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LARGE-SCALE TRACING OF GROUND WATER WITH SULFUR HEXAFLUORIDE

By Jeff D. Gamlin,1 Jordan F. Clark,2 Greg Woodside,3 and Roy Herndon4

ABSTRACT: Sulfur hexafluoride (SF6) was injected into a 9 km reach of the Santa Ana River in Orange County, CA, over a period of two weeks. The entire flow of this river, which averaged 2.8 m3 s−1, percolated into the ground in the field area. The tracer was monitored at wells near the river to determine subsurface flow patterns and flow times with an accuracy much greater than could be achieved using numerical simulations of ground-water flow. During the experiment, SF6 effectively tagged 3.7 × 103 m3 of water. The tracer plume was mapped in the subsurface for 18 months and indicates that linear ground-water velocities averaged about 2 km year−1. The tracer reached two wells adjacent to the river (about 200 m away) within three weeks, giving evidence that SF6 was not retarded significantly relative to the ground-water flow. This is in agreement with previous laboratory experiments.

INTRODUCTION

A fundamental approach of investigating flow paths, mean residence times, and dispersivity in ground-water systems relies on geochemical tracers. Commonly used tracers in young systems include chlorofluorocarbons (CFCs), 3H/He, and 85Kr (e.g., Cook and Solomon 1997). These methods use preexisting signatures within the water to determine ground-water ages (up to 50 years) that typically have uncertainties of ±2 years. The tracer ages are used to estimate hydrologic parameters. Unfortunately, these tracers are unable to resolve ground-water ages between 0 and 2 years, important time scales in many hydrologic problems such as those associated with artificial recharge and some plume studies.

Purposeful tracers such as trace ionic substances and gases can be used to evaluate flow and transport during a period of 0–2 years (e.g., Mackay et al. 1986; LeBlanc et al. 1991). In order for such a tracer to work, it must be chemically nonreactive and cost effective. Sulfur hexafluoride (SF6) has been used as a tracer in continental waters for more than a decade (e.g., Wanninkhof et al. 1987, 1990; Clark et al. 1994, 1996). Background concentrations of SF6 are extremely low (<1 fmol/1, fmol = 10−15 mol) and it is relatively inexpensive. Thus, it is economical to use SF6 to trace water bodies that exceed 103 m3 in volume. Laboratory experiments have shown that SF6 and bromide have similar breakthrough curves (Wilson and Mackay 1993, 1996). Hence, SF6 has the potential to be an ideal ground-water tracer.

In an experiment that began in late July 1998, SF6 was injected into a 9 km section of the Santa Ana River (SAR) in Orange County, CA, for a period of 15 days. The Orange County Water District (OCWD) maintains this section as a principal recharge area to the Orange County ground-water basin. SF6 was chosen as the tracer because approximately 3.7 × 103 m3 (3,000 acre-ft) of surface water was needed to be tagged and SF6 was the most cost-effective tracer available. The purpose of this experiment was to identify flow paths and travel times from the SAR to selected wells with an accuracy better than what could be achieved using numerical simulations of ground-water flow, which rely upon average aquifer properties.

STUDY LOCATION

A 9 km section of the SAR and a series of spreading basins (Warner Basin, Anaheim Lake, Miller Basin, and Kraemer Basin) located near Anaheim, CA, are used by OCWD as a principal recharge location for the Orange County ground-water basin [Fig. 1(a)]. OCWD recharges approximately 250,000 acre-ft/year, of which about one third is recharged through the SAR (OCWD 1998).

During periods of low summer flow, the 100 m wide SAR is divided into three subchannels by constructing sand levees. These levees act in such a way that the river must travel a tortuous path that is about three times longer than its natural course. Thirteen artificial drop structures (0.4–2.8 m in total vertical height) act to decrease the natural slope of the river by dividing it into sections separated by steps. During high flow, the SAR frequently reaches the ocean. However, during low flow, the entire flow percolates into the ground within the study area.

The recharged surface water flows into an alluvial aquifer system consisting of sand, gravel, silt, and clay layers. The spatial extent of any given layer is poorly known. The recharge facilities lie near the northern edge of the basin where the ground-water levels are the highest and the aquifer is relatively thin (OCWD 1998). The piezometric surface is the highest near Imperial Dam and decreases approximately to the west (Fig. 1).

TRACER METHODS

For a period of 15 days (July 20 to August 4), 99.8% pure SF6 gas was released along the length of the SAR by bubbling through diffusing stones placed at the sediment-water interface. Each injection device, which was housed in a weighted, watertight, PVC tripod container, consisted of a 450 ml lecture bottle, an in-line regulator, and a needle valve set so that the release rate was approximately 20 cc min−1. Three of these devices were deployed just under the surface of the water (to keep them near a constant temperature), along the bank of the river at locations below alternating large (2.8 m) drop structures (Fig. 1). Every other day, the lecture bottle in each tripod was replaced.

