The Journal of Engineering Geology, Vol.3, No.3, December., 1993., pp.241-252

# Irregular Distribution of Lead in Groundwater in Door County, Wisconsin
# 위스컨신주 도어지역의 지하수내 납성분의 불규칙한 분포에 관한 연구

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## Abstract/요약

Lead has been found in the groundwater in Door County, Wisconsin, with temporally and spatially irregular distribution in concentration. Correlation coefficients were calculated among lead indicators in groundwater(frequency of lead detections, mean and maximum concentration of lead detections) and seven independent variables(stucture and geographic factors of wells, hydrogeological factors at lead-arsenate mixing sites and the level of soil contamination) which are possibly related to the lead level in groundwater. The significance of relationships was determined statistically by a t-test at the 90% confidence level, and indicated that the spatially located lead-arsenate mixing sites provided the lead in groundwater in the study area.

A total of 112 groundwater samples were collected from 5 house wells with previous lead detects. Lead was detected in particles on filter papers with 0.45 μm pore size, but not in filtrates. The result of chemical analysis for lead indicates that lead is associated with particles in groundwater in Door County. Subsequently, the irregular distribution of lead in the county results from the transport of particulate lead along the advective groundwater movement through the preferential pathways such as vertical and bedding-plane joints.

미국 위스컨신주 도어 지역의 지하수내에서 납성분이 시기적, 장소적으로 불규칙하게 산출되었다. 본 연구에서, 지하수의 납성분의 오염도를 나타내주는 지시자들(검출빈도, 납성분의 평균, 최고 검출농도)과 지하수의 납 오염에 관계가 있을 가능성이 있는 7개의 독립변수들과의

상관계수가 계산되었다. 이 관계의 통계적 중요성은 t-test에 의하여 90%의 신뢰도에서 이루어졌으며, 이 지역에 산재하는 농약(lead-arsenate)의 처리장소가 지하수내 납성분의 오염 원인을 통계학적으로 입증하였다.

납성분의 산출농도가 높았던 5개의 가정우물로부터 총 112개의 지하수시료가 채취분석되었다. 납성분은 0.45μm의 공극 크기를 지닌 여과지에 걸러진 미립자에서 검출되었으며, 여과액에서는 검출되지 않았다. 이러한 결과는 납성분이 이 지역의 지하수내에서 미립자의 상태로 존재함을 지시한다. 따라서, 이 지역에서 납성분의 불규칙한 산출은 미립자상의 납이 수평과 수직으로 발달된 파쇄대와 같은 유동경로를 따라 이동하면서 나타나는 현상으로 고려된다.

## INTRODUCTION

From 1983 to 1986, the Wisconsin Department of Natural Resources(WDNR) sampled 333 drinking water supply wells in Door county, Wisconsin(Fig. 1 ; Stoll, 1986). Lead detections occurred in 169 samples out of 446 groundwater samples in 119 wells. The WDNR conducted a continuous groundwater survey from the 33 wells in the county, which showed lead detections of at least 20 μg/l in concentration during the previous study by Stoll(1986). Subsequently, the 33 wells were called the worst cases of lead contamination.

In fact, the concern over groundwater contamination in Door County results from the discovery of abandoned lead-arsenate mixing sites. The WNDR identified at least 35 abandoned mixing sites around the county(Fig. 1). Lead-arsenate was a dominant pesticide of the fruit growing industry in the county from the late 1800's to as late as the early 1970's. Lead-arsenate was obtained in insoluble powder and mixed with water before application to orchard areas(Cremlyn, 1978). Mixing and handling activities usually occur in designated areas, called mixing sites. Today the sites still contain relatively high concentrations of lead and arsenic

