Pesticides in Ground Water of the United States, 1992-1996

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Abstract

During the first cycle of the National Water Quality Assessment (1992–1996), ground water in 20 of the nation's major hydrologic basins was analyzed for 90 pesticide compounds (pesticides and degradates). One or more of the pesticide compounds examined were detected at 48.4% of the 2485 ground water sites sampled. However, approximately 70% of the sites where pesticides were detected, two or more pesticide compounds analyzed were present—documenting the prevalence of pesticide mixtures in ground water. The pesticide concentrations encountered were generally low, with the median total concentration (summation of concentrations for the 90 pesticide compounds) being 0.046 µg/L. Pesticides were commonly detected in shallow ground water beneath both agricultural (60.4%) and urban (48.5%) areas. This is an important finding because, although agricultural activities have long been associated with pesticide contamination, urban areas have only recently been recognized as a potential source of these types of compounds. Pesticides with higher frequencies of detection were generally those with more extensive use, greater environmental persistence, and greater mobility in ground water (lower soil-water partition coefficients).

Introduction

In 1991, the U.S. Congress appropriated funds for the U.S. Geological Survey's (USGS) National Water-Quality Assessment (NAWQA) Program to determine the quality of the nation's water resources. The NAWQA Program is being conducted in 59 areas (or "study units"), which include some of the nation's largest river basins and aquifers (Gilliom et al. 1995; Leahy et al. 1990). Collectively, these study units encompass about 50% of the conterminous United States, as well as parts of Alaska and Hawaii. Intensive investigations in about one-third of the study units are ongoing at any particular time. Sampling within the first group of 20 study units was conducted between 1992 and 1996. This paper provides an initial summary of the results from the sampling of ground water for pesticides in those units.

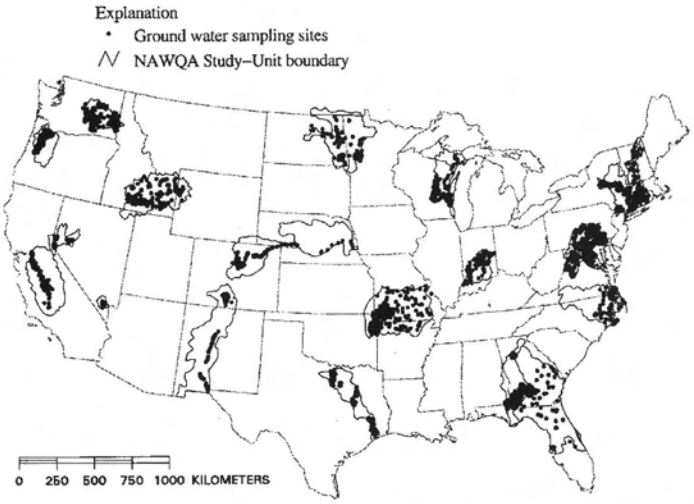

Figure 1. Locations of the 2485 ground water sites sampled during the first 20 NAWQA study units.

Materials and Methods

Sampling Sites

More than 3000 ground water samples were collected from a total of 2485 sites from the first 20 NAWQA study-unit investigations and analyzed for pesticides from 1992 through 1996 (Figure 1). About 1400 of the sites sampled, however, were derived from recently recharged shallow ground water systems. To eliminate the bias caused by sites having more than one pesticide analysis, only one pesticide sample per site was used for this summary of the data. This selection was based on the water sample that had the greatest number of pesticide compounds analyzed. These ground water sites consisted primarily of monitoring (1090 sites), domestic (1025 sites), and public-supply (170 sites) wells, but also included a small number of other well types (i.e., irrigation, stock), springs, and tile drains. To ensure that the results between study units are comparable, the NAWQA design employs consistent guidelines for the selection of either existing wells or locations for the installation of new wells for sampling (Gilliom et al. 1995; Lapham et al. 1995; Scott 1990).

