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Dye study tracks historical pathway of VOC-bearing industrial waste water from failed pond at metals coating facility

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ABSTRACT

Nearly ten years after initiating a ground-water assessment and documenting a stable mass of VOC-bearing ground water in the upper karst aquifer, the Post-Closure Care regulatory process required a dye study to trace ground-water movement. The regulatory impetus was the location of a major spring, regionally downgradient from the facility, used as a municipal drinking water supply.

A well drilled strategically in the area of highest concentrations of VOCs of the stable plume was used as the point of injection of fluorescein. Charcoal packets and water samples were collected from 29 locations (on-site monitoring wells, monitoring wells and productions wells at an adjacent facility, surface water sites, and the major spring – a diffuse flow, conduit spring with three points of emission). The bedrock fracture system in the vicinity of the facility is not well developed or well connected. The distances from the injection point to monitoring/sampling locations ranged from 45.7 meters (150 feet) to 4.9 kilometers (3.05 miles). The duration of the test, including pre-injection monitoring, was 118 days.

A cocktail of potable water (7,182 liters or 1900 gallons) preceded the injection of the dye solution, and a chaser of about 4,158 liters (1100 gallons) followed the injection of fluorescein. The initial movement of the dye was a combination of the head created by the injection of drinking water and dye, and the non-pumping conditions at the adjacent facility. Within less than 24 hours, the imposed head dissipated and water levels in the injection well returned to pre-injection levels.

The head imposed or induced with injection resulted in an almost unique occurrence that pushed water out to the northwest fractures, in pathways other than normal. The only other time that those northwest pathways were probably used, temporarily completing the connection between the facility and the major spring, was when the pond failed, and a slug of VOC-bearing water was induced into the ground-water system.

HYDROGEOLOGIC SETTING AND CONTAMINANT CHARACTERIZATION

The geology at the site is typical of the terrain associated with limestone bedrock. At the site, the irregular surface of the limestone is overlain by a mantle of clay-rich, unconsolidated material (overburden). Ground water in the limestone aquifer, occurs under semi-confined to confined conditions. Variations in the direction of ground-water movement at the site are controlled by pumping of production wells at the adjacent manufacturing facility, immediately west of the site.

Movement of ground water containing volatile organic compounds (VOCs) occurs within fractures/bedding in the upper portion of the bedrock, and locally within the chert-rubble zone at the bedrock-overburden contact. Measured hydraulic conductivities in bedrock, observations that the abundance of fractures and ground water decrease with depth, and analyses of ground water from deep wells demonstrate the presence of little, if any, VOCs at depths exceeding 27 meters (90 feet) below land surface. The horizontal and vertical distribution of Tetrachloroethylene (PCE) in ground water has been assessed and monitored using the wells installed at the site and adjacent to the facility. Monitoring of ground water since 1989 indicates that the mass of VOC-bearing groundwater has been spatially stable.

PCE in ground water at the facility is most probably the result of an historical (1977) failure of a wastewater pond, which was near the southwestern corner of the facility. The bedrock pinnacle, which underlies the northern edge of the former pond, provided a potential pathway for PCE to reach the chert-rubble zone at the bedrock-overburden contact (Figure 1). PCE has been confirmed in the chert-rubble zone in this area (south of the pinnacle) at some of the highest concentrations identified in the sub-soil at the site. The chert-rubble zone in this area is within the zone of fluctuation of the potentiometric surface as a result of pumping at the adjacent manufacturing facility and/or seasonal fluctuations in the water surface.

Ground-water monitoring and the distribution of PCE in soil south of the bedrock pinnacle suggest that this compound is sorbed to soils near the top of bedrock or sorbed to soils in fractures near the top of bedrock. No evidence of Dense Non-Aqueous Phase Liquids (DNAPL) has been identified in soil or ground water at the site. Fluctuation of the water surface, as a result of pumping at the adjacent facility and recharge at the bedrock pinnacle, provides a flushing mechanism which desorbs some PCE from soil, supplies dissolved PCE to ground water, and maintains the spatially stable VOC mass.

The entire course of the nearest perennial stream was observed by canoe within a 1.6 km (1-mile) radius of the facility. No springs were observed along this portion of the creek, nor were any springs observed along the entire 10.5 km (6.5-mile) course, which was traversed. The stream has been sampled at locations upstream and downstream of the facility. No VOCs have been detected.

The system of fractures (joints) in the vicinity of the site consists of two sets, which trend toward the northeast and northwest. Both sets of fractures occur throughout the area. However, the northwest-trending set is dominant, as evidenced by the alignment of the stream valley, and alignment of karst depressions in the area.