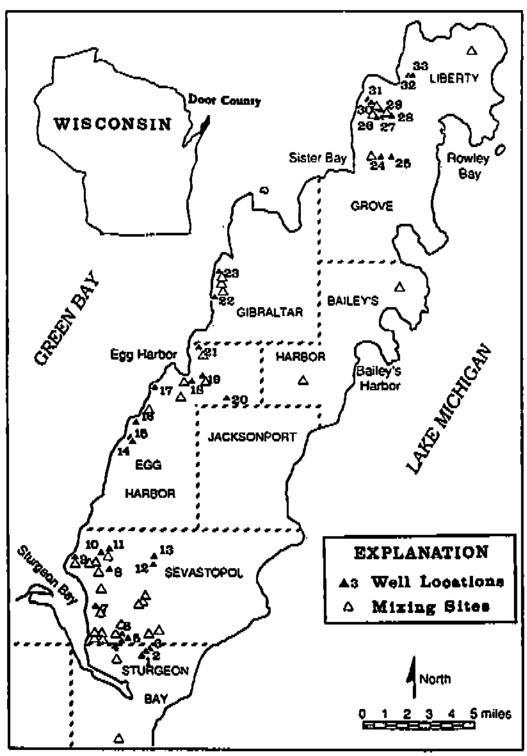


Fig. 1 Locations of Stoll's 33 worst case lead-contaminated wells and 35 identified lead-arsenate mixing sites in Door county, Wisconsin(Modified from Stoll, 1988)

in the surrounding soils, ranging from 20 to 48,000 mg/kg and from 20 to 19,000 mg/kg, respectively. The background levels of lead and arsenic in soils in the northern county are 10 mg/kg and 3 mg/kg, respectively(RMT, 1987). Soil contamination by lead and arsenic is vertically limited to the top 20 to 25 cm(Wiersma and Stieglitz, 1989). Therefore, the high concentrations of lead and arsenic in soils at the mixing sites pose strong potentials of being sources of groundwater contamination by the chemicals.

However, the detection of lead in groundwater has not been consistent, either temporally or spatially. Concentration variations occurred rapidly in a given well. The well with the highest contamination of lead in 1987(2400 μg/l) had concentrations below the detection limit(3 μg/l) at other times(Stoll, 1988). Lead concentrations in several water-supply wells have changed by as mush as 40 μg/l within a three day period. In addition, while wells with high lead concentrations tended to be near the lead-arsenate mixing sites, which were presumed to be the sources of lead in groundwater, many other nearby wells had no lead detection.

Therefore, this study was objected to (1) answer the question of the mixing sites being the source of lead in groundwater, and (2) explain the unpredictable occurrences of lead in the groundwater system of the county.

## HYDROGEOLOGIC SETTING

The Silurian dolomite aquifer system includes the Niagaran aquifer and the underlying Alexander aquifer, and is the most important source of water supply to wells in the county (Fig. 2). Unconfined conditions predominate in the upper part of the Niagaran aquifer due to the abundance of vertical joints(Sherrill, 1978). Bedding-plane joints transmit most of the water into the lower part of the Niagaran aquifer and the Alexandrian aquifer. The major orientations of the vertical joints are at azimuths of approximately 70° and 155°(Rosen, 1984 ; Craig, 1989).

As the dolomite is recharged, water moves downward through an unsaturated zone. When the water reaches the zone of saturation, the

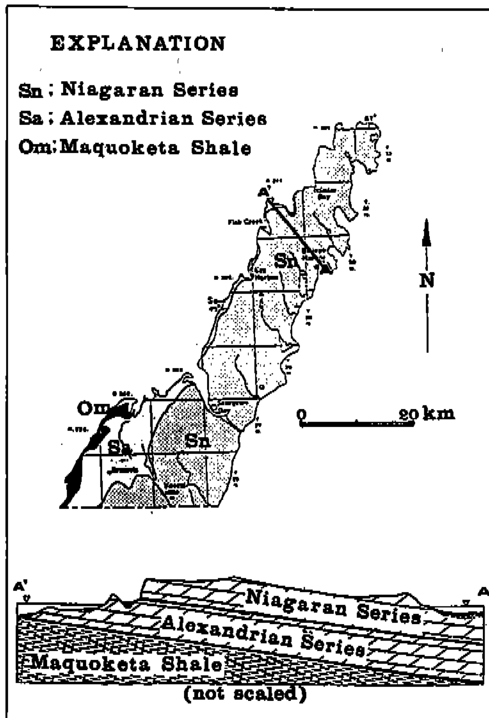


Fig. 2 Bedrock geology(top) and geologic cross section(bottom) of Door County, Wisconsin(modified from Craig, 1989 and Sherrill, 1978)

movement becomes nearly horizontal along bedding-plane joints. Bradbury and Muldoon(1992) measured hydraulic conductivity($K$) of the dolomite aquifer by slug tests and obtained ranges of $K$ from $1.8\times10^{-4}$ to 95 m/day with a geometric mean of $8.1\times10^{-2}$ m/day. The wide range of $K$ reflects the heterogeneity of the dolomite aquifer.