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Table 1
Select Results for Pesticide Compounds Examined for This Investigation
[N, number of sites; % Detect, frequency of detection, Max Conc, maximum concentration]

Compound	N	% Detect	Max. Conc. (µg/L)	Compound	N	% Detect	Max. Conc. (µg/L)
2,4-D	2306	0.43	0.54	ethalfluralin	2459	0.12	0.09
2,4-DB	2306	0.04	0.06	ethoprop	2460	0.04	0.009
1,2-dichloropropane	1834	1.14	7.50	fenuron	2301	0.04	0.14
1,3-dichloropropene, cis	1831	0	<0.20	fonofos	2459	0.12	0.059
1,3-dichloropropene, trans	1831	0	<0.20	fluometuron	2317	0.34	0.43
2,6-diethylaniline	2459	0.65	0.049	HCH, alpha	2459	0.04	0.059
3-hydroxycarbofuran	2301	0	<0.014	HCH, gamma	2459	0.12	0.032
2,4,5-T	2306	0	<0.035	linuron	2459	0.08	0.029
1,2,3-trichloropropane	1831	0.76	1.10	malathion	2459	0.28	0.10
acetochlor	1064	0.09	0.023	MCPA	2306	0	<0.17
aciflurofen	2308	0.04	0.19	MCPB	2306	0	<0.14
alachlor	2460	2.07	3.80	methiocarb	2315	0	<0.026
aldicarb	2306	0.04	0.01	methomyl	2258	0	<0.017
aldicarb sulfone	2258	0.09	0.32	methyl bromide	1831	0.06	0.50
aldicarb sulfoxide	2258	0.31	1.80	methyl parathion	2459	0.08	0.062
atrazine	2460	30.0	4.20	metribuzin	2459	1.91	0.30
azinphos-methyl	2451	0.16	0.18	metolachlor	2460	12.0	5.40
benfluralin	2459	0.12	0.006	molinate	2459	0	<0.004
bentazon	2306	1.04	6.40	napropamide	2460	0.16	0.07
bromacil	2318	1.04	14.0	neburon	2315	0	<0.015
bromoxynil	2306	0	<0.035	norflurazon	2317	0.04	0.22
butylate	2459	0.24	0.004	oryzalin	2315	0.09	0.08
carbaryl	2460	0.81	0.036	oxamyl	2255	0	<0.018
carbofuran	2460	0.57	1.30	parathion	2459	0	<0.004
chloramben	2317	0	<0.42	pebulate	2459	0.12	0.052
chlorothalonil	2305	0	<0.48	Pendimethalin	2459	0.08	0.012
chlorpyrifos	2459	0.37	0.013	permethrin	2459	0.08	0.007
clopyralid	2293	0	<0.23	phorate	2459	0	<0.002
cyanazine	2460	1.38	0.16	picloram	2257	0.04	2.20
dacthal	2459	0.65	10.0	prometon	2460	11.6	40.0
dacthal, mono acid	2305	0.04	1.10	propachlor	2460	0.12	0.004
DBCP	1831	0.76	2.90	pronamide	2460	0.08	0.052
DDE	2454	3.30	0.006	propanil	2460	0.49	0.015
deethylatrazine	2460	28.4	2.60	propargite	2459	0.08	0.009
diazinon	2459	1.30	0.16	propham	2316	0	<0.035
dicamba	2305	0.13	0.21	propoxur	2286	0.09	0.30
dichlobenil	2317	0.30	0.21	silvex	2308	0	<0.021
dichlorprop	2306	0.09	0.01	simazine	2460	14.8	1.30
dieldrin	2459	1.63	0.068	tebuthiuron	2460	2.72	0.79
dinoseb	2307	0.22	7.90	terbacil	2441	0.49	0.33
disulfoton	2459	0	<0.017	terbufos	2459	0.08	0.012
DNOC	2306	0	<0.42	thiobencarb	2459	0	<0.002
Diuron	2313	1.95	2.00	triallate	2459	0.20	0.002
EDB	1831	0.27	1.40	triclopyr	2309	0	<0.25
EPTC	2459	0.77	0.45	trifluralin	2459	0.32	0.014

