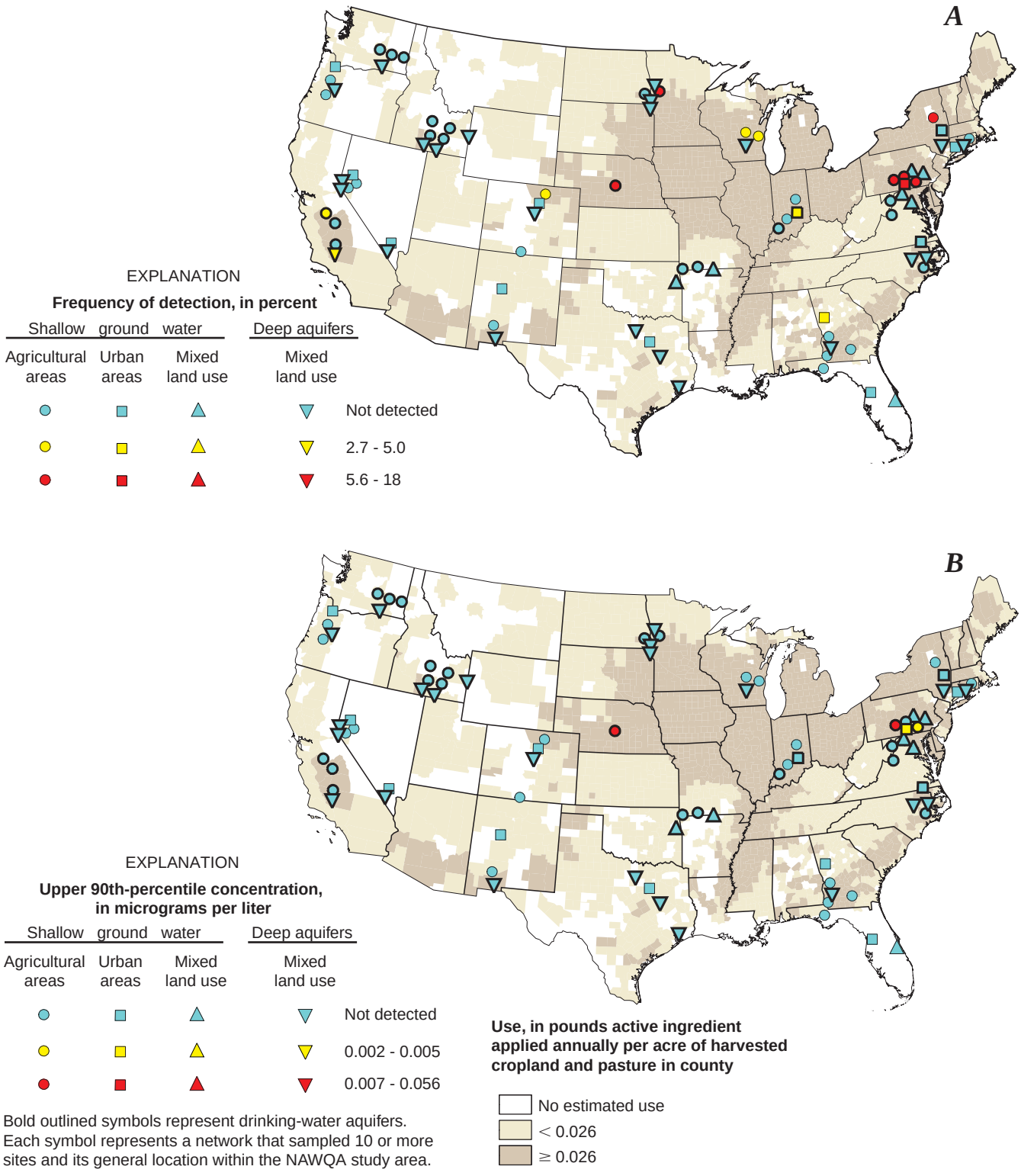

Figure 31. Cyanazine occurrence in ground water for the NAWQA study in relation to agricultural use (A) Frequencies of detection. (B) Upper 90th-percentile concentrations. See figures 3 and 4 for areas sampled.