A major spring (Spring), 4.9 kilometers (3.05 miles) northwest of the site, is a municipal water supply. The Spring is considered a diffuse flow conduit spring with emissions visible at three nearby locations. The estimated 7-day Q_2 (low flow) is 34 million liters (9 million gallons) per day (mgd) and average flow is about 158.76 million liters per day (42 mgd). The computed variability in flow is 200 percent. Water is withdrawn by the treatment plant by a 24-inch intake pipe approximately 3 meters (10 feet) from the largest flowing of the springs. The plant is permitted by the state environmental agency as a surface water filtration plant. The recharge area of the Spring is 218 km^2 (84 square miles). The quality of water at the spring is influenced by many potential sources of inorganic and organic constituents.

THE INJECTION WELL

A recovery well, designated RW-1, was installed to extract ground water with elevated concentrations of PCE and degradation products and to prevent the migration of PCE-containing ground water from the source area. The well was also installed for use as the injection well for this dye study.

A borehole for the well was advanced through the overburden (residuum) from land surface to the top of bedrock. To aid in the characterization of hydrogeologic conditions, soil samples in the overburden were collected continuously using a split-spoon sampler, and bedrock was drilled by air rotary method as opposed to previously specified coring. The air rotary method was used, after reconsideration of coring using drilling water and/or drilling mud, so as to enable easy recognition of water-bearing zones and to eliminate the potential for the introduction of drilling fluids into the ground-water system. Lithologic descriptions based on returns (drill cuttings), rig behavior and penetration rate, and geophysical logging allowed the identification of fracture zones at 7-8.84 m (23 to 29 feet), 12.80 to 15 m (42 to 49 feet, 16.76 to 17.37 m (55 to 57 feet), 23.45 to 23.77 m (77 to 78 feet) and, 25.60 to 26.21 m (84 to 86 feet) below land surface. The total depth drilled was 29.38 m (96.38 feet) below land surface. The borehole was drilled to 8 inches in diameter to facilitate installation of 4-inch diameter PVC casing and screen for the recovery well. The well was screened from 18.60 to 29.26 m (61 to 96 feet) below land surface.

THE DYE TEST

The injection of dye was timed to be dependent on the hydrologic conditions in the vicinity of the injection well. As tracing the potential connectivity from the site to the Spring was the primary focus of the study requirements, hydrologic conditions conducive to dye migration off site should occur at, or just prior to, dye injection. Ground-water withdrawals at the adjacent manufacturing facility occur on an as-needed basis during the operation of the plant and affect the movement of ground water at the site. The pumps are not typically operating during weekends and holidays. To simulate conditions that would be most conducive to off-site ground-water movement, the dye was injected on December 22, 1996, after a period of 2 non-operation (non-pumping) days at the manufacturing facility.

Natural or man-made background fluorescence of the ground water was monitored prior to injection of the dye (background concentration). Passive dye detectors (charcoal packets) were placed at selected locations, including wells, surface water, springs and storm sewers 18 days prior to injection of the dye at well RW-1. Some potential sources for background fluorescence are detergents, bathroom cleaners, pigments for inks and dyes, antifreeze, industrial wastes, naturally-occurring mineral fluorescence, and residual dye from previous studies. The first set of charcoal packets for the background portion of the dye was installed on December 4 through 6, 1996. The detectors were removed for evaluation at 9 days and 2 days prior to injection of the dye, (on December 13 and December 20, respectively).

On December 22, 1996, between 15:16 hours and 15:31 hours, 2.27 kg (5 pounds) of fluorescein (powdered fluorescein mixed with a total of 26.46 liters or seven gallons of distilled water) were injected in well RW-1 at the site. A drop hose was installed in the injection well (RW-1) such that the bottom opening was within the screened interval of the well and approximately 3 m (10 feet) below the top of bedrock. A slug of potable water (7,182 liters or 1900 gallons) preceded the injection of the dye-containing mixture (dye and water) via two drop hoses. Care was taken to avoid contamination of clothing and the area around the point of injection. A chaser, of approximately 4,158 liters (1100 gallons), followed the injection of fluorescein.

Water-level changes before injection (during the slug) at RW-1 are presented in Figure 2. At the beginning of the slug, the water level rose about 4.27m (14 feet) above the initial level and then declined, as the water infiltrated, to 2.1 m (7 feet) above the initial level (about 3,780 liters or 1,000 gallons were added at this time). Thence the water level stabilized for the rest of the injection.