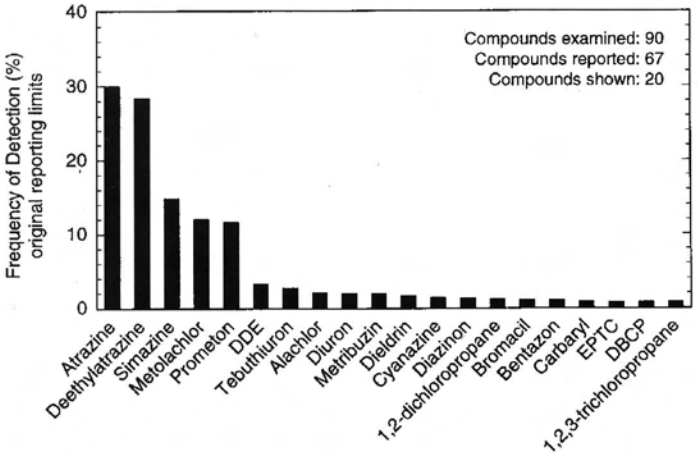

Figure 2. Frequencies of detection of select pesticide compounds in ground water sites sampled during the first 20 NAWQA study units.

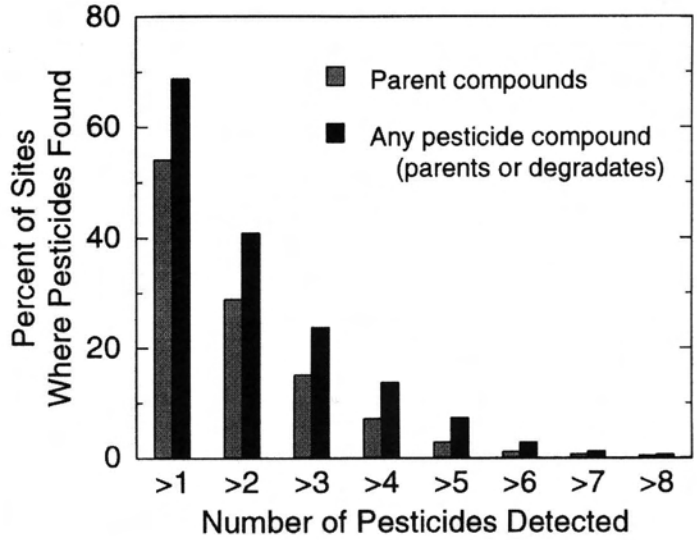

Figure 3. Number of pesticide compounds detected at a given site.

Sampling and Analytical Procedures

All samples were collected by USGS personnel using established protocols and procedures designed to obtain water samples representative of the targeted aquifer (Koterba et al. 1995). Before water samples were collected, each site was pumped until field-measured values of pH, water temperature, specific conductance, and dissolved-oxygen concentration stabilized.

All of the samples analyzed for pesticide compounds were collected in an enclosed chamber to prevent airborne contamination. Following collection, all of the pesticide samples were chilled and shipped to a laboratory for chemical analysis. The target list of pesticide compounds included 52 herbicides, three herbicide degradates, 22 insecticides, five insecticide degradates, seven fumigants, and one fungicide (Table 1). These pesticides represent those that have, or have had, extensive agricultural or nonagricultural use in the United States, are a degradation product of the aforementioned compounds, are of potential toxicological concern, and are sufficiently sensitive to the analytical methods (Rose and Schroeder 1995; Werner et al. 1996; Zaugg et al. 1995) used. The method detection limits for these compounds generally ranged from 0.001 to 0.20 µg/L. Additional details on the compounds analyzed and their method detection limits are available via the Web at http://water.wr.usgs.gov/pnsp/anstrat. Not all 90 pesticide compounds were measured in every ground water sample collected.

Ancillary Data

For each pesticide, the total amount of active ingredient applied to agricultural crops and pasture (Gianessi and Anderson 1996) was estimated within a 1 km radius of each ground water sampling site by multiplying the amount applied within the county by the fraction of the cropland and pasture area in the county that was located within 1 km of the site. This approach probably underestimated the total use for each pesticide because no data are currently available on applications to nonagricultural areas other than overall national estimates. All of the data on selected physical and chemical properties for each pesticide (aerobic soil half-lives, field dissipation half-lives, and K_{oc}) were obtained from the U.S. Department of Agriculture, Agricultural Research Service's (USDA-ARS) Pesticide Properties Database (U.S. Department of Agriculture 1995), with the exception of acetochlor, where properties were obtained from the U.S. Environmental Protection Agency's "One-Liner Database" (U.S. Environmental Protection Agency 1994). When multiple values were available in the USDA-ARS database, the value recommended by the database authors was used. Field dissipation half-lives were used for those compounds for which laboratory-based aerobic soil half-lives were not available. A significance (α) level of 0.05 was used for all statistical tests.