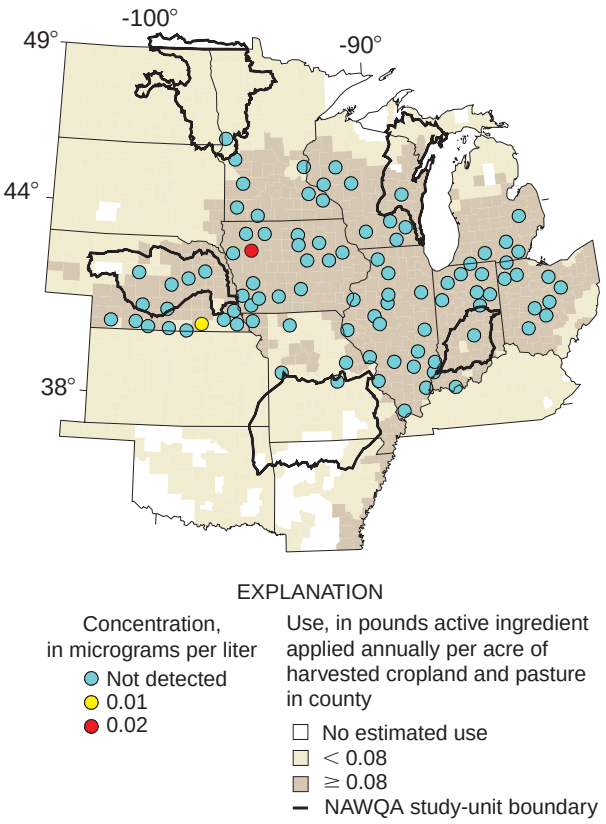

Figure 32. Concentrations of cyanazine in near-surface aquifers of the northern midcontinent for the 1992 sampling of the MWPS in relation to agricultural use.

In accord with the exclusively agricultural use of cyanazine (table 2), most of the NAWQA studies that detected the herbicide in ground water were located in regions of high agricultural use (fig. 31*A*), although the detections occurred at relatively low concentrations (figs. 30 and 31*B*). While several of the studies with cyanazine detections were in urban settings, as noted earlier these detections might have been the result of atmospheric or subsurface transport of the herbicide from applications in nearby agricultural areas. The MWPS results for cyanazine (fig. 32) were consistent with these results, with few detections, even in areas of high use.

As with alachlor, the low frequencies of cyanazine detection, even in areas where its agricultural use is considerable, may be a result of its comparatively rapid rate of transformation in aerobic soil (table 3). This hypothesis is supported by the relatively high frequency with which one of its principal degradates, cyanazine amide, was encountered in ground water in the northern midcontinent; this compound was detected in ground water at or above 0.05 µg/L nearly five times as frequently as cyanazine during the MWPS (table 11), and more than 10 times as frequently as cyanazine at or above 0.20 µg/L during the statewide sampling in Iowa (table 12 and fig. 12). The low rates of detection for cyanazine may also have been partly a result of voluntary reductions in its use over the past decade.

Acetochlor

At present, the use of acetochlor is restricted largely to the corn-growing areas of the Central Lowland and northern High Plains, with some additional use in Oregon and Delaware. Analyses for acetochlor were carried out at 953 of the sites sampled for the NAWQA study (table 4), but the herbicide was detected in only two locations; one well sampled for the cnbrsus1 study in central Nebraska (see table 8) with a concentration of 0.11 µg/L, and one well sampled for the nvbrlusag2 study in the Carson Desert of western Nevada with a concentration of 0.023 µg/L. These detections occurred in areas where the herbicide was known to have been used (Gianessi and Anderson, 1995). No acetochlor was detected at or above 0.05 µg/L in any of the 38 wells sampled for the MWPS during the summer of 1994 (table 13).

