Monitoring to Improve the Understanding of Herbicide Fate and Transport in the Southern Sierra Nevada, California

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Reforestation efforts on the Sierra and Stanislaus National Forests (California) in the 1990s included use of the herbicide, hexazinone. This herbicide is the most water-soluble and water-mobile of those approved for use under a Regional EIS (Environmental Impact Statement) for vegetation management. To better understand hexazinone movement in soil and water typical of areas being reforested, monitoring at operational units was carried out. Two monitoring efforts are described here. In the first, instrumentation to detect hexazinone in the soil, vadose zone water, and surface water runoff was used to monitor the effectiveness of a stream buffer zone. Initial chemical application was to a 6.1 by 7.6 m test plot upslope from an instrumented area. Water and soil samples were collected after five significant storm events. The following year, an operational chemical application was made to the unit containing this instrumentation. Samples were collected after four significant storm events. Hexazinone was detected in the vadose zone to a depth of 1.8 meters and for a slope distance of 6.1 meters. The difference between the test plot and operational applications was an order of magnitude greater concentration detected in both vadose water and surface runoff. While hexazinone did penetrate into the buffer zone, it was well below concentrations of concern. In the second monitoring effort, on the Stanislaus National Forest, groundwater monitoring wells were installed in selected reforestation units to verify levels of hexazinone entering groundwater and examine its persistence. A total of five monitoring wells were sampled. Wells sampled the unconfined upper aquifer above competent bedrock and typically varied between 2.9 m to 10.1 m in depth. If hexazinone was detected, typically 1 to 2 years had passed between hexazinone application and detection in groundwater samples. While the concentrations generally dropped after initial detection, it took between 2 to 3 more years before it fell below the detection limit. Monitoring demonstrated that hexazinone detected in groundwater was well below the concentration established by the State of California as a water quality goal.

Keywords: *herbicides, monitoring, water quality, groundwater, surface water, soil*

Introduction

The Forest Service Pacific Southwest Region (R-5) (Figure 1) published an Environmental Impact Statement (EIS) in 1974 to address reforestation methods (USDA Forest Service 1988). In the years following, public concern regarding use of herbicides and possible effects of vegetation management on the forest environment changed, as did environmental regulations and the herbicides registered for silvicultural use (USDA Forest Service 1988). A moratorium on herbicide use in the Pacific Southwest Region was instituted in 1984 in response to federal court rulings.

In response to these changes, a final Environmental Impact Statement (FEIS) for vegetation management for reforestation was issued in December 1988 (USDA Forest Service 1988). The Record of Decision (ROD) issued in February 1989 selected an alternative that provided for the limited use of herbicides in reforestation efforts (USDA Forest Service 1989). The ROD reiterated several mitigation measures for herbicide use from Table 2-7 of the FEIS. One of these measures stated, "Do not use hexazinone, picloram, dalapon, or dicamba where they are expected to enter groundwater or surface water, such as when soils are very sandy or have low clay or organic matter contents." A clarifying letter to Forest Supervisors on 30 October 1990 from the Regional Forester noted that this mitigation measure did not mean a zero tolerance

M Furniss, C Clifton, and K Ronnenberg, eds., 2007. *Advancing the Fundamental Sciences: Proceedings of the Forest Service National Earth Sciences Conference, San Diego, CA, 18-22 October 2004*, PNW-GTR-689, Portland, OR: U.S. Department of Agriculture, Forest Service, Pacific Northwest Research Station.

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Figure 1. The Pacific Southwest Region showing the location of National Forests relative to geographic features in California. Note the Sierra and Stanislaus National Forests in the southern Sierra Nevada.

for the stated herbicides entering groundwater or surface water. Rather, a site specific analysis should identify high risk situations where this mitigation would be applied. Another mitigation measure in Table 2-7 of the FEIS stated, "Monitor herbicide residues in soil and ground and surface water to identify patterns of herbicide persistence and mobility at sensitive sites." This direction clearly required monitoring to validate site specific analyses and decisions on hexazinone use.