Results

Pesticide compounds were commonly detected in ground water sampled in the first 20 NAWQA study units investigated. In fact, at least one of the 90 compounds examined was found at 48.4% of the 2485 ground water sites sampled between 1992 and 1996. Sixty-seven pesticide compounds were detected (Figure 2). Concentrations, however, were generally low, with the median total concentration (summation of concentrations for the 90 pesticide compounds) being 0.046 µg/L. The maximum concentrations measured for each pesticide compound are listed in Table 1.

An interesting result of this study was the prevalence of pesticide mixtures in ground water. For this study, 69% of the ground water sites with pesticide detections had more than one compound present, 24% had more the three compounds present, and 3% had more than six compounds present (Figure 3). As many as 13 pesticide compounds were found in a single ground water sample. The most frequent two-compound mixture was atrazine and deethylatrazine (an atrazine degradate). At 49.4% of the sites with pesticide detections, both atrazine and deethylatrazine were present in the water sampled. In fact, in only 11.5% of the sites with pesticide detections was atrazine detected in the absence of deethylatrazine, and in only 8.4% was deethylatrazine detected in the absence of atrazine. Because deethylatrazine has similar acute (Kaufman and Kearney 1970) and chronic (Babic-Gojmerac et al. 1989) toxicity as atrazine, the presence of both compounds in ground water likely increase the health and environmental risks involved. Thus, future research on the toxicity of pesticides to humans and other nontarget organisms should consider the effects of mixtures of the pesticide compounds and the effects of individual pesticides. Indeed, previous studies have shown that the toxic effects of some combinations of pesticides may be additive or, in some cases, synergistic

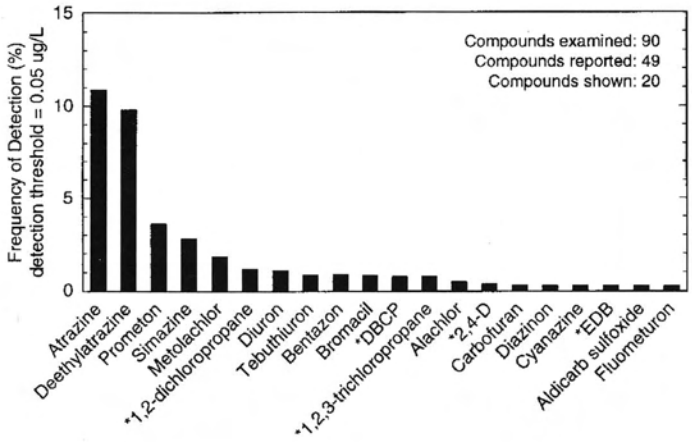

Figure 4. Frequencies of detection of select pesticide compounds in ground water sites sampled during the first 20 NAWQA study units, adjusted to a common detection threshold of 0.05 µg/L. Compounds marked with an * had method detection limits greater than 0.05 µg/L.

(Marinovich et al. 1996; Pape-Lindstrom et al. 1997; Thompson 1996).

The pesticide degradates examined had an insignificant effect on the overall frequency of pesticide detection. The frequency of detection decreased slightly from 48.4% when both parent compounds and degradates are included to 44.3% when just the parent compounds are considered. However, the effect of degradates on the mixture of pesticide compounds present was more apparent (Figure 3). Most of this effect was the result of frequent detections of the deethylatrazine. It is felt that pesticide degradates would have had a much more pronounced effect on both the overall frequency of detection and the number of mixtures present if other degradates documented to be commonly detected in ground water (Kolpin et al. 1998b; Kolpin et al. 2000) could have been included in this study.

Correcting for Variations in Detection Limit

The data must be corrected to account for variations in method detection limits among the different compounds before the underlying factors responsible for the differences in detection frequency among the pesticide compounds can be examined. Not surprisingly, frequencies of detection have been found to be inversely related to reporting limits, not only for individual studies but also among different studies with similar sampling designs (Burkart and Kolpin 1993). However, because a large number of the pesticide compounds were detected only at concentrations lower than the highest detection limit among the 90 compounds examined (0.42 µg/L), a value intermediate between the highest and lowest detection limits was employed as a common detection threshold. Thus, for this study, a common detection threshold of 0.05 µg/L was selected as a compromise between the need to accommodate comparisons with compounds exhibiting higher detection limits and the desire to minimize the loss of information on compounds detected only at low concentrations. This approach, however, probably caused detection frequencies to be underestimated for those compounds having a method detection limit greater than 0.05 µg/L, relative to those with detection limits less than or equal to this value (Figure 4).