The low frequencies of detection reported for acetochlor are consistent with the fact that sampling for the herbicide by the USGS studies took place relatively soon after it was first registered for use in the United States in 1994. Indeed, differences in the timing of sampling relative to the first applications of acetochlor may explain why it was not detected at or above 0.05 µg/L in shallow ground water by the MWPS during the summer of 1994, but was detected above this concentration in shallow ground water—though not in deeper aquifers—by Kolpin and coworkers during statewide sampling in Iowa in the summer of 1995 (Kolpin and others, 1997) and the summer of 1996 (table 12). These findings agree with results from several field studies, discussed by Barbash and Resek (1996), indicating that some pesticides may reach shallow ground water in detectable concentrations within the first year following their application.

As noted earlier for alachlor and cyanazine, however, the low frequencies of acetochlor detection in ground water may also be related to its comparatively high rate of transformation in soil (table 3), a point noted previously by Kolpin and others (1996a) for the northern midcontinent. As with alachlor and

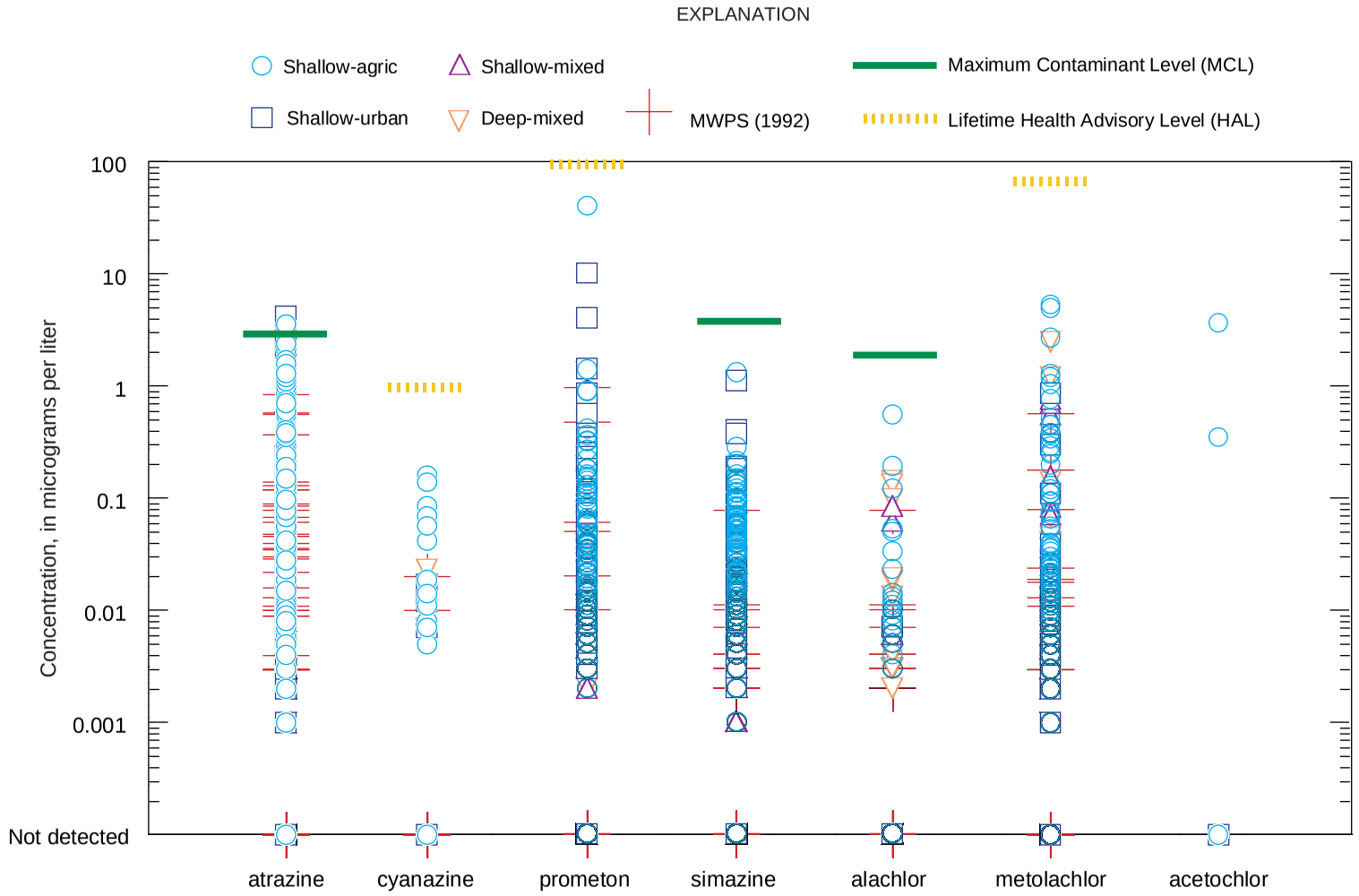

Figure 33. Concentrations of herbicides measured in ground water at individual sites during the NAWQA and MWPS investigations in relation to drinking-water criteria. Lifetime health advisory level (HAL) shown for herbicides for which no maximum contaminant level (MCL) has been established (U.S. Environmental Protection Agency, 1996). No drinking-water criteria have yet been established for acetochlor. agric, agricultural.

cyanazine, this hypothesis is supported by the high frequencies of detection of acetochlor degradates, relative to the parent compound, during the statewide sampling in Iowa by Kolpin and others (1998b); acetochlor ESA was detected at or above 0.20 µg/L over eight times as frequently, and acetochlor OA more than three times as frequently, as the parent compound during the Iowa study (table 12).

Comparisons of Observed Concentrations with Drinking-Water Criteria

Because pesticides were usually detected at relatively low concentrations, drinking-water criteria—that is, those established for the protection of human health—were rarely exceeded in ground-water samples collected during the USGS investigations. Among the seven herbicides of interest to this report, maximum contaminant levels (MCL), which are legally enforceable standards, have been set for only atrazine, simazine, and alachlor (fig. 33). Of the four other herbicides, lifetime health-advisory levels (HAL), which are nonenforceable guidelines, have been established for cyanazine, prometon, and metolachlor; to date, neither an MCL nor an HAL have