Samples were collected daily along the length of the river at designated locations to determine the tracer input function to the ground-water aquifer. At each location, three or four samples were collected across the channel. All river samples were stored submerged and analyzed on a gas chromatograph (GC) within four hours of collection at a temporary lab set up just downstream of the sampling locations.
at OCWD field headquarters, using the head space method described by Wanninkhof et al. (1987) and Clark et al. (1994). The GC detector response was calibrated about every 30 minutes with standards (148 pptv and 1947 pptv) certified by Scott-Marrin Inc. The precision and detection limits of this method were ±3% and 0.04 pmol l⁻¹, respectively.

OCWD staff collected ground-water samples either biweekly or monthly, based on a predetermined sampling strategy, from three public supply wells and 20 monitoring wells, of which eight were screened at multiple discrete depths (Fig. 1). SF₆ samples were collected in 3/8 in. copper tubes (10 ml) that were sealed with steel pinch-off clamps and sent to the University of California, Santa Barbara, where they were analyzed. Tests have shown that SF₆ samples can be stored in copper tubes for at least three months.

In the laboratory, each copper tube was attached to a low vacuum extraction apparatus, where the water sample was transferred into a 25 ml glass bulb. The sample was stirred vigorously for 5 min to extract the SF₆ from the water. A known quantity of ultra-high purity N₂ gas was then added to the system. 20 ml of the head space gas (about half of the gas) was drawn into a glass syringe and analyzed with the GC. The uncertainty determined with replicates was ±15% and is probably related to poor head space mixing.

FIG. 1. Map of Field Area Showing: (a) Water Table Elevation, Santa Ana River Recharge Area, and Off-River Recharge Ponds; (b) Well Locations and SF₆ Plume with Contours of Peak Concentration Arrival Time (Water Table Elevation Determine Using Only Well Data)

EXPERIMENTAL RESULTS

SAR Tracer Distribution

Prior to the start of the tracer injection, background samples for SF₆ were collected along the SAR and were found to be below the detection limit (~0.04 pmol l⁻¹). This was also the case for all samples collected upstream of the first injector during the 15 day injection period and subsequent samples collected thereafter.

Based on inferred concentrations immediately downstream of the injectors, it was estimated that approximately 4% of the SF₆ released dissolved in the river; the rest escaped to the atmosphere. Mean cross-sectional concentrations at each sampling location varied by about 30% during the fifteen day injection. There was considerable spatial variation found along the axis of the river related to the location of the injectors and drop structures leading to a complex input function to the ground-water system. Mean cross-sectional SF₆ concentrations ranged from ~20 pmol l⁻¹ to ~300 pmol l⁻¹ (Fig. 2). Axial distributions showed that tracer concentrations decreased with travel distance from the injectors, with the sharpest decreases at the large drop structures (Fig. 2). The river typically became well mixed across the channel (standard deviation <5% of mean) after traveling about 2.5 km in the subchannels.

Gas Exchange Rates

The main loss of tracer downstream from the injectors occurred at the thirteen hydraulic drop structures, which ranged in height between 0.4 and 2.8 m. The gas exchange efficiency (E) for these structures was calculated using the following equation (Rindels and Gulliver 1991):

\[ E = \frac{(C_{up} - C_{dn})(C_{up} - (C_g/H))}{(C_{up} - C_{dn})} \]  

where \( C_{up} \) = concentration upstream; \( C_{dn} \) = concentration downstream; \( C_g \) = concentration in the air; and \( H \) = Henry coefficient. In this case \((C_g/H)\) is approximately equal to 0.

The 2.8 m drop structures experienced a mean efficiency of 47 ± 4%, while the 0.4 m drop structures had a mean efficiency of 5 ± 3%. Loss of the tracer also occurred along lengths of the river that did not contain drop structures. Concentrations decreased by about 20% per kilometer of flow in the subchannels.