Of the four chemicals noted in the mitigation measure, only hexazinone was considered appropriate to the treatment prescriptions. Neary et al. (1983) noted hexazinone has high water solubility (33,000 ppm by weight at 25°C), making it likely to move off-site with storm runoff and leach to groundwater. Therefore, it became the focus of concern for much of the groundwater and surface water monitoring on reforestation projects in the southern Sierra Nevada. Earth scientists working on national forests in the southern Sierra Nevada were faced with predicting the pattern for mobility and persistence of hexazinone as input to environmental documents prepared for reforestation projects. They also needed to define locations where hexazinone should not be used in vegetation management prescriptions.

Unit 505 Monitoring – Sierra National Forest

Test Plot Applications

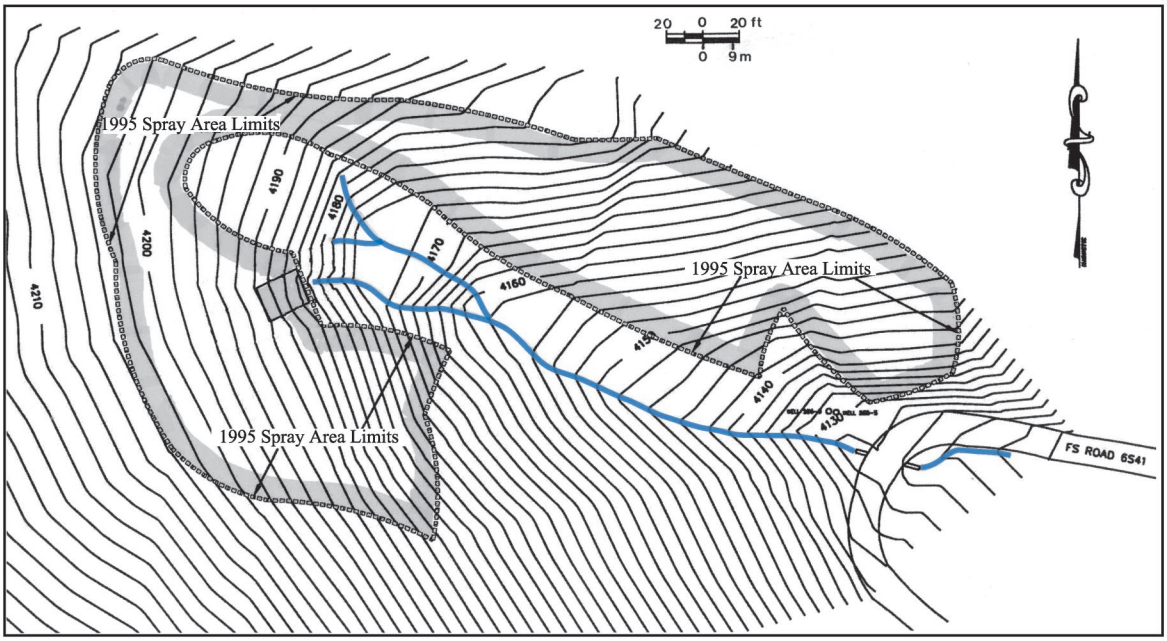
The Sierra National Forest undertook a number of small reforestation projects in the early 1990s. The mixed conifer forests in these areas were generally second-growth, following the cutting of the old-growth during the early 1900s. Harvest of this second-growth forest in the 1980s had occurred partly in response to the combined effect of drought and insect attack. Unlike earlier logging efforts, the recent harvest had included clear-cut units. In some of these harvest units, grass and brush competition significantly hampered re-establishment of conifers in subsequent reforestation efforts. Mechanical methods were not wholly successful in reducing this competition. Herbicide application was then undertaken to improve the success of reforestation efforts.

Buffer zones are a common mitigation to prevent impacts to water quality. In 1993, a question arose about the adequacy of buffer zones applied to Class 4 channels. Class 4 channels are the minor drainage ways that typically channel water into more defined channels only during snowmelt runoff or during intense rain storms. A buffer 7.6 meters on either side of the Class 4 channel centerline was excluded from application of herbicides for individual reforestation projects on the Sierra National Forest. The widespread distribution of Class 4 channels would lead to serious problems in operations to apply herbicides if the buffer zones needed to be wider than 7.6 meters.