## METHODS

### Statistical Approach

**Variables** : A statistical study was undertaken to identify statistically significant relations among 10 variables, which are potentially related to lead in groundwater. The variables include :

1. the frequency of lead detections in groundwater(%)
2. the mean concentration of lead detections in groundwater(μg/l),
3. the maximum concentration of lead detections in groundwater(μg/l),
4. the casing depth of a well(m),
5. the total depth of a well(m),
6. the distance from the well to HWY 42 (km),
7. the distance from the well to the nearest upgradient mixing site(km),
8. the thickness of soil at the nearest upgradient mixing site(m),
9. the mean concentration of lead(mg/kg) in the top 15 cm of soils at the nearest upgradient mixing site, and
10. the depth to water level(m) at the nearest upgradient mixing site.

**Data Acquistition** : Variables 1 to 3 are indicators of lead level in groundwater. As it is conceived that contaminants would predominantly travel horizontally along bedding-plane joints in the lower part of the Silurian dolomite aquifers(Sherrill, 1978), information on well construction was selected for variables 4 and 5. Data for variables 1 to 5 for the 33 worst cases and locations of specific wells were obtained from the WNDR's report(Appendix I ; Stoll, 1988).

Distance from wells to HWY 42(variable 6) was chosen to examine the possibility of atmospheric deposition from combustion of leaded gasoline as the source of lead in groundwater (Appendix I). HWY 42 connects major towns along the western part of northern Door County, thus major industrial and sightseeing traffic occurs on this highway. Garrels et al. (1975) estimated that about 99% of lead in the atmosphere results from combustion of leaded gasoline.

Distances for variables 6 and 7(distance from wells to the nearest upgradient mixing sites) were measured on the county map along directions of groundwater flow and major vertical joints(Appendix I). Groundwater flow direction was determined based on the Sherrill's(1978) potentiometric surface map of the county(Fig. 3). Distances were measured on the lines drawn parallel to joint directions(70° and 155° in azimuth ; Rosen, 1984 ; Craig, 1989), assuming an even distribution of vertical joints in this area.

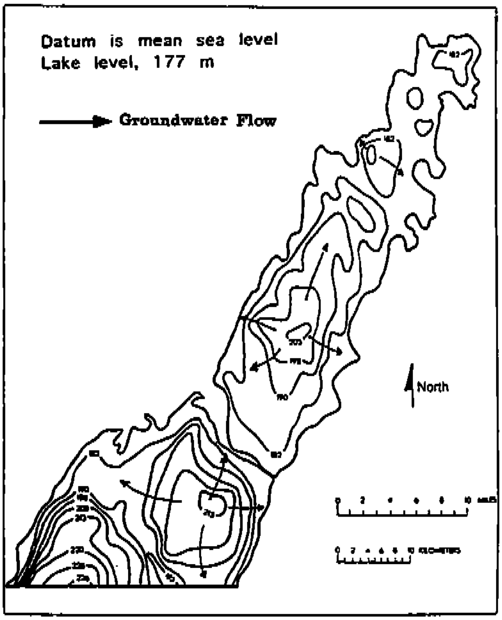


Fig. 3 Potentiometric surface map of Door County, Wisconsin (From Sherrill, 1978)

Upgradient mixing sites were not identified for nine(12,13,14,15,16,17,20,32,33) out of the 33 worst case wells. The nine wells may have upgradient mixing sites, but the sites have not been identified by the WDNR because of no obvious remnant of mixing facilities above the ground. They were excluded in the search for a possible relationship of lead in groundwater to upgradient mixing sites.

The thickness of soils at mixing sites were calculated from the topographic map and the bedrock surface map of Door County(Sherrill, 1978). Lead concentration in soils at mixing sites were obtained from RMT's(1987) report. Depths to water level at mixing sites were calculated from elevations of mixing sites and water level based on sherrill's(1978) potentiometric surface map. Data for these variables are presented in the Appendix I.

**Statistical Analysis :** Correlation coefficients (r) among variables were calculated from data for all variables with a computer program, STATS-2(STATSOFT, 1985). An inherent bias was introduced by using only the 33 worst case wells. However, information on lead indicators in wells(variables 1-3) were available from only those wells.