No significant change in water levels in monitoring wells was measured during the injection process. At wells MW-1S, MW-2, and MW-3, water levels rose slightly in the range of 0.030 m (0.10 feet), and at MW-1D, a slight decline in water level was measured (less than 0.015 m or 0.05 feet). These wells are located between 45.70 to 61.00 m (150 to 200 feet) from RW-1 (Figure 1). The water level at RW-1 was measured at 0.018 m (0.06 feet) above the pre-injection level at 13:00 hours on December 23, 1996, about 22 hours after the dye injection.

DYE TEST RESULTS

The data from a single dye trace generally reflect conditions for that particular test, and especially for that particular discharge. The shape and magnitude of dye-recovery curves are influenced, primarily, by the amount of dye injected, the velocity and magnitude of flow, the mixing characteristic within the flow system, the sampling interval, and whether the discharge was diluted by non-dyed waters (i.e., other waters from the recharge area).

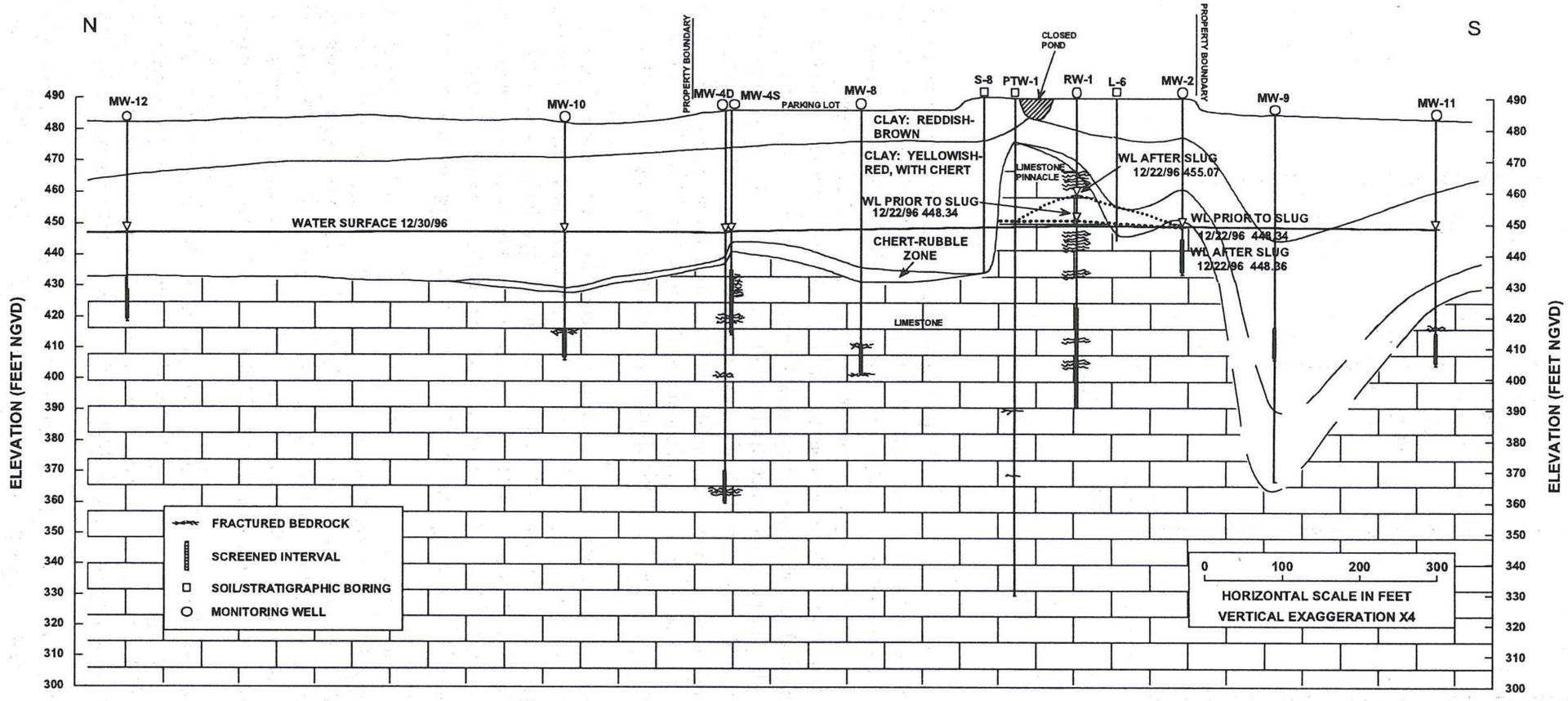

Figure 1. Local Hydrogeologic Cross-Section

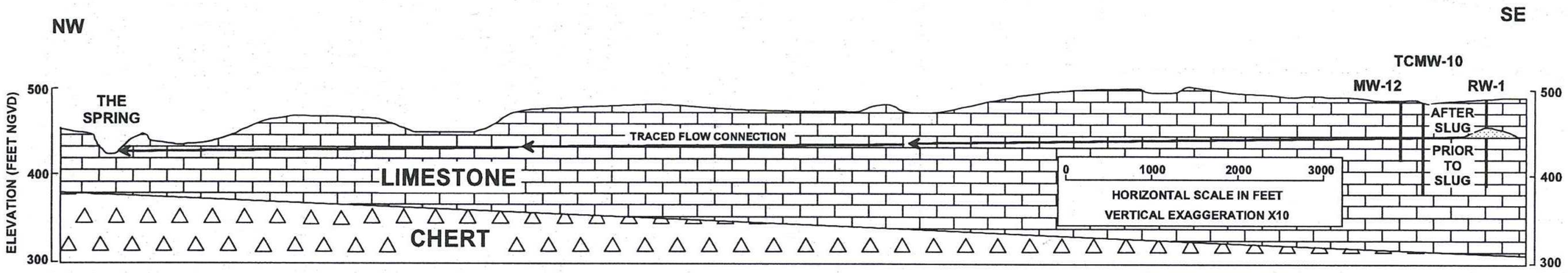

Figure 3. Regional Hydrogeologic Cross-Section

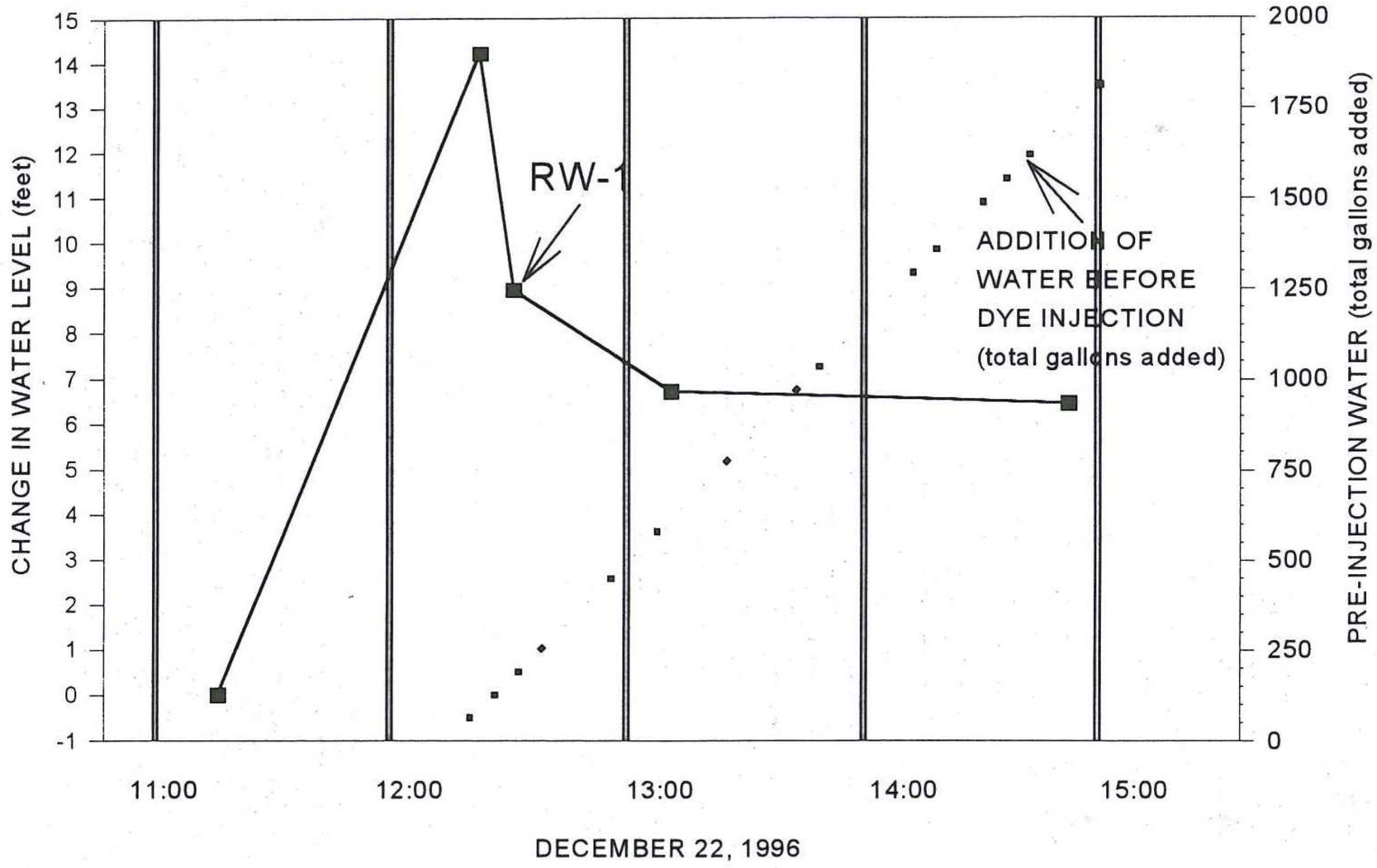

Figure 2. Water-Level change at RW-1 before the injection

Fluorescein was detected in the elutant from all four charcoal packets (SP-1, SP-2, SP-3, SB-1) at the Spring. Figure 3 is a generalized hydrogeologic cross section showing the site and the Spring and shows the pre- and post-injection water levels at the site. Time of travel (time to leading edge) was a maximum of 48 hours (maximum hours based on time charcoal packets were in place) after injection (on December 24, 1996), with the peak dye concentration being detected on December, 30, 1996. Dye detections at the springs are shown on Figure 4.