A site was chosen in 1993 for an intensive monitoring effort to examine the adequacy of the 7.6-meter buffers on Class 4 channels. This monitoring effort clearly represented implementation of the mitigation measure, "Monitor herbicide residues in soil and ground and surface water to identify patterns of herbicide persistence and mobility at sensitive sites." While scientific literature on hexazinone persistence and mobility was available for the southeastern United States, no studies had been done for the southern Sierra Nevada, and it was questionable whether results from such a different and distant region could be applied to central California conditions.

The chosen site, Unit 505, was one of several herbicide treatment units in an approved vegetation management plan for part of the Bass Lake Ranger District of the Sierra National Forest. Unit 505 lies within the Lewis Creek watershed about 5 km north of the town of Oakhurst, California. The vegetation, slope, and soil present in Unit 505 are representative of many areas where hexazinone was being considered as a treatment option. The elevation is 1340 m, on generally east-facing slopes. Soil mapped as Holland Series overlies the Tonalite of Bass Lake at this

Figure 2. A detailed topographic map showing the outline of the Unit 505 operational area (wide gray shaded border). The alignment of the Class 4 channel is shown in blue. The larger rectangle is the outline of the protective fence with the inner rectangle delineating the application area and instrumented area within the buffer zone.



site. At the beginning of the study, the ground surface was covered by grasses with scattered concentrations of bear clover.

The monitoring plan specified establishment of a 6.1 m by 7.6 m plot (46.5 m^2) for applying hexazinone, adjacent to the upper boundary of the buffer zone of a Class 4 channel (DeGraff et al. 1994) (Figure 2). Transect lines for soil samplings were established within the application area and the adjacent buffer zone in order to determine the concentrations of hexazinone at the ground surface where the herbicide was applied, and where herbicide attached to soil particles might be washed down slope. Leaching to shallow groundwater was evaluated using pressure vacuum lysimeters, consisting of PVC tubes with a ceramic tip, installed on three lines parallel to the slope contour. The first line was at the application plot/buffer boundary, while the second and third lines were placed at 3.1 m and 6.1 m down slope from the boundary, respectively. Along each line, three sets of 0.6-m- and 1.2-m-long lysimeters were installed, with a set in the center of the line and the others 1.5 m to either side of the center set (Figure 3). The exception to this array was along the line placed 6.1 m down slope, where a 1.8-m-long lysimeter was added to this arrangement. Vadose zone water was collected from the lysimeters by placing a vacuum on each one following each storm event. Fluid was extracted two to five days later into sample bottles.

Surface runoff was intercepted at a 4.6-m distance down slope from the application plot/buffer boundary where a stiff plastic barrier was embedded into the soil. The barrier was attached to a pipe that directed the collected surface water into a container housing a 1000-mL sample bottle. After each storm event, the filled or partially filled sample bottle was collected for analysis.

On 15 March 1994, liquid hexazinone was applied to the plot using a backpack sprayer (Figure 4), at the treatment rate of 1.4 kg of active ingredient per 0.4 ha prescribed for the operational treatment of Unit 505.

Figure 3. A view down slope from the application area toward the Class 4 channel that runs from left to right across the background. The clusters of lysimeters are visible on the boundary of the application area and buffer zone and along the line 3.1 m down slope. A black plastic barrier at 4.6 m diverts surface runoff to the surface water sampler.

Figure 4. A view of the monitoring installation in Unit 505 surrounded by a protective fence. The application area is visible in brown following the initial hexazinone treatment.

