Memorandum

To: RACER Project Files

From: Pete Shanahan

Date: June 25, 2004

Re: Review of Robert Gilkeson Report

LANL/General/Ground Water

This memorandum provides a review of a draft report *Groundwater Contamination in the Regional Aquifer Beneath the Los Alamos National Laboratory* written by Robert Gilkeson and provided to the Northern New Mexico Citizen's Advisory Board (Gilkeson, 2004).

Gilkeson (2004) presents the conclusion that various drilling fluids used to construct the monitoring wells installed by the Los Alamos National Laboratory under their Hydrogeologic Workplan prevent the collection of representative samples of groundwater from these wells. The drilling fluids alter the chemistry of the aquifer near the wells such that radionuclides that should be detected cannot be detected. He also concludes that the wells, therefore, do not satisfy the requirements for monitoring wells under the Resources Conservation and Recovery Act.

I found that Mr. Gilkeson's report raises concerns that merit consideration. The following provides some background on the issues raised by Mr. Gilkeson, a technical review of the issues he raises, and an evaluation of the regulatory consequences of those issues.

Background

The Hydrogeologic Workplan was prepared by LANL in March 1998 and lays out a program to develop a more complete understanding of the complex hydrogeology on the LANL site and its vicinity (LANL, 1998). The centerpiece of the Workplan is a program to install 32 boreholes into the regional aquifer and to complete those boreholes as monitoring wells in the intermediate perched aquifer and the regional aquifer. Well installation commenced in 1997 and has been continuing since.

LANL installed the first of the wells (R-9 in September–December, 1997; R-12 in March-June 1998) without the use of drilling fluids. Later wells installed by LANL were drilled using various proprietary drilling fluids including TORKease polymer, EZ-MUD (polyacrylamide-polyacrylate copolymer), QUIK-FOAM, Ben-seal Bentonite, Bentonite Gel, Aqua-Guard Bentonite, Pel-Plug Bentonite, Cellophane, Mag Fiber, and Nylon. A list of drilling fluids used based on the LANL well completion reports is provided as Table 1. Short descriptions of the additives are provided in Table 2.

Table 1. Summary of Information on Reported Use of Drilling Additives in LANL Wells

Well ID	Additives used	Quantities reported?
R-5	EZ-MUD, QUIK-FOAM	No
R-7	EZ-MUD, foam polymer	No
R-8	EZ-MUD, QUIK-FOAM	No
R-9	none	
R-9i	none reported	No
R-12	none	
R-13	"Polymer additives," QUIK-FOAM, EZ-MUD	No
R-14	Bentonite, LIQUI-TROL, QUIK-FOAM, soda ash, N-Seal, Magma Fiber	Yes
R-15	TORKease polymer, EZ-MUD	No
R-16	QUIK-GEL, LIQUI-TROL, foam, soda ash, EZ-MUD, Magma Fiber, Pac-L, N-Seal	Yes
R-19	TORKease polymer, EZ-MUD, QUIK-FOAM	No
R-20	QUIK-GEL, LIQUI-TROL, QUIK-FOAM, soda ash, EZ-MUD, Magma Fiber, Pac-L, N-Seal	Yes
R-22	QUIK-FOAM, EZ-MUD	No
R-23	Bentonite, LIQUI-TROL, QUIK-FOAM, soda ash, Pac-L, N-Seal, Magma Fiber	Yes
R-25	QUIK-FOAM, EZ-MUD, MF-1, Ben-seal Bentonite, Bentonite Gel, Aqua-Guard Bentonite, Pel-Plug Bentonite, Cellophane, Magma Fiber, Nylon, TORKease	Yes
R-31	TORKease polymer, EZ-MUD	No
R-32	QUIK-GEL, LIQUI-TROL, QUIK-FOAM, soda ash, EZ-MUD, Magma Fiber, Pac-L, N-Seal	Yes
CdV-R-15-3	QUIK-FOAM, EZ-MUD	No
CdV-R-37-2	QUIK-FOAM, EZ-MUD	No
MCOBT-4.4	EZ-MUD, foam polymer	No