Because of significant temporal and spatial variations in lead detections, it was difficult to pick up one variable as a representative indicator for level of lead contamination in groundwater. Therefore, variables 1, 2 and 3 were separately correlated with the other independent variables. In addition, two correlation coefficient sets were calculated with different numbers of cases, because information on the total depth and the casing depth was not available from 14 out of 33 worst case wells and upgradient mixing sites were not identified for nine wells.

The statistical significance of the relationship was determined by a t-test at the 90% confidence level(Till, 1974). The equation is given as,

$$t=r\times SQRT\{(n-2)/(1-r^2)\}$$

where n denotes the number of cases, and (n−2) the degree of freedom. This equation could be converted to,

$$r=t/SQRT(n-2+t^2)$$

The t value can be obtained for 90% confidence level in the Student t-distribution table, subsequently $r_{90}$(correlation coefficient showing the significance of the relation at the 90% level) could be calculated.

### Groundwater Sampling

Groundwater samples were collected from five house water-supply wells(1,5,9,13 and 34 in Fig. 4) in the Sturgeon Bay and Sevastopol areas. Wells 1, 5, 9 and 13 have shown relatively high lead detections in previous studies by the WNDR(1988), and well 34, the newly constructed one, posed strong potential of lead contamination due to a nearby mixing site. The samples included three components : water samples filtered through 0.45 μm pore size Whatman membrane filters, unfiltered counterparts and filter cakes. Samples were collected from the nearest tap to the pump after flushing for at least 10 minutes to remove possible contaminant residues in the pump tank. During the groundwater survey for lead and arsenic by the WDNR(Stoll, 1988), water samples were collected after three to five minutes of flushing. This study's protocol was established to exceed the WDNR's flushing but to remain relatively consistent with it.

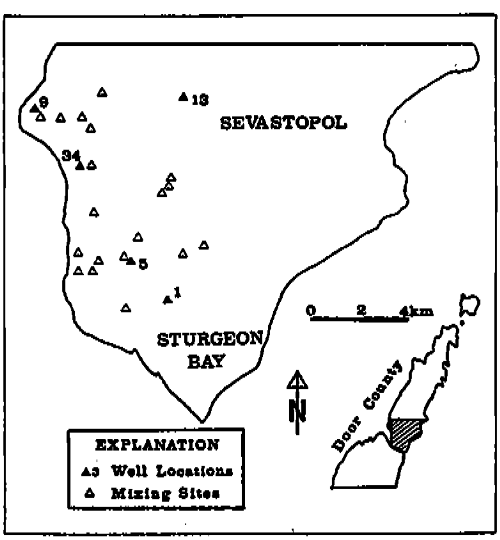


Fig. 4 Location of sampled wells and known mixing sites in Sturgeon Bay and Sevastopol area, Door County.

Water samples were collected into a 1-gallon (3.8 l) plastic jug and immediately filtered through a pre-weighed 0.45 μm filter. The amount of filtered water varied from 1,500 to 3,500 ml depending on filtering time. Pre-weighed filters were stored in sealed plastic Petri dishes to prevent hydration before use. Approximately 500 ml of filtered water was acidified with sufficient concentrated nitric acid(trace metal grade) to bring the solution's pH below 2, and eventually analyzed for lead. Filter cakes were saved in the original Petri dishes.

For quality control of the sampling procedure, complete field blanks were also taken using distilled deionized(DSDI) water from a 5-gallon plastic jug, which was brought to the field for cleaning equipment and mixing titration reagents. These blank samples could indicate if sampling procedures contributed any lead.

### Analysis of Groundwater and Particles on Filter

Lead in groundwater was analyzed by graphite furnace atomic absorption spectrometry (FAAS) with detection limits of 0.3 μg/l, following the APHA standard methods(#3113; American Public Health Association, 1989). Concentrations of lead were calculated from calibration curves prepared with standard stock solutions.

The filters were digested in 125-ml Erlenmeyer flasks with concentrated nitric acid according to APHA standard method # 3030E. The resulting solutions were analyzed by FAAS for lead. Field and laboratory blanks with DSDI water and a blank filter were also prepared through the same steps for quality control.