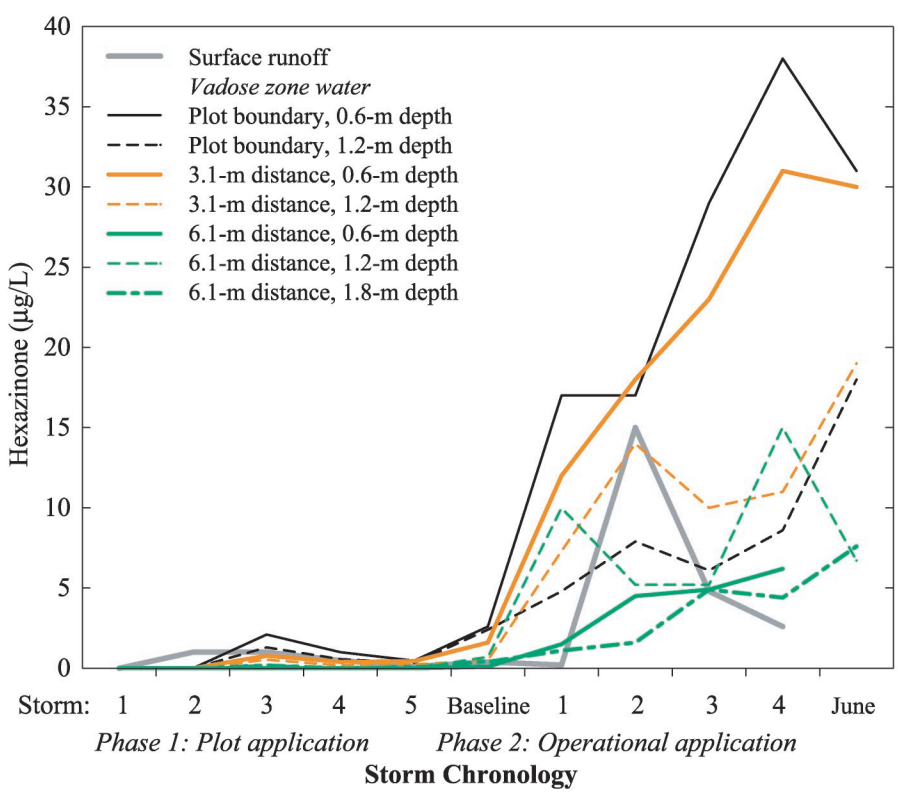
No hexazinone applications had previously occurred in this unit or anywhere else within the watershed where this unit is located. The absence of hexazinone residues was confirmed by sampling of soil transects in the application area and by testing soil excavated during lysimeter installation prior to application.

Five significant precipitation events affecting the application area took place between 15 March 1994 and 31 May 1994. A total of 185.4 mm of precipitation was recorded between these dates at the daily storage gauge located at Batterson Work Center, about 4 km from the site and 1305 m lower in elevation. Daily precipitation totals ranged from 27.9 mm to 2.5 mm, with the events on 24 March, 8 April, and 19 May being generally light rainstorms. Some of these events were preceded by one to two days of lower precipitation. A rainstorm on 23 April continued through 25 April and turned to snow with falling temperatures. The fifth storm event was a thunderstorm on 30 May yielding some intense rainfall.

No hexazinone was detected in surface water after the first storm event on 24 March (Figure 5). The highest concentration in surface water of 1.0 µg/L was found after the second storm on 8 April. The 23 April sample was lost due to breakage during transport. However, the remaining two storm events yielded surface water hexazinone concentrations of 0.49 µg/L on 19 May and 0.11 µg/L on 30 May. It should be noted that the surface water sample for the 19 May storm event was taken earlier from preceding rainfall. The change to snowfall on 19 May resulted in no surface runoff at that time.

No hexazinone was detected in vadose zone water withdrawn from the lysimeters after the first storm (24 March) or the second storm (8 April) (Figure 5). After the third storm (23 April), hexazinone was detected in the 0.6-m lysimeters at the application plot/buffer boundary and in the line 3.1-m down slope, but not at 6.1 m down slope, where the 0.6-m lysimeters are down slope from the surface water intercepting barrier. It was also detected in the 1.2-m lysimeters on the plot boundary, and at the lines

Figure 5. Chart comparing concentrations of hexazinone found in the surface runoff and vadose zone water following plot and operational applications.