Table 2. Description of Drilling Additives Used in LANL Wells

Product Name	Purpose	Description
AQUAGUARD®	Subsurface grouting material	One-sack sodium bentonite grout (granular-30 mesh)
BENSEAL®	Sealing and plugging agent	Coarse ground, granular sodium bentonite (8-mesh)
Bentonite/Bentonite Gel	Sealing and plugging agent	Bentonite clay
Cellophane	Control of lost circulation	Cellophane - form not specified
EZ-MUD®	Borehole stabilizer/Viscosifier	PHPA liquid polymer emulsion
LIQUI-TROL™	Filtration control/viscosifier	Modified natural cellulosic polymer suspension
Magma Fiber	Circulation control	Extrusion spun mineral fiber
MF-1	Well-bore stablizer	Polyacrylamide
N-SEAL™	Control of lost circulation	Acid soluble extrusion spun mineral fiber
Nylon	Control of lost circulation	Nylon - form not specified
PAC™-L	Filtration Control Agent	Polyanionic cellulose
PEL-PLUG	Sealant	High swelling western sodium bentonite pellet
QUIK-FOAM®	Foaming agent	High expansion, biodegradable liquid surfactant blend
QUIK-GEL®	High-yield gelant / viscosifier	High yield treated sodium bentonite
Soda ash	pH and hardness control	Sodium carbonate
TORKease	Lubrication	Polymer

The LANL well-completion reports vary in the degree of detail provided regarding the types and amounts of drilling fluids used. There are four general styles of presentation:

- A highly detailed tabulation of the types and quantities of drilling fluids used by depth interval: R-25 (Broxton et al. 2002)

- A summary table showing the types and total quantities of drilling fluids used in completing the well over large depth intervals: R-14 (LANL 2003d), R-16 (LANL 2003e), R-20 (LANL 2003f), R-23 (LANL 2003g), R-32 (LANL 2003h)

- A tabulation of the depth intervals over which drilling fluids were used, but not a complete record of the types and quantities of fluids: R-15 (Longmire et al. 2001), R-19 (Broxton et al. 2001b), R-31 (Vaniman et al. 2002)

- Acknowledgement that drilling fluids were used, but no detailed information on drilling fluid use: R-5 (LANL 2003a), R-7 (Stone et al. 2002), R-8 (LANL 2003b), R-9i (Broxton et al. 2001a), R-13 (LANL 2003c), R-22 (Ball et al. 2002), CdV-R-15-3 (Kopp et al. 2002), CdV-R-37-2 (Kopp et al. 2003), and MCOBT-4.4 (Broxton et al. 2002).

There are some seeming contradictions in the information available. For example, Dr. Patrick Longmire has reported the water quality of well R-9i to be compromised by drilling additives (Longmire and Counce 2003), but the well completion reports do not indicate the use of additives in this well.

Impact of Drilling Fluids

The drilling fluids used during well installation contain two potentially problematic classes of constituents: organic materials and clay minerals. Organic materials biodegrade over time, consuming oxygen in the process. There results anaerobic conditions in the well and adjacent groundwater and a chemically reduced state. The creation of reducing conditions in turn alters the chemical state of some metals and radionuclides. Some chemicals transition from essentially immobile species that precipitate as solids or adsorb strongly to aquifer solids to highly mobile dissolved species. Thus, the apparent chemical makeup of the groundwater as found in collected samples may be significantly altered from natural conditions.

Bentonite drilling muds and other additives containing clay alter the chemistry in a different way. Many clay minerals have high ion exchange capacity and thus the capability to adsorb some metal and radionuclide ions. As a result, clay minerals may make contaminants that are mobile in the natural groundwater immobile in the well and nearby aquifer invaded by clay drilling muds.

LANL has recognized and acknowledged that the use of drilling fluids compromises the characterization of water quality. The well completion reports include numerous references to water-quality samples compromised by drilling fluids. In addition, LANL has described the problems in public meetings. For example, at the October 2003 LANL Quarterly Groundwater Meeting, Dr. Patrick Longmire of LANL discussed the water chemistry compromises associated with EZ-MUD. He said that several of the wells showed anaerobic conditions and other indications that the EZ-MUD was undergoing biodegradation and changing the water chemistry in the process. It appeared this effect was diminishing over time as the EZ-MUD was being degraded, but that it could persist for years. The implication is that water-quality samples from these wells were not currently indicative of actual groundwater conditions. Longmire said that residual drilling fluids compromised water-quality data from R-7, R-9i, R-19, R-22, and R-32. The reported results for uranium in R-9i were specifically cited as compromised. Dr. Longmire presented compelling data to illustrate the alteration of the redox state and water chemistry in at least some of the monitoring wells.

Geochemistry reports have been issued for several of the characterization wells and further discuss the compromises to water quality due to drilling fluids. We have reviewed reports for R-7 (Longmire and Goff 2002), R-9 and R-9i (Longmire 2002b), R-12 (Longmire 2002c), R-15 (Longmire 2002a), R-19 (Longmire 2002d), and R-22 (Longmire 2000c). The compromises to water chemistry associated with degradation of the organic EZ-MUD additive are discussed in all reports except for R-15, which appears to not have been significantly affected by drilling fluids. The effects of EZ-MUD are observable in elevated alkalinity, a reduction in oxidized species (for example, nitrate and sulfate), an increase in reduced species (ammonia and sulfide), and an increase in dissolved (reduced) iron and manganese. The effects of the EZ-MUD appear to be decreasing over time in some wells, such as Screen 4 at Well R-22, as evidenced by slow decreases in the concentration of dissolved iron, manganese, and ammonium (Longmire 2002d). In other wells, such as Screen 1 at Well R-22, there is no apparent change over time (Longmire 2002d).