The peak dye concentration of elutant from charcoal packets from SP-1 is 16.20 parts per billion (ppb), and was detected in the packet collected 4 days (96 hours) after dye injection. At SP-1 the last charcoal packet where the dye was detected, was recovered on January 13, 1997, 528 hours after dye injection, with a concentration of 2.93 ppb. The peak dye concentration of the dye in charcoal packets from SP-2 is 104 ppb, and was detected 4 days (96 hours) after injection. The highest concentration of the dye detected in SP-3 is 233 ppb, and was encountered 4 days (96 hours) after injection. The highest concentration of the dye detected in SB-1 is 146 ppb, and was encountered 4 days (96 hours) after injection. The highest concentration of dye (Figure 4) was detected in SP-3 (maximum reading was 233 ppb) followed by SP-2 (104 ppb) and SP-1 (16.20 ppb). At SP-2, SP-3, and SB-1, the dye was detected in all the charcoal packets recovered during the 100 days period.

The results suggest that the ground water is traveling between the site and spring through a fracture system. However, because of differing concentrations detected and times of arrival of peak concentrations at the 3 springs, ground-water movement through the fracture system and the discharge at the springs is affected by conditions throughout the recharge area (218 km^2 or 84 square miles) for the Spring. The travel time (time to leading edge) of the dye from RW-1 (injection well) to the Spring is a maximum 48 hours. The dye traveled between the injection well and the spring in a maximum 48 hours, a distance of approximately 4,900 meters (16,100 feet) (flow velocity of 102 m/hour (335 feet/hour)). The difference between the water level elevation in RW-1 and spring elevation is approximately 9.14 m (30 feet), corresponding to a gradient of the water surface of about 0.1 degree, toward the spring (Figure 3).

The response of the system and the movement of the dye is a combined result of water injected at RW-1 before and after dye injection (head of water in RW-1, Figure 1 and 2), and non-pumping conditions (natural condition) at adjacent manufacturing site. The injected water resulted in an imposed artificial head at the injection well. The water and dye moved out of the injection well through the north-west trending fracture. Within less than 24 hours, the imposed head dissipated and water levels in RW-1 returned to pre-injection levels and fluctuations. The imposed or induced head resulted in an almost unique occurrence, which pushed water out to the northwest fractures, other than normal flow paths. The only other time that those northwest pathways were probably used was in 1977 when the pond failed, and a slug of water was induced into the ground-water system.

On December 29, the pumps were turned on at the adjacent manufacturing (normal conditions), and resulted in appearance of low concentrations of dye in the pumping wells. Under "normal conditions" (pumps on at the manufacturing facility, no artificially induced head in RW-1), significant concentration of dye remained in the well RW-1 indicating the well documented, low conductivity of the system. The last charcoal packet collected in RW-1 on March 31, 1997 still had a concentration of 1,430,000 ppb of dye, and the water sample had 288,000 ppb of dye (approx. 9% of the dye).