3.1-m and 6.1 m down slope. Hexazinone was detected in the 1.8-m lysimeters installed only along the line 6.1 m down slope. As expected for a material leaching into the soil, the concentrations detected at any depth were highest for the points closest to the application area and decreased down slope. Similarly, the concentrations at any particular line of lysimeters decreased with depth. With subsequent storms, this pattern of concentration values persisted but with decreasing concentrations. The highest concentration detected in vadose water was 2.1 µg/L.

Surface soil was collected from both the application area and the buffer zone. Within the application area, soil samples were collected at the beginning of the months of April, May, June and September. Samples were soil plugs 7.6 cm deep, collected along two transects and combined for analysis. The highest 1994 concentrations found in the application area were 910 µg/L (April), 880 µg/L (May), 330 µg/L (June), and 40 µg/L (September). Surface soil sampling within the buffer zone took place along transects aligned at 1.5 m and 3.1m down slope from the application plot/buffer zone boundary. These samples were 2.6 cm-deep plugs of soil collected after the first, third, and sixth storm events. Buffer zone sampling attempted to determine how much hexazinone was transported down slope via attachment to surface soil particles. Hexazinone was detected at 15 µg/L (1.5-m transect) and 13 µg/L (3.1-m transect) in the surface soil of the application area after the third storm. It was not detected in samples taken after the first or sixth storm events. Hexazinone may have been present at concentrations less than 10 µg/L, as the extraction process from soil resulted in this higher detection level than analysis of water samples.

The plot results showed that hexazinone would penetrate at least 4.5 m into the 7.6-m buffer zone in surface water, at least 3.1 m in surface soil, and leach to a depth of 1.8 m at 6.1m into the buffer zone. This demonstrated that a larger quantity of the chemical was moving into the buffer zone than anticipated. Sampling also found higher concentrations of hexazinone in vadose zone water than in surface runoff. However, no concentration detected exceeded the then-current 200 µg/L regulatory limit for hexazinone. This 200 µg/L limit was the State of California adopted water quality goal, and represents the Environmental Protection Agency's lifetime health advisory value (EPA 1988), which represents the portion of an individual's total hexazinone exposure from drinking water, and is meant to limit lifetime adverse health effects due to exposure to carcinogenic substances. The highest detected value within the buffer zone, the 15 µg/L surface soil value, is clearly significantly lower than the State of California regulatory limit for this chemical.

Operational Application

To ensure that plot results represented actual operational results, a second phase of monitoring involved the planned treatment of Unit 505 with hexazinone (Figure 4). The same instrumentation was left in place to monitor surface soil, surface runoff , and leaching into the soil. Treatment of the unit took place on 15-16 March 1995. Again, backpack sprayers were used to apply the hexazinone at the rate of 1.4 kg of active ingredient per 0.4 ha.

Prior to the first storm following treatment, the samples were collected of surface water runoff, soil from transects within the buffer zone, and vadose zone water from the lysimeters. Nearly one year after the plot treatment, surface water runoff was found to have concentrations of hexazinone at 0.9 µg/L, at non-detectable concentrations in surface soil, and at maximum concentration of 2.6 µg/L in vadose zone water. This represented the baseline situation prior to the operational application in March 1995.

Four significant storm events occurred following the operational application of hexazinone to Unit 505, between 20 March and 15 May 2004. As measured at the storage gauge at Batterson Work Center, these storms generated a total of 160 mm of precipitation. Daily totals on 22 March, 17 April, 5 May, and 15 May ranged from 16.5 mm to 30 mm. Rainfall the day preceding the storm peak day ranged from 0 to 17.3 mm.

Hexazinone detected in surface water during the 22 March storm event was at 0.2 µg/L (Figure 5). During the second storm (14 April), the maximum concentration of 15.0 µg/L was found in the surface water sample. The third storm sample (5 May) and fourth storm sample (15 May) were found to have 4.8 and 2.6 µg/L of hexazinone present, respectively.