## RESULTS AND DISCUSSION

### Source of Lead in Groundwater

Statistically significant relations at 90% confidence level were identified between lead indicators in groundwater and the other variables. When correlation coefficient(r) is equal to or greater than 0.36 in the set of 14 cases and 0.26 in that of 24 cases, the relations between variables are significant at a 90% confidence level(Appendix II). In case of inverse correlations, the coefficient(r) would be negative.

A negative correlation was found between mean concentration of lead(variable 2) and the distance to the nearest mixing site(variable 7) (Table 1). This represents that lead levels in groundwater varied inversely with the distance between wells and mixing sites.

A negative correlation was also found between mean and maximum concentration of lead(Table 1) and depth to water table at mixing sites. This implied that lead concentrations

Table 1. Significant relations between lead indicators in groundwater and other variables

| Variables* | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
|---|---|---|---|---|---|---|---|
| 1 | | | | | + | − | |
| 2 | + | + | | − | + | | − |
| 3 | + | | | | + | | − |

*Variables include:

1. the frequency of lead detection,
2. the mean concentration of lead detection,
3. the maximum concentration of lead detection,
4. the casing depth of well,
5. the total depth of well,
6. the distance from the well to the nearest highway(HWY 42)
7. the distance from the well to the nearest upgradient mixing site,
8. the thickness of soil at the nearest upgradient mixing site,
9. the mean concentration of lead in soils at the nearest upgradient mixing site, and
10. the depth to water level at the nearest upgradient mixing site.

in groundwater increased where the nearest mixing site has a shallow water level.

Positive correlations with the total and casing depths(Table 1) represented that lead concentrations became higher in deeper wells with longer casing depths. The relations suggest that lead travels horizontally with groundwater flow along the lower part of the Silurian dolomite.

Relations between lead indicators in groundwater and the thickness of soil at mixing sites (Table 1) were also positive. This probably indicates that contaminated soils are the major source of lead-carrying particles in the groundwater system. Consequently, the relations imply the possibility of the particle transport of lead in groundwater in the study area.

A negative relation was found between frequency of lead detection in groundwater and lead concentration in soils(Table 1). The relation is anomalous and not understood.

No significant relation was found between lead indicators in groundwater and the distance from worst case wells to HWY 42(Table 1), which is a major source of lead in the atmosphere from the combustion of leaded gasoline in this area. Assuming no significant effect of wind direction, this implies that lead in groundwater did not originate from atmospheric deposition due to leaded gasoline.

Another possible source of lead in water samples is plumbing in old houses. In this study, however, water samples were collected after 10 to 15 minutes of flushing. Therefore, introduction of possible residual lead from the pump tank was assumed to be negligible based on the flushing process. In addition, Stoll's(1988) survey showed significant temporal variation in lead concentration at particular wells. A Pb-plumbing source would not produce such temporal inconsistency, therfore, plumbing is assumed not to be the lead source.

The other possibility is that lead in groundwater originated from surface spills of gasoline and leakage from underground fuel tanks. Because information on locations of spills and underground tanks is not available, it cannot be determined whether the possibility is reasonable.

In summary, the lead-arsenate mixing sites are indicated to be sources of lead in groundwater in Door County by the statistically significant relations between lead indicators in groundwater(in frequency, mean and maximum of lead concentration) and the distance to the nearest upgradient mixing site and the thickness of soil and the depth to water table at the mixing site.

### Distribution of Lead in Groundwater

A total of 56 groundwater samples and 56 filter cakes were collected from five house wells(Fig. 4). For lead, each filtered groundwater sample was analyzed three times and the standard deviation was calculated by the built-in program in the FAAS. The results were examined using a Q-test(Skoog and West, 1971) to identify outliers at the 95% confidence level. No lead was detected in field and laboratory blanks.

No lead was found in the filtered groundwater samples. Lead detections, however, occurred on 44 out of 56 particle samples filtered from groundwater(Table 2). This indicates that lead is not in the dissolved state, but sorbed onto particles in groundwater. Mean Pb concentrations ranged from 0.3 to 4.3 μg/mg, and individual sample detection varied from 0.2 to

Table 2. Particle analysis for lead in samples 5 house wells Sturgeon Bay and Sevastopol areas, Door County

| Wells | 1 | 5 | 9 | 13 | 34 | Total |
|---|---|---|---|---|---|---|
| # of samples | 8 | 15 | 9 | 15 | 9 | 56 |
| # of Pb detect | 2 | 9 | 9 | 15 | 9 | 44 |
| Mean Pb conc.(ug/mg) | 0.3 | 0.3 | 4.3 | 0.8 | 1.3 | |

7.1 μg/mg at the detection limit of 0.1 μg/mg.