In its reports and presentations, LANL has focused on the effects of organic drilling fluids and has not discussed the potential effects of clay drilling muds (bentonite) in compromising well water quality. Longmire (2002c) mentions ion exchange from drilling muds as a possible source of elevated ammonium in Well R-12, but does not address the potential for drilling muds to remove radionuclides, metals, and other ions. In his report, Mr. Gilkeson places great emphasis on the potential effects of bentonite in adsorbing radionuclides that would otherwise be detected in the monitoring wells. The following examines the potential effect of bentonite clay mud on water chemistry.

Bentonite is a clay with a high percentage of sodium montmorillonite. It has a high cation exchange capacity and, as such, can influence the chemistry of water in contact with it (Fetter 1993). The potential for bentonite-based drilling fluids and well cements to compromise monitoring wells has been recognized by various authors including Claassen (1982), Walker (1983, as cited by Driscoll 1986), Ericson et al. (1985), Brobst and Buszka (1986), Gibb and Jennings (1987), Puls and Barcelona (1989), ASTM (1990), and Hix (1993). The consensus of these authors is clear: where possible, drilling fluids should not be introduced into the borehole during drilling. That said, the authors recognize that drilling in some geologic formations requires drilling fluids if a well is to be successfully installed. One of those situations is in drilling deep wells or in unstable formations. Both situations are encountered in drilling at LANL. For these situations, the authors recommend that the completed well be thoroughly developed to purge residual drilling fluids from the well. Significantly, even guidance documentation from the EPA recognizes that drilling fluids may be necessary. For example, Puls and Barcelona (1989) give the following advice:

> If no alternative to the use of drilling muds or fluids exists, these materials must be removed from the well bore and adjacent formations by careful well development.

Similarly, mud-rotary drilling methods are included among the methods presented in EPA guidance documents for the RCRA (EPA 1986) and Superfund (EPA 1987 and 1993) programs.

Hix (1993) presents a useful summary of mud-drilling techniques for monitoring wells. He stresses that a key requirement is the use of appropriate equipment. The mud system should include a mud mixer and appropriately sized mud pump among other equipment. The use of proper components ensures that the mud will be used appropriately during the drilling process and that the drilling will proceed faster, with less opportunity for mud to invade the geologic formation. The mud system employed during the LANL drilling was a complete and appropriate system as described in the well completion report for Well R-14 (LANL 2003d):

> The system included a mixing tank and pump assembly, a generator to power the mixing unit, a de-sander unit for removing solids from the discharged drilling fluids, and a large auxiliary pump.

Use of drilling fluids was an unavoidable or pragmatic option for the LANL wells given their considerable depth and the character of the geologic formations penetrated. As such, these wells then have a need for thorough well development and cleaning after completion. The LANL well-completion reports include detailed descriptions of the well development process at each well. The process was generally extensive; for example, development of well R-14 began on July 19, 2002, and continued through October 10, 2002. Altogether, over 200,000 gallons were removed from the well during the development process. The extensive development effort is no guarantee that the well is free of contaminants, but certainly illustrates a good-faith effort to develop it properly. LANL has also continued to monitor the wells with specific attention to the effects of drilling fluids. The Laboratory continues to label some wells as compromised by drilling fluids and not representative of actual groundwater chemistry.

Review of Gilkeson Analysis

Gilkeson (2004) draws upon detailed analyses of the chemistry at wells R-7 and R-22 to conclude that the aquifer is contaminated by radionuclides but that the contamination is masked in the monitoring wells by the altered geochemistry. While his overall conclusion is clearly stated, I found his arguments difficult to follow in detail. He completes a review of the levels of strontium and strontium-90 in Well R-7, showing over four quarterly sampling rounds that concentrations decrease consistently. He then writes, "The trend analyses presented in Figure 5 of the analytical results for well R-7 confirm that the radionuclide contaminant strontium-90 is present in the regional aquifer below Los Alamos Canyon." The chemical mechanism to support this conclusion is not clearly stated. I presume that Gilkeson takes the decrease in concentration over time to be the result of progressive removal of strontium-90 from the groundwater by ion exchange with the clay minerals in the drilling mud and the earliest and higher concentrations are more representative of the aquifer. I am not convinced that this explanation is valid. Ion exchange ought to be much more rapid than the quarterly time scale of these samples, and I see no reason for its effects to become increasingly more pronounced over time. An alternative explanation is that the initially rapid biodegradation of the organic drilling muds created reducing conditions, increased alkalinity, and caused more dissolution of natural ions from the formation in response to the altered chemical equilibrium. As the chemical conditions in and near the well gradually return to normal after drilling, there is a decrease in concentrations of dissolved ions, total dissolved solids (as indicated by specific conductance), and alkalinity. Under this hypothesis, the wells are not progressively deviating from equilibrium conditions in the aquifer, but progressively returning to equilibrium conditions. This trend analysis points to the end point, with low concentrations, as chemistry representative of natural aquifer conditions. Another problem in Gilkeson's analysis is that the concentrations of strontium-90 are all below detection limits and thus of uncertain reliability.