**TABLE 1. Tracer Arrival Times and Calculated Velocities at Selected Wells**

<table>
<thead>
<tr>
<th>Well</th>
<th>Screen depth(^1) (m msl)</th>
<th>Distance direct(^a) (km)</th>
<th>Flow path(^b) (km)</th>
<th>First arrival(^c) (weeks)</th>
<th>Peak arrival(^d) (weeks)</th>
<th>Duration(^e) (weeks)</th>
<th>Mean velocity(^f) (km/year)</th>
</tr>
</thead>
<tbody>
<tr>
<td>WBS-4 #1</td>
<td>52</td>
<td>0.24</td>
<td>0.30</td>
<td>1</td>
<td>3</td>
<td>10</td>
<td>5.2</td>
</tr>
<tr>
<td>SAR-8 #1</td>
<td>58</td>
<td>0.14</td>
<td>0.20</td>
<td>3</td>
<td>3</td>
<td>34</td>
<td>3.5</td>
</tr>
<tr>
<td>SAR-7 #1</td>
<td>38</td>
<td>0.33</td>
<td>0.60</td>
<td>7</td>
<td>15</td>
<td>8</td>
<td>2.1</td>
</tr>
<tr>
<td>AM-4</td>
<td>11</td>
<td>0.89</td>
<td>1.00</td>
<td>&lt;17</td>
<td>17(?)</td>
<td>&gt;24</td>
<td>&gt;3.1</td>
</tr>
<tr>
<td>OCWD-LV1</td>
<td>36</td>
<td>0.23</td>
<td>0.55</td>
<td>11</td>
<td>17</td>
<td>&gt;67</td>
<td>1.7</td>
</tr>
<tr>
<td>SAR-1 #1</td>
<td>14</td>
<td>0.16</td>
<td>0.21</td>
<td>13</td>
<td>17</td>
<td>16</td>
<td>0.6</td>
</tr>
<tr>
<td>WBS-2A #2</td>
<td>47</td>
<td>0.72</td>
<td>0.77</td>
<td>13</td>
<td>19</td>
<td>44</td>
<td>2.1</td>
</tr>
<tr>
<td>WBS-3 #1</td>
<td>54</td>
<td>0.45</td>
<td>0.70</td>
<td>17</td>
<td>21</td>
<td>36</td>
<td>1.7</td>
</tr>
<tr>
<td>OCWD-FH1</td>
<td>36</td>
<td>0.15</td>
<td>0.20</td>
<td>17</td>
<td>31</td>
<td>36</td>
<td>0.3</td>
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<tr>
<td>AM-5A</td>
<td>19</td>
<td>1.24</td>
<td>1.80</td>
<td>25</td>
<td>33</td>
<td>28</td>
<td>2.8</td>
</tr>
<tr>
<td>WBS-2A #3</td>
<td>33</td>
<td>0.72</td>
<td>0.90</td>
<td>27</td>
<td>34</td>
<td>38</td>
<td>1.4</td>
</tr>
<tr>
<td>WBS-4 #3</td>
<td>8</td>
<td>0.24</td>
<td>0.30</td>
<td>17</td>
<td>35</td>
<td>32</td>
<td>0.4</td>
</tr>
<tr>
<td>SAR-6 #1</td>
<td>5</td>
<td>0.23</td>
<td>0.98</td>
<td>33</td>
<td>39</td>
<td>&gt;74</td>
<td>0.9</td>
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<tr>
<td>YLWD-15</td>
<td>28</td>
<td>0.49</td>
<td>0.65</td>
<td>20</td>
<td>39</td>
<td>&gt;74</td>
<td>0.9</td>
</tr>
<tr>
<td>AM-5</td>
<td>13</td>
<td>1.24</td>
<td>1.85</td>
<td>29</td>
<td>39</td>
<td>66</td>
<td>2.5</td>
</tr>
<tr>
<td>SAR-2 #1</td>
<td>13</td>
<td>0.23</td>
<td>0.67</td>
<td>13</td>
<td>53</td>
<td>48</td>
<td>0.7</td>
</tr>
<tr>
<td>YLWD-5</td>
<td>11</td>
<td>1.27</td>
<td>1.69</td>
<td>29</td>
<td>53</td>
<td>24</td>
<td>1.7</td>
</tr>
<tr>
<td>WBS-3 #2</td>
<td>11</td>
<td>0.45</td>
<td>0.70</td>
<td>53</td>
<td>70</td>
<td>&gt;48</td>
<td>0.5</td>
</tr>
<tr>
<td>AM-11</td>
<td>−4</td>
<td>0.66</td>
<td>0.95</td>
<td>—</td>
<td>—</td>
<td>&gt;38</td>
<td>—</td>
</tr>
</tbody>
</table>

\(^a\)Total screen lengths range between 3 m and 5 m except for the YLWD wells.
\(^b\)The direct distance is the shortest (perpendicular) distance to the river while the flow path distance was inferred from the migration of the plume and the water table elevations.
\(^c\)The velocity was determined from the peak arrival time and the flow path distance. It is the mean linear velocity rather than the Darcy velocity.