Hexazinone concentrations in vadose zone water jumped from the maximum pre-application value of 2.6 µg/L to 17 µg/L after the first storm event (Figure 5). It was 18 µg/L after the second storm event. Concentrations increased to 29 and 38 µg/L following the third and fourth storm events, respectively. All the samples extracted from the lysimeters, regardless of depth or position on the slope, were found to have concentrations of hexazinone greater than the those found during the baseline analysis before the first storm event. The distribution of concentrations among the lysimeters at each sampling event was similar to that encountered following the plot application; the values detected at any depth were highest for the points closest to the application area and decreased down slope. Likewise, the concentrations at any line were greatest at the shallowest depth and decreased with depth. The difference between the plot application and the

operational application was that while this pattern of concentration values persisted, it was with increasing rather than decreasing concentrations. A general decrease in concentrations of hexazinone was not detected until post-rainy season sampling in June 1995 when the highest concentration had dropped to only 31 µg/L.

Again, transects aligned at 1.5 m and 3.1 m down slope from the application plot/buffer zone boundary were used for surface soil sampling. Samples were collected in the upper 2.5 cm of soil after the first and third storm events, and in follow-up monitoring in June 1995. Unlike sampling after the first storm following plot application, hexazinone was detected in samples taken after the first storm event. The maximum concentration along the 1.5-m transect was 1200 µg/L and along the 3.1-m transect at 20 µg/L. Sampling after the third storm detected hexazinone at 20 µg/L (1.5-m transect) and 30 µg/L (3.1-m transect) in the surface soil of the application area. This dropped to 10 µg/L along both transects in the follow-up sampling done in June 1995.

The concentrations of hexazinone detected after operational application were significantly higher in surface and vadose zone water and in surface soil compared to the plot application. However, the patterns of change in concentrations found over time and space were similar (Figure 5). Hexazinone concentration was highest in the surface water from the second storm event following each application. In the vadose zone, the peak concentrations lagged one to two storms behind the surface water peak, and the pattern of changing concentrations down slope and at depth was similar. Again, the maximum concentration of hexazinone in surface water was about half the maximum concentration detected in vadose zone water. However, the concentrations of hexazinone for both plot and operational applications were below the water quality goal of 200 µg/L, except for one surface soil sample at the 1.5-m transect taken after the first storm event following operational application. Higher concentrations persisted longer in the vadose zone after the operational application than after the plot application.

The Unit 505 monitoring clearly showed that hexazinone in the surface water runoff and on surface soil penetrates a significant distance into the buffer zone for Class 4 channels. It also demonstrates that leaching of hexazinone in the vadose zone water moves laterally down slopes into the buffer zone and vertically to at least a 1.8-m depth. However, it also shows that the concentrations are unlikely to approach or exceed the State of California water quality goal for hexazinone, except for surface soil washed about a meter into the buffer zone during the first storm following operational application.

Post-wildfire Reforestation Monitoring – Stanislaus National Forest

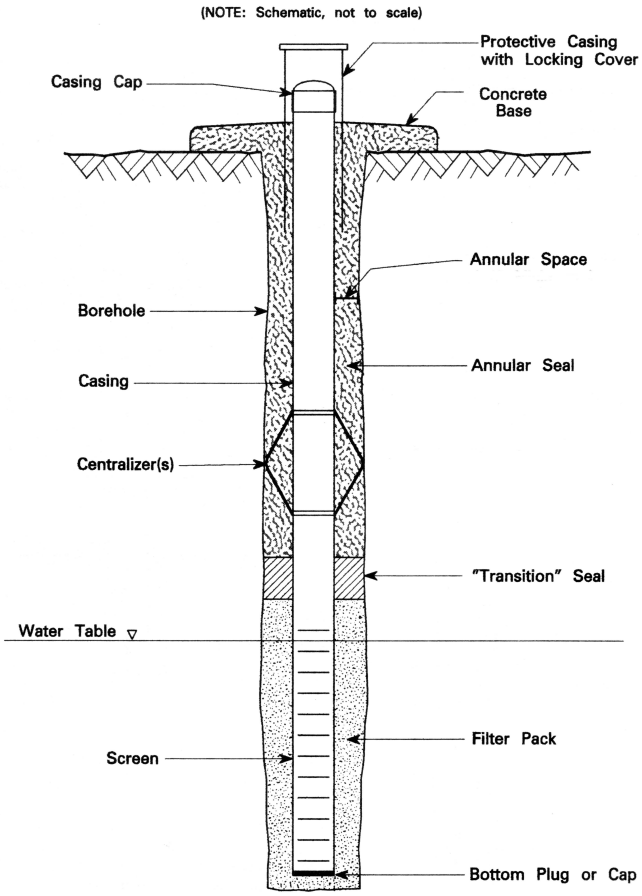
In August 1987, several major wildfires were ignited by dry lightning on the Stanislaus National Forest. These wildfires expanded into each other to create the Stanislaus Complex wildfire that eventually burned 59,489 ha on the Mi-Wok and Groveland Ranger Districts (Frazier and Grant 2003). Reforestation efforts experienced significant problems in grass and brush competition and resulted in plans to use herbicides to improve seedling survival and growth. Public opposition delayed implementation of this work until the late 1990s. Several additional wildfires adjacent to the Stanislaus Complex, such as the Arch Rock and Ruby fires, added more areas in need of reforestation during this time.