been set for acetochlor. These criteria (MCL or HAL) were exceeded only for atrazine during the NAWQA study (fig. 33), at two of the 2,227 sites sampled. No drinking-water criteria for any of these herbicides were exceeded during the 1992 phase of the MWPS. For the NAWQA investigation, both of the sampling sites where the MCL for atrazine (3 µg/L) was exceeded were shallow (LUS) wells. One was located in an agricultural area; the other was a well used for drinking water in an urban area.

Limitations of Existing Drinking-Water Criteria for Assessing Overall Health Risks

Comparisons of pesticide concentrations measured in the hydrologic system with criteria established for the protection of drinking-water quality provide an initial approximation of the level of concern that might accompany the detection of these compounds in water resources. However, the low frequencies of drinking-water criterion exceedance observed during the USGS studies discussed here may underestimate the overall health risks associated with the presence of these pesticides and degradates in shallow ground water for several reasons. First, water-quality criteria for the protection of human health have been established for only a relatively small number of all pesticides registered for use. For example, as noted earlier, among the seven herbicides of interest, enforceable standards (MCLs) have been established for only three (fig. 33); HALs, which have been specified for three of the other herbicides, are guidelines recommended for use "in the absence of regulatory limits" (Nowell and Resek, 1994). Second, the drinking-water criteria consider only the effects of individual pesticides and do not account for the additional effects of other pesticides or degradates that might be present. As shown earlier in figure 9 and table 9, detections of more than one pesticide at a given site were relatively common during both the NAWQA and MWPS investigations, and recent research has indicated that some combinations of pesticide compounds may show additive or even synergistic toxicity (Marinovich and others, 1996; Thompson, 1996). Third, other pesticide compounds not examined by either study, including degradates of several of the parent compounds that were investigated, have been detected in ground water (for example, Potter and Carpenter, 1995; Barbash and Resek, 1996; Barrett, 1996; Kolpin and others, 1997, 1998b) that could also cause adverse health effects (for example, Kauffman and Kearney, 1970; Babic-Gojmerac and others, 1989; Tessier and Clark, 1995; Bain and LeBlanc, 1996; Reddy and others, 1997). Finally, drinking-water criteria do not account for potential impacts on the health of aquatic ecosystems into which contaminated ground water may discharge (for example, Squillace and others, 1993; Kim and Hemond, 1998).