Since lead is associated with particles in groundwater, its detections would occur only where particles could pass. The hydrogeological settings, well developed vertical and bedding-plane joints in the Silurian aquifer, probably contributes major part of the unpredictable occurrences of lead in study area. Wells connected with joints, where groundwater flows through dynamically with lead-carrying particles, could show higher and more frequent detections of lead than those without connection. Since groundwater flow through the joints would be much faster than through the pores in aquifer media, the spatial distribution of lead would be rather irregular following the joint system.

The temporal variation of lead concentration in a given well could also be explained by the particulate nature of lead in groundwater. Lead concentrations would depend on the amount of lead-carrying particles in groundwater. The amount of particles originated from surface sources would be fluctuated by the particle-introducing mechanism into the groundwater system such as recharge events. Subsequently, the temporal variation of lead occurrences in groundwater could be complexed by the randomness of the recharge events and the particle travel-time in the groundwater system with joints.

## CONCLUSION

From the statistical analysis of the numerical data and groundwater sampling and analysis for lead, following conclusions were drawn in this study.

(1) Abandoned lead-arsenate mixing sites are the major sources of lead in groundwater in Door County, Wisconsin. Relations between lead indicators in groundwater(detection frequency, mean and maximum concentrations) and other variables related to the lead-arsenate mixing sites(the casing and total depth of a well, the distance to the nearest upgradient mixing site, the depth to the water level and the soil contamination by lead at the mixing site) were statistically significant at the 90% level.

(2) Lead is associated with particles in groundwater in Door County, Wisconsin.

(3) Particulate phase of lead is considered to produce spatial and temporal variation in lead detections in Door County, because particles would move advectively in groundwater along the preferential flowpaths such as joints under influences of recharge events in occurring times.

## ACKNOWLEDGEMENTS

I am deeply grateful to R. Stoll and K. Erdmann in the Wisconsin DNR at Green Bay, Wisconsin, USA. Without their cooperation, this study would not have been possible. Thanks to K. Graff and P. Anderson in the Center for Great Lake Studies for their invaluable help during laboratory analysis. Special thanks to Professors D. Cherkauer and T. Grundl in the

Dept. of Geosciences, UW-Milwaukee, for their assistance and encouragement.

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Appendix I.

Data for calculation of correlation coefficient(r) for variables related to groundwater contamination by lead in Door County, Wisconsin