The slug flow produced by the injection procedure demonstrated connection between the two locations; but further demonstrated that the connection only functioned for a limited time in response to increasing the head at the well by 2.13 to 4.27 m (7 to 14 feet). After the water levels recovered from the injection, water was drawn to the pumping wells at the adjacent manufacturing facility. Three months after injection, significant dye remained in RW-1, and under "normal conditions" dye had not reached any on-site monitoring wells, except MW-3. This is an

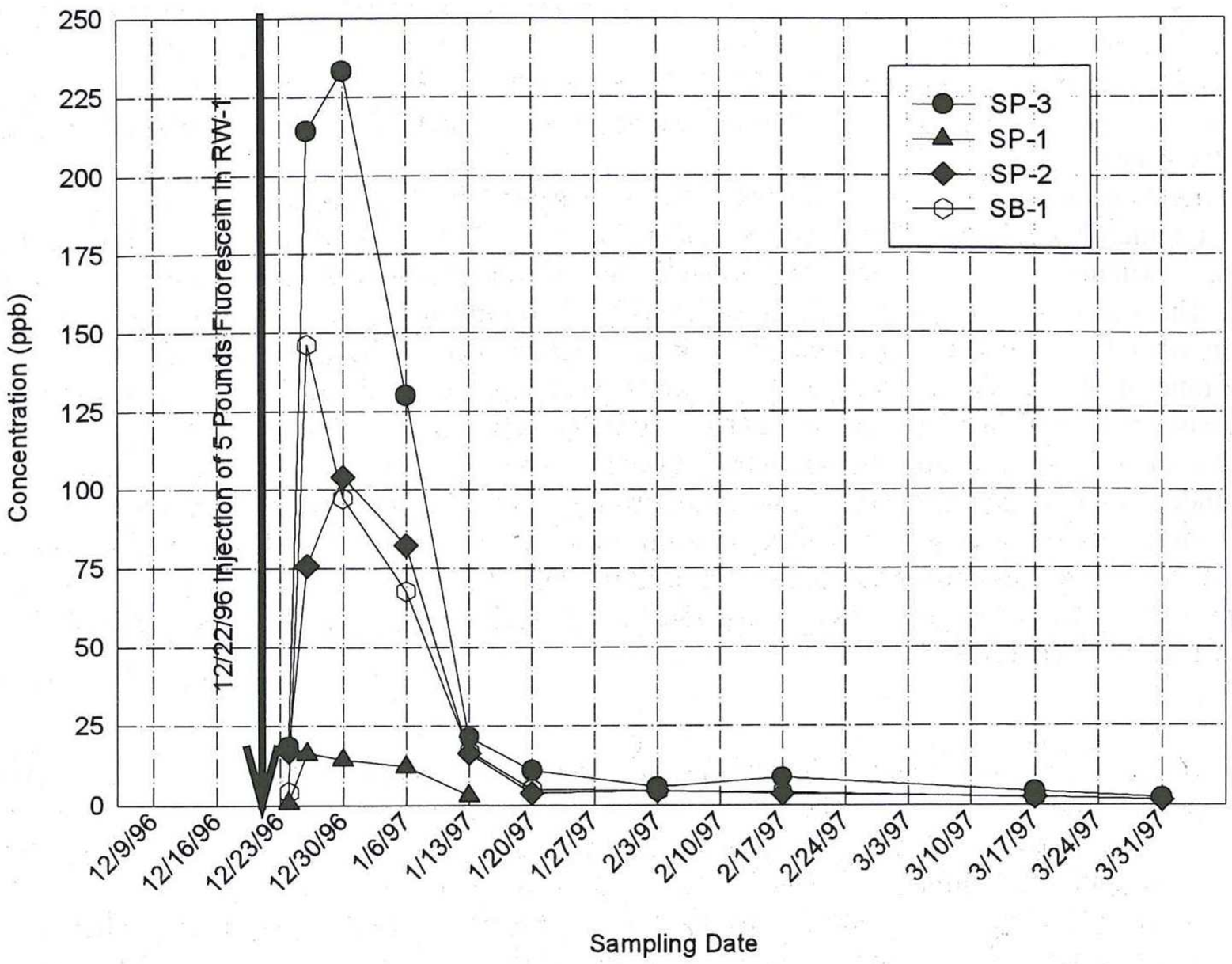

Figure 4.: Fluorescein Concentration in Charcoal packets at spring

expected result of the low gradient and slow movement of ground water under natural conditions.