SUMMARY AND CONCLUDING REMARKS

As part of an effort to initiate and develop Pesticide Management Plans (PMPs) for selected pesticides, the U.S. Environmental Protection Agency is currently evaluating information regarding the occurrence and distribution of five high-use herbicides in ground water of the United States—atrazine, cyanazine, simazine, alachlor, and metolachlor. (At the time of writing, however, the removal of cyanazine from this list was under consideration.) This report provides an overview of data on detections in ground water for these five compounds, along with two chemically related herbicides (prometon and acetochlor), primarily on the basis of the results from two

recent multistate studies by the U.S. Geological Survey (USGS)—the National Water-Quality Assessment (NAWQA) Program and the Midwest Pesticide Study (MWPS). These two investigations detected the five PMP herbicides and prometon in drinking-water aquifers and other shallow ground water in a variety of agricultural and nonagricultural settings across the Nation. Acetochlor, the use of which began in 1994 in the United States, was detected at only two of the 991 sites sampled for the herbicide by the two programs through 1995; its detection within this time period, however, supports the observation from previous studies that pesticides can sometimes be detected in ground water within the first year following their application.

Consistent with the results from previous large-scale studies of pesticide occurrence in ground water, more than 98 percent of the pesticide detections during these USGS studies were at concentrations less than 1 μg/L. Consequently, criteria for the protection of drinking-water quality were rarely exceeded. However, these guidelines may underestimate overall health risks because they (1) have been established for only a relatively small number of pesticides, (2) do not account for additive or synergistic effects among combinations of pesticides, (3) neglect the potential toxicity of pesticide degradates, and (4) do not consider effects on aquatic ecosystems influenced by ground-water discharge. Multiple pesticide detections at individual sampling locations were common during the USGS studies; among all of the sites examined in this report for the NAWQA and MWPS investigations, 19.7 and 13.8 percent, respectively, had detections of two or more of the seven herbicides. Furthermore, degradates were detected frequently—in many cases, more often than their parent compounds.

The likelihood of detecting a particular pesticide in ground water is dependent upon a broad range of natural and anthropogenic factors (for example, climate, soil properties, hydrogeologic setting, well construction, pesticide properties, rates of pesticide use, and other agricultural management practices), as well as study design. The examination of relations between these factors and pesticide occurrence in ground water during the NAWQA Program is being carried out in a stepwise fashion. After correcting for many of the confounding effects of study design (through the use of consistent procedures for well selection, sampling and chemical analysis across the Nation, and a common analytical reporting limit among the compounds examined), the first factor examined in this regard was pesticide use. Limitations on the available data on use, however, restricted this analysis to the five PMP herbicides, on the basis of nationwide use estimates for nonagricultural settings, and county-level use estimates for agricultural areas.