Hexazinone was one of the herbicides planned for use in this reforestation effort. Development of the alternatives excluded use of hexazinone on any treatment unit where a reasonable chance existed for it to contaminate existing wells. In order to validate the mitigation measure, "Do not use hexazinone, picloram, dalapon, or dicamba where they are expected to enter groundwater or surface water, such as when soils are very sandy or have low clay or organic matter content," monitoring of both water sources was undertaken.

Logistical and physical conditions constrained the location of groundwater monitoring. Where possible, groundwater and surface water monitoring was co-located to demonstrate whether concentrations of hexazinone in surface water were strictly due to surface runoff or included a base flow concentration from groundwater. Efforts were made to identify monitoring sites within each major drainage of the project area where units to be treated with hexazinone were located. A major physical constraint on final selection was whether a road existed to allow the drill rig to install a groundwater monitoring well at the down slope end of the treatment unit.

Monitoring wells were installed to enable sampling of groundwater at the base of the unconfined upper aquifer extending from the ground surface to the base of the overlying regolith. This permitted installation of the monitoring wells using a hollow-stem auger. Drilling would cease at auger refusal when it penetrated to the regolith-rock interface. The drill hole was logged, as was the installation of the monitoring well. Each monitoring well was a 5.1 cm diameter well cased with a combination of PVC pipe and stainless steel screening (Figure 6). Wells were installed to the standards for monitoring wells promulgated by the California Department of Water Resources Department (CDWR 1991).

Figure 6. A schematic diagram illustrating a typical monitoring well installation and its main components (CDWR 1991). Wells used on the Stanislaus National Forest were too shallow to require a centralizer.



On the four reforestation projects, a total of twelve groundwater monitoring wells were installed. While most monitoring wells were drilled in areas of granitic bedrock, several were within volcanic or metamorphic bedrock areas. Of the twelve wells, one was destroyed by reforestation work after its initial baseline sampling took place. Seven others were rendered unusable because of a decision to modify the treatment by omitting use of hexazinone, or due to lack of sufficient rainfall following application for mobilizing hexazinone.

Groundwater was sampled from each well using a bailer (Figure 7). Bailers were dedicated for each well to avoid cross-contamination, and water present within the well casing was purged prior to taking a sample. The volume of water present was determined by using an electric water level indicator to measure the depth to water. Calculations would then determine how many times the bailer would have to be used to remove the entire stored volume. The temperature and pH of the water was taken prior to starting the purge and after the first and second purges were completed. Typically, the temperature dropped and a shift in pH was noted between the pre-purge and first purge values. If the second purge values were identical to those after the first purge, it was concluded that a water sample would be representative of the groundwater present around the well screen. In some instances, a third purge was done because of continuing differences in temperature and pH. Following purging of the well, a water sample was collected using the bailer and placed in 1000-mL glass bottles provided by the commercial laboratory where the samples would be taken for analysis. Duplicate samples were included among those collected from different wells to provide a check on laboratory analysis. The bottles were placed in cooled chests and transported to the laboratory to avoid exceeding the appropriate hold time prior to analysis. The same analysis laboratory was used for all the samples taken from the Stanislaus National Forest projects.

