoir. The index reservoir that both the FIRST model and PRZM/EXAMS are meant to model has no vegetative buffers or noncropped land. There is no temporal component between when a storm event begins and when it reached the index reservoir so that as soon as a rain event occurs, it immediately enters the water-receiving body. The scenario and it is implementation are more similar to an agricultural field with an adjacent farm pond than to a drinking water reservoir watershed system. Because the watershed and reservoir system modeled are unrepresentative of an actual watershed, it should not be surprising that predictions do not approximate actual concentrations found in drinking water supply reservoirs.

Development of a Modeling to Monitoring Correction Factor. Because our backward stepwise regression analysis confirmed that there was a relationship between the total amount of compound applied in the models and their overprediction, our findings are consistent with those of other researchers (10-12). Therefore, an examination was made to determine if an overprediction correction factor could be developed. Although chronic exposure followed the trend that total pounds applied was proportional to overprediction ($r^2 = 0.41$), degradation



Figure 14. Comparison of tier I with tier II chronic exposure predictions.



Figure 15. Relationship between total active applied and PRZM/EXAMS overprediction used to develop the overprediction factor.

processes were an influence on their usefulness for development of an overprediction factor. Therefore, acute exposure predictions provided a stronger indication ($r^2 = 0.72$) that an overprediction correction factor could be developed from total pounds applied being proportional to overprediction. Acute (instantaneous) model predictions do not include the influence of compound degradation because they are meant to be instantaneous concentration values. Therefore, the influence of degradation was minimized as a confounding factor for the development of a correction factor.

The regression developed in **Figure 15** describes the relationship between total pounds applied and the tier II acute PRZM/ EXAMS model predictions.

From the relationship in Figure 15, eq 1 was developed

$$y = 2.156 + 1.03584x \tag{1}$$

where $y = \log$ of model overprediction and $x = \log$ of total active applied.

As a computational example, the dicamba use pattern has a maximum application rate of 0.5 lb/ac \times two applications for a total seasonal active application rate of 1.0 lb/ac.

Table 2. Summary of NAWQA Monitoring Results and Overprediction Factor Corrected Concentrations^a

					observed		
		total	tier II model	corrected	NAWQA (95th)		
compound (class)	EPA model scenario used	appl (lb)	conc (µg/L)	conc (μ g/L)	conc (µg/L)	п	
aldicarb (I)	CA fruit (noncitrus)	10	98.7	0.063	<0.55 (LOQ)	5980	
bromoxynil (H)	MS cotton	1.5	88.3	0.405	<0.035 (LOQ)	5887	
acifluorfen (H)	MS soybean	0.5	13.4	0.192	<0.035 (LOQ)	6014	
bentazon (H)	MS soybean	2	32.5	0.111	0.1 (LOQ)	6488	
dicamba (H)	CA alfalfa	1	32.5	0.227	<0.035 (LOQ)	6043	
imazaquin (H)	MS soybean	0.13	3.09	0.179	0.058	1214	
imazethapyr (H)	OH corn	0.1	3.06	0.232	0.028	1128	
MCPA (H)	OR wheat	4	81	0.135	<0.07 (LOQ)	6218	
2,4-D (H)	FL sugarcane	2	145	0.494	0.362	7129	
2,4-DB (H)	TX alfalfa	2	140	0.477	<0.24 (LOQ)	5938	
clopyralid (H)	OH corn	0.5	15.8	0.226	<0.23 (LOQ)	5976	
flumetsulam (H)	OH corn	0.07	2.25	0.247	<0.11 (LOQ)	1084	
oryzalin (H)	NC apples	6	161	0.176	<0.31 (LOQ)	6056	
picloram(H)	OR wheat	0.25	5.27	0.155	<0.05 (LOQ)	5763	
tebuthiuron (H)	FL sugarcane	20	1350	0.423	0.049 (LOQ)	14133	
benomyl (F)	GA Onions	3	9.34	0.021	0.024	1182	
linuron (H)	PA vegetable	2	35	0.119	<0.08 (LOQ)	259	
methomyl (I)	PA vegetable	9	16.3	0.012	<0.017 (LOQ)	105	
nicosulfuron (H)	OH corn	0.063	1.92	0.235	<0.013 (LOQ)	1089	
sulfometuron-methyl (H)	OR Christmas	0.375	1.87	0.036	0.025	1165	
terbacil (H)	LA sugarcane	2.4	125	0.352	<0.034 (LOQ)	10223	
atrazine(H)	LA sugarcane	10	438	0.282	1.4	23726	
metalaxyl (F)	GA peaches	12	101	0.054	0.04	656	
norflurazon (H)	FL citrus	7.6	215	0.184	<0.042 (LOQ)	6101	
propiconazole (F)	PA turf	7.12	125	0.114	<0.021 (LOQ)	1112	

^a These data do not include the USGS reservoir study.

Then

log of 1.0 lb/ac = 0.0 $x = \log \text{ of } 1.0 \text{ lb/ac or } 0.0$ $y = 2.156 + 1.03584 \times 0.0$

taking 10^{y} , $y \approx 143.21$ over prediction factor (modeling/monitoring)

residue correction = $1/143.21 \times \text{model prediction of } 47.4 \,\mu\text{g/L}$

or

 $1/143.21 \times 47.4 \,\mu g/L = 0.33 \,\mu g/L$

The PRZM/EXAMS scenario yielded an acute concentration of 47.4 μ g/L. Taking into account the overprediction factor, a reasonable monitoring concentration value of 0.33 μ g/L might be expected in larger bodies of water using the overprediction correction factor. The actual maximum concentration found in the USGS reservoir study was 0.19 μ g/L. To address the concern that the two year monitoring study might not adequately describe the magnitude and frequency of residues occurring in water, available monitoring data were obtained from the U.S. Geological Survey National Agricultural Water Quality Assessment program (NAWQA) data server. The data are from all available sources of surface water in the database and are presented as an indication that the data in the reservoir study are consistent with other data collected to describe pesticide residues and the analyses of ILSI and RTI. The overprediction correction factor was also applied for each of the presented compounds summarized from the NAWQA program. Based on a paired t test, the predicted concentrations after application of the overprediction factor to modeled data and the NAWQA monitoring data were not significantly different (p = 0.05). A summary of the data used in the comparison can be found in Table 2.

Conclusions. The historical development of the index reservoir model concept begins at the development of the EPA's farm pond scenario. The farm pond, like the index reservoir, has a field surrounding a water body that all of the designated area drains into. Spray drift moves into the water body at a fixed percentage of applied as well. Whereas the farm pond is a more plausible representation of an actual environment, the same representation of an actual environment for the index reservoir has many more conceptual problems. It is suggested that if the conceptual model representing an actual environment is incorrect to begin with, it should not be possible for the conceptual model to reflect actual exposure. The inability of the models and the appropriate scenarios to predict surface water concentrations is strongly indicated from our work. We suggest that the failure of the conceptual index reservoir model to reflect actual environmental factors is the dominant issue for poor model predictions. Because comparisons presented in this paper indicate that model predictions can range several orders of magnitude greater than monitoring results, where monitoring data are available, it is most scientifically sound to use them. Additionally, the use of the overcorrection should be useful to indicate more realistic exposure estimates.

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