Variations in Compounds Examined

Previous research has shown that the intensity of use, mobility through the subsurface (as quantified using the water/soil organic carbon partition coefficient, or K_{oc}) and environmental persistence may all affect the likelihood of detecting pesticides in ground water (e.g., Barbash and Resek 1996; Kolpin et al. 1998a). For this study, frequencies of pesticide detection in ground water (adjusted to 0.05 µg/L) were found to be significantly related to the estimated amount of agricultural use within a 1 km radius of each of the sampled sites (ρ [Spearman's rank correlation coefficient] = 0.286; P = 0.013). This significant, but weak, correlation could probably have been substantially strengthened with use, if it had been possible to account for nonagricultural use.

Variations in the adjusted frequencies of detection among the pesticide compounds were also found to be significantly related to variations in their persistence and subsurface mobility. Frequencies of detection were positively related to aerobic soil half-life (ρ = 0.323, P = 0.003) and inversely related to K_{oc} (ρ = -0.319, P = 0.003). Thus, the pesticides that were more persistent and had greater subsurface mobility were generally detected more frequently in ground water.

Improvements in explaining the frequencies of pesticide detection determined from this study are obtained by using a multivariate approach, taking information on both estimated use and physical properties into account. All variables were log-transformed for this analysis, which resulted in relatively normal distributions. The amount of variance in the log-transformed frequencies of pesticide detection that was explained by these parameters was found to be 11% for aerobic soil half-life alone (R^2 = 0.11), 24% for aerobic soil half-life + K_{oc} (R^2 = 0.24), and 27% for aerobic soil half-life + K_{oc} + estimated agricultural use (R^2 = 0.27). The results of this multivariate analysis suggests that a compound's aerobic soil half-life (relative persistence) and K_{oc} (relative mobility) have a more pronounced effect on determining pesticide occurrence in ground water (accounted for more of the variation in detection frequency) than chemical use. As previously mentioned, the affect of use on pesticide occurrence may have been more pronounced if total use (both agricultural and nonagricultural) could have been obtained. This multivariate analysis also demonstrates that although a chemical's persistence, mobility, and use affect the transport of pesticides to ground water, 73% of the variance in pesticide detection frequencies was unaccounted for. Thus, other factors such as hydrogeology, soil properties, ground water age, and agricultural management practices must also be considered.

Variations Among Land-Use Settings

One objective of NAWQA is to determine the effects of specific land-use and hydrogeologic settings on the quality of shallow ground water. The two principal land-use settings examined during the first 20 NAWQA study-unit investigations were both associated with frequent detections of pesticides in the underlying shallow ground water—agricultural (60.4%) and urban (48.5%). This is an important finding because although agricultural areas have long been identified as a source of pesticide contamination in ground water, urban areas have only recently been recognized as such a source on a national scale (USGS 1999).

The frequencies of detection were statistically higher under agricultural than under urban areas for herbicides (57.6% versus 44.3%; P < 0.001; Wilcoxon rank sum test) and fumigants (2.8% versus 0.3%; P = 0.01; Wilcoxon rank sum test). There was, however, no statistical difference in the frequency of insecticide detection between areas underlying agricultural (7.4%) and urban areas (10.0%; P = 0.144, Wilcoxon rank sum test).