At the site, the dye was detected only in MW-3 in concentrations from 1.43 ppb (March 3, 1997) to 0.53 ppb (March 17, 1997). The dye was not detected in the other wells sampled and tested. The fluorescein was not detected in other wells most probably because the other wells were not influenced by the slug induced at RW-1, as the bedrock at the site is predominantly of low hydraulic conductivity. When stressed the system exhibits a northwestern anisotropy due to the abundance of similarly oriented fractures. Under normal conditions and under conditions of limited stress (pumping at the adjacent manufacturing facility) interconnection of the northeast and northwest oriented fracture systems provides a weak diffuse flow system for the movement of ground water and movement of PCE bearing ground water.

The MW-3 well is 45.72 m (150 feet) (flow velocity based on computation, 0.027 m/hour or 0.09 feet/hour) southwest of RW-1. The dye detected in this well indicates that the well has a hydraulic connection to RW-1 and that the movement of the dye and ground water was influenced initially by the induced head at RW-1 and subsequently water withdrawal at the manufacturing facility.

At the manufacturing facility the dye was detected in four wells (two pumping wells, and two monitoring well). The pumping wells (TCMW-2 and TCMW-4) are reportedly 91.44 m (300 feet) deep. TCMW-2 is a pumping well 107 m (350 feet) west of RW-1. Water samples only were collected during the dye study. Dye was detected for the first time in the water sample collected on March 3, 1997, arrival time of the dye (time to leading edge) was 71 days after injection, or 1,032 hours (flow velocity 0.104 m/hour or 0.34 feet/hour). The peak dye concentration dye was detected on March 31, 1997, 100 days after injection, or 2,400 hours. TCMW-3 is a deep monitoring well (depth is 67 m or 220 feet below top of casing) 76.20 m or 250 feet northwest of RW-1. Charcoal packets were used in well TCMW-3 during the dye test study. Dye was detected for the first time (time to leading edge) in elutant from the charcoal packet collected on December 30, 1996, arrival time of the dye being 8 days after injection or 192 hours (flow velocity o.40 m/hour or 1.3 feet/hour). The peak concentration of the dye was encountered on February 17, 1997, 57 days after injection.

TCMW-4 is a pumping well situated 110 m (360 feet) southwest of RW-1. Water samples only were collected during the dye test study. Dye was detected for the first time in the water sample collected on January 6, 1997, arrival time (time of leading edge) of the dye being 15 days after injection, or 360 hours (flow velocity 0.3 m or 1.0 feet/hour). The peak concentration of the dye was encountered on January 20, 1997, 29 days after injection, or 696 hours, which correspond to the lowest water level elevation in TCMW-4, during the dye study. TCMW-8 is a monitoring well situated 53 m (170 feet) northwest of RW-1. Charcoal packets were used in well TCMW-8 during the dye test study. Dye was detected only in the charcoal packet collected on January 20, 1997, arrival time of the dye being 29 days after injection or 696 hours (flow velocity 0.07 m or 0.24 feet/hour).

The appearance of the dye in the wells at the adjacent facility is a result of the water withdrawal during pumping at TCMW-2 and TCMW-4. Dye was not encountered at the facility when the pumps were not operating.

At the Spring the dye was detected only in the charcoal packets. Dye was not detected in any water samples collected at the springs during the 100 day test. The passive samplers continuously absorb the tracer dye passing through them; thus they are cumulative samplers. The concentration of any dye in a sampler is, therefore, greater than the concentration in the water. Fluorescein dye is preferentially sorbed on activated charcoal when the charcoal is present in water containing fluorescein. The distribution coefficient (Kd - ratio of concentration of

fluorescein in charcoal to that in water) is not available. However, based on the results at RW-1 the concentration of dye in the elutant is 3 to 60 times greater than the concentration in water.

The concentration of fluorescein in the elutant is about 3.5 times less than that in charcoal, because 15 ml of elutant are used to extract fluorescein from 4.5 grams of charcoal. As a consequence the charcoal in packets from RW-1 should have contained 10 to 210 times more fluorescein than that in the water.

The very high concentrations of fluorescein in water from RW-1 cause saturation of the charcoal, and the apparent Kd's may be too low - but would represent a minimum for Kd. In addition, wide variations in the concentration of fluorescein in water from RW-1 indicate that fluorescein may be stratified within the well bore. If we use the data for fluorescein packets from SP-3 (Spring) we can determine a maximum value for Kd, as follows: The detection limit of fluorescein in water is 0.0005 ppb and the reporting limits is 0.0015 ppb. Using the reporting limit as the concentration, when fluorescein is not detected; and concentrations reported for the elutant from charcoal packages in SP-3 at 2.03 to 21.4 ppb range; the ratio of fluorescein in elutant to that in water has a range of about 1,350 to 14,500. Obviously the Kd for the charcoal/water exchange is 3.5 times higher than that for the elutant/water or about 4,700 to 50,000.