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**Ground-Water Tracer Data**

\(\text{SF}_6\) concentrations in ground-water samples collected prior to and within three weeks of the start of the tracer injection were below the detection limit. The tracer first arrived at two shallow wells, SAR-8 #1 and WBS-4 #1, adjacent to the river (about 200 m) within three weeks [Fig. 1(a); Table 1]. The tracer plume remained at both wells for about six weeks. Maximum \(\text{SF}_6\) concentrations were, respectively, 104 pmol l\(^{-1}\) and 10.6 pmol l\(^{-1}\) (Fig. 3). The large difference in the maximum concentration is probably related to the location of the wells relative to the injectors and drop structures. SAR-8 is located just downstream of Injector 2, while WBS-4 is located upstream of Injector 2 and far from Injector 1.

Eleven weeks after the start of the tracer injection, \(\text{SF}_6\) arrived at two more wells, OCWD-LV1 and WBS-2A #2 (Table 1). Thereafter, the tracer was observed at a sufficient number of wells to map its migration through the ground-water system. At 30 weeks, two plumes could be mapped that extended to the north approximately perpendicular to the SAR up- and down-gradient from Warner Basin [Fig. 1(b)]. Warner Basin was recharging about 0.8 m\(^3\) s\(^{-1}\) of surface water to the ground-water system at this time, thus creating a water mound that diverted the ground-water flow. By 60 weeks, these two plumes merged and began to flow to the west. Ground-water flow velocities can be determined using both the first and peak arrival times at each well and the inferred flow path. Linear velocities calculated with the peak arrival times varied by an order of magnitude (0.6 km year\(^{-1}\) to 7 km year\(^{-1}\)) and averaged 2 km year\(^{-1}\). The first arrival times, which indicate the quickest flowpaths from the SAR to the wells, are important when considering the transport of “time-sensitive” contaminants such as microbes.

The migration of the tracer was better resolved for the upper two-thirds of the river than the lower third because of the higher density of wells. However, the arrival time at a few wells (AM-45, SAR-1, and SAR-2) suggests that the tracer was moving at a horizontal rate approximately five times slower. The slower ground-water velocities probably reflects both differences in the hydrogeology (i.e., hydraulic conductivity) and deeper screened intervals.

**DISCUSSION AND SUMMARY**

The tracer injection tagged approximately \(3.7 \times 10^6\) m\(^3\) of surface water with \(\text{SF}_6\) concentrations that ranged between \(\sim 20\) pmol l\(^{-1}\) and \(\sim 300\) pmol l\(^{-1}\). The variation in concentration along the axis of the SAR followed a predictable pattern. Concentrations were the highest just below injectors and decreased nearly exponentially with distance as the gas was lost across the air-water interface. Gas-exchange efficiencies over the 2.8 m drop structures averaged about 50% and were in good agreement with other estimates of the gas exchange efficiencies over hydraulic drops (Rindels and Gulliver 1991).

As a result of gas loss, the spatial input function of the tracer...
into the ground-water system was very complex and intensive sampling was required to define this function.

There was surprisingly little temporal variation in the mean cross-section concentration at each sampling location considering the simple design of the injectors. After setting the regulator and needle valve, the flow remained nearly constant. Much of the temporal variability in concentration was caused by replacing the lecture bottles every other day. Injectors designed to accommodate larger gas cylinders would eliminate this problem. Another source of variability was caused by the occasional movement of the diffusion stones, because the tracer injection rate was very sensitive to water depth.

After percolating into the subsurface, the tracer was transported by the ground-water flow system, creating plumes that could be easily mapped. Linear ground-water velocities averaged 2 km year$^{-1}$. The plume reached two wells, WBS-4 and SAR-8, three weeks after the start of the injection. The distance between the SAR and the screens of these two wells is on the order of 200 m. Assuming the tracer traveled the shortest possible distance, a minimum ground-water velocity of about 5 m day$^{-1}$ was calculated. Given this rapid transport rate and estimates of the hydraulic gradient and conductivity, SF$_6$ does not appear to be retarded relative to the ground-water flow in this aquifer system, which is in agreement with previous laboratory experiments of Wilson and Mackay (1993, 1996). At many wells, tracer was observed for more than 20 weeks, showing the effects of dispersion and probably the effects of the flow system, which tended to focus the flow initially into two plumes around Warner Basin and later in a narrow zone between the SAR and Anaheim Lake.

Due to the low cost and fast analysis time of samples, SF$_6$ gas proved to be an effective tracer for tagging water in a river and following its movement in the aquifer after recharge. The authors expect to be able to follow the movement of the SF$_6$ patch for at least another 12 months. Ultimately, this data will be used to refine numerical models of ground-water dynamics near the SAR.

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APPENDIX. REFERENCES