Frequencies of detection in shallow ground water beneath urban areas during the NAWQA study were significantly higher for the PMP herbicides with greater nonagricultural use nationwide ($P=0.026$; simple linear correlation). In agricultural settings for both the NAWQA and MWPS studies, the frequencies of PMP herbicide detection in shallow ground water were generally higher in areas of more intensive agricultural use, but the strength of this relation varied considerably among different compounds and different regions of the country. Of the five PMP herbicides, statistically significant relations between agricultural use and the frequency of detection at or above 0.01 μg/L in shallow ground water beneath agricultural areas during NAWQA, on the basis of simple linear correlations of log-transformed parameters, were observed only for metolachlor ($P=0.0006$) and atrazine ($P=0.003$). Nonparametric correlations between detection frequency and use in agricultural settings were statistically significant ($P<0.05$; Spearman rank correlations) for all of the PMP herbicides except for simazine. The absence of statistically significant relations between occurrence and use for simazine was caused largely by the fact that it was detected at relatively high frequencies in areas where its reported agricultural use was low or zero, perhaps because of substantial use in nonagricultural settings or, in at least two of the study areas, extensive irrigation.

Frequencies of alachlor and cyanazine detection in many agricultural areas were considerably lower than would have been anticipated from their extensive agricultural use in these settings. The comparatively rapid rates at which both herbicides undergo transformation in aerobic soils, coupled with the frequent occurrence of their degradates in ground water, suggest that the infrequent detections of these herbicides may have been a reflection of their relatively low environmental persistence. The infrequent detections of acetochlor may also have been caused by its low field persistence, but an alternate explanation for the low detection rates for this herbicide is that its use did not begin until 1994, after the NAWQA sampling had commenced. The observation of a highly significant linear correlation between detection frequency and agricultural use for

metolachlor—despite an aerobic soil half-life comparable to those for alachlor and cyanazine, which did not show such a relation—may have been a statistical consequence of the considerably higher use of metolachlor in many of the study areas.

The examination of relations between PMP herbicide detections in shallow ground water and the various natural and anthropogenic factors with which such detections may be associated was extended beyond agricultural use through a multiple correlation analysis of the NAWQA results involving a subset of these factors. The frequencies of detection of the five PMP herbicides in shallow ground water beneath the agricultural areas were significantly correlated with their agricultural use in each of the sampled areas and with their aerobic soil half-lives ($P \leq 0.0001$ for both parameters), but not with the predicted mobilities of the compounds in ground water (as approximated by their soil organic carbon partition coefficient, or K_{oc}; $P=0.19$) or the median well depths of the sampled networks ($P=0.72$).

The highly significant correlation between detection frequency and aerobic soil half-life is consistent with the aforementioned hypothesis that the infrequent detections of alachlor and cyanazine, particularly in areas of high agricultural use, may have been a result of their comparatively low persistence in soil. The absence of significant relations with well depth or K_{oc} was attributed to the relatively narrow range examined for both of these parameters. In addition to these effects, results from the MWPS and other investigations also indicate that frequencies of detection are generally higher following periods of pesticide application or enhanced recharge.

The fact that variations in pesticide persistence and agricultural use accounted for less than 40 percent of the variability in the frequencies of PMP herbicide detection in shallow ground water beneath agricultural areas demonstrates the need to incorporate other parameters into this analysis. Future examination of the NAWQA data will involve consideration of additional natural and anthropogenic factors associated with pesticide detections in ground water, including those relating to soil properties, hydrogeologic setting, climate, and agricultural management practices.

The observations from these studies underscore the need for more detailed information on pesticide use and on the occurrence of pesticide degradates in ground water. Limitations on current information regarding the spatial distributions of pesticide use in the United States, particularly for pesticide applications in nonagricultural settings, may have contributed to the relatively poor geographic correspondence often seen between herbicide detections and use across the Nation during the multistate USGS studies. The limited data on the occurrence of selected degradates for some of the herbicides provided a more complete picture of the effects of pesticide use on ground-water quality, indicating that for several of the herbicides examined, particularly those that are less persistent, some degradates may be detected considerably more frequently than their parent compounds. Chemical analyses during ground-water monitoring studies should, therefore, routinely include the major degradates for the parent compounds of interest, especially for those pesticides that are more reactive. The incorporation of more explanatory factors, as well as refinements in the data on pesticide use and more extensive coverage of degradates, will help advance current understanding of how environmental and land-use setting influence the likelihood of detecting pesticides in ground water after they are applied to the land.

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