For any particular monitoring well, groundwater sampling took place just prior to the initial application of hexazinone to the treatment unit. This provided a baseline to demonstrate that hexazinone was not already present in the groundwater. Initial post-treatment sampling took place after the first major storm event following application of hexazinone. A major storm was defined as one that provided fifty or more millimeters of rainfall to the unit. In subsequent years, samples were collected in the spring following the winter rainy season, or after snowmelt took place, depending on the elevation of the monitoring well location.

Figure 7. The senior author (DeGraff) purging monitoring well BS-45 with a bailer on the Stanislaus National Forest. A second bailer is used to collect the sample after well purging. A housing installed flush to the ground surface protects the well between sample events. See Figure 6 for details.

Among the four wells successfully used to sample groundwater, hexazinone was not detected at concentrations that approached, much less exceeded, the State of California water quality goal of 200 µg/L (Table 1). It should be noted that the Environmental Protection Agency revised its health advisory during this period. This resulted in the State of California easing its water quality goal for hexazinone to 400 µg/L. The rate of application for hexazinone, in pellet form, for the treatment units where the groundwater monitoring wells were sited varied from 33.6 to 41.4 kg/ha.

Hexazinone was not detected during the year of application in any of the monitoring wells (Table 1). For wells where application was ground based, it was detected in the first year following application (second year sampling). For wells where aerial application took place, hexazinone was not detected until the second year following treatment (third year sampling). However, the sample size is too small to make any inference from this observation. What is not surprising is the delay before detectable amounts of hexazinone were found in groundwater samples. Neary et al. (1993) had noted that movement of hexazinone in shallow groundwater took from months to a year following hexazinone application before detection. Based on the Stanislaus National Forest sampling, hexazinone demonstrated multi-year persistence, lasting from one to four years following detection. Again, this is too small a sample upon which to base broad conclusions.

Conclusions

Both the Unit 505 monitoring and post-fire groundwater monitoring well results demonstrate that hexazinone used in reforestation efforts in the southern Sierra Nevada does enter unsaturated and saturated groundwater zones. Monitoring to date does not find those detected concentrations approaching or exceeding the initial State of California water quality goal of 200 µg/L or the later less restrictive value of 400 µg/L.

On the Sierra National Forest, Unit 505 monitoring demonstrated that hexazinone can penetrate a significant distance into the 7.6-m buffer on either side of a Class 4 channel centerline. The detectable concentrations are a full magnitude lower than the State of California water quality goal. The pattern of mobility at these sensitive sites clearly shows peak concentrations of hexazinone in surface water following the first storm event and a gradual rise to peak concentrations of hexazinone in the vadose zone water after several storm events.

On the Stanislaus National Forest, monitoring well results yielded persistence information that has implications for groundwater monitoring in future reforestation projects. Plans for monitoring of the saturated groundwater zone should extend for two years of sampling following the year of application to ensure detection of hexazinone. Once detected, hexazinone will likely persist for one to four more years.

Monitoring for herbicide impacts to groundwater requires a significant commitment of time, personnel and funding. However, monitoring information is important to improving project design and demonstrating expected performance to a sometimes skeptical public.

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Table 1. Well number, application data, and detected concentrations of hexazinone (in µg/L) for monitoring wells used on Stanislaus National Forest reforestation projects.

Well	Application Method	Application Rate (kg/ha)	Concentration of Hexazinone (in µg/L) for Year 1	2	3	4	5	6	7
MW 26-12	Ground broadcast	34.7	ND	**2.6**	**3.1**	*NS*	**0.56**	**0.44**	ND
MW 26-21	Ground broadcast	41.4	ND	**0.99**	**2.2**	**0.16**	ND	---	---
MW 26-56	Aerial broadcast	33.6	ND	ND	**0.29**	**0.76**	**0.71**	ND	---
MW BS-45	Aerial broadcast	33.6	ND	ND	**0.17**	**0.28**	ND	---	---

ND - substance absent or below detection limit of test
***NS* - no sample taken**

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