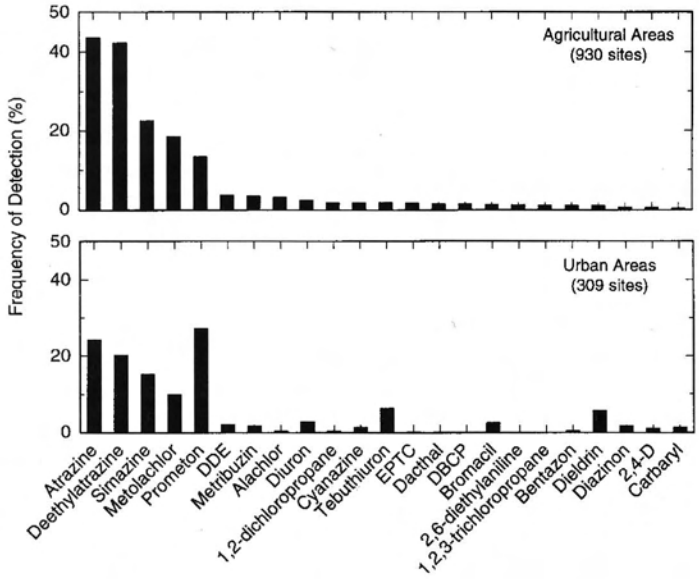

Figure 5. Frequencies of detection of individual pesticides in shallow ground water beneath two different land-use settings. Only compounds having detection frequencies exceeding 1.0% in at least one of these settings were included.

Because of the considerable differences between agricultural and urban settings with respect to the types of pesticides used and the methods and timing of their application, it is not surprising that the pesticides detected varied by land use (Figure 5). Atrazine, a triazine herbicide primarily used in corn production, and deethylatrazine, a degradate of atrazine, were the most frequently detected compounds in shallow ground water within agricultural areas (Figure 5). Whereas prometon, a triazine herbicide primarily used for various nonagricultural purposes, was the pesticide detected most frequently in urban areas.

A statistical comparison (Mann-Whitney rank sum test) between agricultural and urban areas identified significantly ($P < 0.05$) fewer occurrences under urban areas for atrazine, deethylatrazine, simazine, metolachlor, alachlor, EPTC, Dacthal, and DBCP than under agricultural areas. With the exception of DBCP, a soil fumigant, these compounds are herbicides primarily used on crops and orchards to increase production. Conversely, significantly ($P < 0.05$) more occurrences under urban areas were identified for prometon, tebuthiuron, dieldrin, and carbaryl than under agricultural areas. These compounds are an equal mix of herbicides and insecticides and represent chemicals that have—or have had—substantial use on noncropland areas such as rights-of-way and residential lawns.

It should be noted that some of the pesticide detections identified under agricultural and urban areas may have been derived from other mechanisms in addition to recent chemical use. Possible mechanisms include atmospheric deposition (Goolsby et al. 1997; Larney et al. 1999; Majewski et al. 1998), historical use from previous agricultural and urban activities (Spencer et al. 1996; Wade et al. 1998), and transport from nearby areas via runoff or subsurface migration (Burkart et al. 1999).

Significance to Human Health

Pesticides were commonly detected in shallow ground water for this study, but their concentrations rarely exceeded current drinking water criteria established by the U.S. EPA for the protection of human health (U.S. EPA 1996). The concentration of DBCP exceeded its established maximum contaminant level (MCL) of 0.2 μg/L at 14 sites, dieldrin concentrations exceeded the 10^{-6} risk specific dose of 0.02 μg/L at 12 sites, EDB concentrations exceeded its established MCL of 0.05 μg/L at five sites, 1,2-dichloropropane exceeded its established MCL of 5.0 μg/L at two sites, atrazine concentrations exceeded its established MCL of 3.0 μg/L at two sites, cyanazine concentrations exceeded its established health advisory level of 1.0 μg/L at one site, and dinoseb concentrations exceeded its established MCL of 7.0 μg/L at one site.

However, the relative infrequency with which pesticides exceeded drinking water criteria may not provide a complete assessment of the overall health risks associated with the presence of pesticides in ground water. First, water quality criteria for the protection of human health have been established for only 56 of the 90 compounds examined for this study. Second, these drinking water criteria consider the effects only of individual compounds and do not account for the presence of more than one pesticide compound. As noted earlier, the presence of pesticide mixtures was a common phenomenon (Figure 3). Third, other pesticide compounds not examined for this study—particularly pesticide degradates—have been detected in ground water (Barret 1996; Kolpin et al. 1996; Kolpin et al. 1998; Potter and Carpenter 1995) that also could have potential acute (Kaufman and Kearney 1970; Tessier and Clark 1995) and chronic (Babic- Gojmerac et al. 1989) health effects. Fourth, recent research also suggests that some pesticide compounds may cause health problems at levels considered safe by current standards (Biradar and Rayburn 1995; Sawicki et al. 1995).

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