| Well | Variables* | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
| I.D | (%) | (ppb) | (ppb) | (m) | (m) | (km) | (km) | (m) | (mg/kg) | (m) |
| 1 | 99 | 22.1 | 180 | 52.7 | 73.5 | 1.9 | 1.4 | 3.0 | 203 | 12.2 |
| 2 | 4 | 3.0 | 8 | 52.7 | 77.4 | 2.1 | 2.3 | 1.8 | 382 | 11.6 |
| 3 | 20 | 4.3 | 43 | NA** | NA | 2.2 | 1.9 | 1.8 | 382 | 11.6 |
| 4 | 55 | 17.5 | 120 | 76.2 | 99.1 | 0.2 | 0.5 | 3.0 | 643 | 16.5 |
| 5 | 99 | 89.5 | 2400 | 76.5 | 91.7 | 0.3 | 0.5 | 3.0 | 643 | 16.5 |
| 6 | 7 | 3.7 | 21 | 51.8 | 70.7 | 1.0 | 0.6 | 0.9 | 340 | 40.8 |
| 7 | 19 | 3.5 | 10 | 51.8 | 97.5 | 3.0 | 0.5 | 0.9 | 906 | 25.9 |
| 8 | 14 | 3.6 | 25 | 59.1 | 89.6 | 2.4 | 0.8 | 0.6 | 6764 | 21.9 |
| 9 | 98 | 12.9 | 34 | 41.1 | 60.0 | 4.8 | 0.5 | 0.9 | 1283 | 53.3 |
| 10 | 70 | 7.2 | 140 | NA | NA | 2.7 | 0.8 | 0.6 | 6764 | 21.9 |
| 11 | 8 | 3.0 | 3 | NA | NA | 2.1 | 0.8 | 0.9 | 491 | 46.3 |
| 12 | 47 | 10.4 | 160 | 51.8 | 71.3 | 1.3 | NA | NA | NA | NA |
| 13 | 26 | 9.3 | 150 | 76.5 | 96.6 | 1.3 | NA | NA | NA | NA |
| 14 | 19 | 4.2 | 42 | 47.2 | 62.8 | 1.6 | NA | NA | NA | NA |
| 15 | 21 | 3.8 | 11 | NA | NA | 1.9 | NA | NA | NA | NA |
| 16 | 13 | 3.2 | 5 | NA | NA | 2.2 | NA | NA | NA | NA |
| 17 | 13 | 3.2 | 7 | 27.4 | 36.6 | 2.4 | NA | NA | NA | NA |
| 18 | 85 | 13.0 | 100 | NA | NA | 0.3 | 0.3 | 1.8 | 350 | 52.4 |
| 19 | 10 | 3.7 | 20 | NA | NA | 0.2 | 1.1 | 2.1 | 849 | 31.7 |
| 20 | 94 | 17.4 | 370 | 56.7 | 97.5 | 2.6 | NA | NA | NA | NA |
| 21 | 100 | 9.1 | 17 | NA | NA | 0.5 | 1.1 | 0.9 | 6700 | 45.7 |
| 22 | 37 | 19.6 | 210 | NA | NA | 0.7 | 0.5 | 0.9 | 393 | 36.0 |
| 23 | 48 | 4.0 | 16 | 58.5 | 79.6 | 0.8 | 0.6 | 0.9 | 63 | 42.1 |
| 24 | 3 | 3.0 | 3 | NA | NA | 1.8 | 1.1 | 0.9 | 303 | 36.0 |
| 25 | 78 | 7.6 | 77 | NA | 187.0 | 2.6 | 1.9 | 0.9 | 303 | 36.0 |
| 26 | 96 | 7.8 | 33 | 46.6 | 92.4 | 0.3 | 1.4 | 0.9 | 575 | 49.4 |
| 27 | 71 | 11.9 | 78 | NA | NA | 0.7 | 0.6 | 0.9 | 575 | 49.4 |
| 28 | 48 | 5.3 | 33 | 6.1 | 58.5 | 1.0 | 1.3 | 0.9 | 575 | 49.4 |
| 29 | 19 | 3.7 | 15 | NA | NA | 0.2 | 1.0 | 0.9 | 575 | 49.4 |
| 30 | 3 | 3.1 | 10 | 52.7 | 70.7 | 1.0 | 0.6 | 0.9 | 855 | 48.2 |
| 31 | 40 | 10.1 | 160 | 52.7 | 74.4 | 1.3 | 1.0 | 0.9 | 855 | 48.2 |
| 32 | 20 | 3.5 | 13 | NA | NA | 0.2 | NA | NA | NA | NA |
| 33 | 5 | 3.1 | 6 | 46.9 | 55.2 | 0.3 | NA | NA | NA | NA |

*Variable index is given in the Table 1.

**NA indicate that data were not available.

Appendix II.

Correlation coefficients for variables possibly related to lead contamination of groundwater in Door County, Wisconsin

Set 1* : 14cases

| Variables** | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
|---|---|---|---|---|---|---|---|
| 1 | −0.1 | −0.07 | 0.05 | 0.04 | 0.41 | −0.39 | 0.03 |
| 2 | 0.4 | 0.27 | −0.28 | −0.24 | 0.67 | −0.2 | −0.38 |
| 3 | 0.38 | 0.27 | −0.31 | −0.23 | 0.56 | −0.15 | −0.34 |

Set 2* : 26cases

| Variables | 6 | 7 | 8 | 9 | 10 |
|---|---|---|---|---|---|
| 1 | −0.02 | −0.1 | 0.26 | 0.09 | 0.11 |
| 2 | −0.22 | −0.26 | 0.59 | −0.14 | −0.29 |
| 3 | −0.21 | −0.21 | 0.51 | −0.11 | −0.3 |

* Different number of cases in set 1 and 2 resulted from missing information on well design and apparent upgradient mixing sites.

** Variable list is given in the Appendix 1.

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