According to Ozark Underground Laboratory the concentration factor or Kd of dye on charcoal is time dependent and demonstrated to be several orders of magnitude after only one week and as demonstrated by the above. The system may or may not be at chemical equilibrium after one week. Under natural flowing conditions the system probably does not attain equilibrium, because the concentration of fluorescein changes continuously as a slug-flow passes through the spring. This confirms the relationships seen during the test, where elutant concentrations at the spring sites range vary from the 21 to 2 ppb and water analysis resulted in below detection. Therefore, it is likely that no dye would have been detected in water samples from the spring.

WATER LEVELS

Water-level fluctuations are related to the amount of precipitation and the water withdrawal at the adjacent manufacturing facility (the rate and duration of pumping at each well is not known). The highest ground-water levels across the site were measured between December 20, 1996, to December 29, 1996, when the pumps where off at the manufacturing facility and, on March 17, 1997, two weeks after a precipitation event, indicating the lag between the rain, infiltration, water-level response in the ground-water system.

Water-level fluctuations, over a period of 112 days (December 9, 1996 - March 31, 1997), at the site range from about 0.625 m (2 feet) at the beginning of the test, and in most of cases are in the range of less than 0.15 m (0.5 feet), illustrating that the water surface fluctuations are not dramatic at the site. The water level change across the area covered by monitoring wells is only about 0.60 m (2 feet) indicating that the ground-water surface is nearly horizontal.

Under natural conditions the ground-water gradient is nearly horizontal and ground-water velocities are low. Ground-water withdrawals at the adjacent manufacturing facility locally affect the direction of ground-water movement and gradient locally.

By contrast to the subtle conditions at the site in responding to climatological events, surface water levels and the pool at the Springs exhibit rapid responses due to precipitation and ground water recharge. The pool level rose over 1.25 m (4 feet) after the rainfall event. These conditions demonstrate the influence of precipitation over the recharge area of the springs and illustrate the contrast in hydraulic conditions between the well developed karst at the springs and the poorly connected system at the site.

VOCs CONCENTRATIONS AT THE SPRING

During the dye test, samples were collected for VOC analysis from selected monitoring sites. The PCE concentrations detected at the springs were consistently within a range from 0.28 to 0.66 ppb, at levels, 10 times below the Maximum Contaminant Level (MCL) of 5.0 ppb. The concentrations detected were fairly constant over the period of testing and could indicate a constant source from some location within the 218 km^2 (84 square mile) area of recharge. In contrast, the dye detected at the spring reflects movement of a slug or pulse through the system as a result of the imposed artificial head.

In addition, in comparing the water quality characteristics of ground water at the site and the springs, the ratio of dye concentrations in water, PCE concentrations and constituents from city water are not consistent. The dye concentration in elutant from charcoal packets changed more than 2 orders of magnitude as the dye moved through the spring area. When detected, PCE varied only 0.2 ppb in the spring discharge. Therefore, the data indicates connection between the site and the springs during slug movement, other sources of PCE, and very limited migration of dye and city water since recovery of water levels after injection.

CONCLUSIONS

Under induced artificial head, affected by the injection of water and dye, a hydraulic connection between RW-1 and the Spring was temporarily realized. A slug or pulse of dye-containing water was pushed rapidly through the system via pathways not active under normal conditions.

The water-level data collected before injection, during the injection of city water and the fluorescein dye, and during the remainder of the dye test period are consistent with and confirm monitoring data collected since initiation of investigations at the site. The regional direction of ground-water movement is to the northwest and the ground-water surface at the site is nearly flat, exhibiting a low gradient resulting in low velocity unless disturbed or influenced by outside influences (i.e., ground-water withdrawal at the adjacent manufacturing facility or slug flow).

The VOC's detected at the Spring remained fairly constant, at generally 0.6 ppb or less (well below the MCL of 5.0 ppb), over the period tested, with the exception of samples collected during the flood event (no VOC's detected). The dilution factor for dye in the elutant is over one million, and the highest concentration of PCE ever detected at site was about 1,200 ppb. Therefore, if PCE can survive the trip from the site to the Spring, the maximum concentration would have been 0.12 parts per trillion. This data in conjunction with the dye results suggests the potential for different sources for the two constituents (dye and VOC's). The PCE at the Spring cannot be directly or solely attributed to the site due to many other potential sources identified (previous dye studies) and sources unknown throughout the 84 square mile recharge area.

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