Gilkeson (2004) concludes that the geochemistry at Well R-22 is compromised by drilling fluids and prevents an accurate understanding of the aquifer at that location. I concur in that conclusion, at least with respect to the current conditions in the well. For the same reasons as discussed above, I am not persuaded that his time-trend analysis proves the presence of radionuclides at Well R-22. This possibility clearly needs to be evaluated over time, but it is premature to draw conclusions while the well chemistry is compromised.

Gilkeson (2004) and LANL differ in that Gilkeson implicitly portrays the consequences of drilling mud as permanent whereas LANL in its various well completion and geochemistry reports implies it is temporary (although on a scale of years). There is no doubt that at least some wells are compromised and do not yield useful information as yet. This is disappointing for all who wish to understand as soon as possible the nature and extent of contamination in the aquifers beneath LANL. Nonetheless, the consensus of the literature is that wells drilled using drilling muds and other additives are far from irreparably damaged. Indeed, EPA guidance documents and many literature references indicate that such wells can be useful after proper well development. In light of the consensus of the literature, it appears premature to write off all LANL wells in which drilling fluids were used as unsuitable for groundwater monitoring.

Gilkeson (2004) further argues that the wells are a violation of the RCRA. I found his arguments here unconvincing in that EPA guidance for the RCRA and CERCLA programs recommends mud-rotary well drilling when required. Further, it is not clear that these wells will serve as RCRA monitoring wells, per se. The hydrogeologic workplan sought to install wells to provide a general characterization of the aquifer systems, particularly in areas not previously explored. Many of these wells could be characterized as "fishing expeditions" trying to find out whether conditions were problematic and warranted further investigation. With this investigatory aspect of the work plan as context, I do not foresee that all, or perhaps even many, of these wells will see use as RCRA monitoring wells. That said, wells in which the chemistry is compromised by drilling fluids need close evaluation before conversion to RCRA groundwater monitoring wells.

Gilkeson (2004) also criticizes the fact that a number of wells have very long screen lengths and that screen lengths should be no longer than 10 feet. This general rule is appropriate at most sites in which contamination is shallow and localized, and in which aquifer units are relatively thin. This is less of a concern at the LANL site because the vertical length scales associated with the groundwater system are relatively long. Longer screen lengths are appropriate in the regional aquifer because the thick and non-uniform vadose zone can be expected to act as a relatively diffuse source of contamination to the regional aquifer, and because the regional aquifer is itself so thick and non-uniform. Furthermore, for the relatively broad characterization sought in the Hydrogeologic Workplan, longer screen lengths are desirable and are more likely to detect the presence of contamination. Eventually, wells with shorter screens may be necessary at particular locations where contaminant plumes are identified and better quantification for contaminants is needed.

Gilkeson (2004) similarly criticized the geologic formations selected for certain wells, saying that zones of high hydraulic conductivity were missed. The uncertain nature of the subsurface makes all hydrogeologists "Monday-morning quarterbacks" to some extent, even for their own work. The type of second-guessing indicated by Gilkeson is not unusual and certainly not unexpected for the Hydrogeologic Work Plan, which was intended as a fairly generalized characterization effort. All hydrogeologic investigations raise questions and suggest additional locations to be explored in the future; obviously, future wells will need to be installed to fill the information gaps identified in this characterization.

Gilkeson (2004) criticizes the analysis of hydraulic conductivity completed by LANL. I found this section of the report to be too incomplete to evaluate. For example, he states that LANL used "wrong analytical methods to interpret the test data" without indicating the test methods used, why they are wrong, and what test methods should have been used instead. He also seemingly criticizes the tests for not recording high hydraulic conductivity values as observed in prior tests. This reflects LANL's choice of screened intervals to some extent, but may also simply be a reflection of natural variability of aquifer materials. It is common to find wide ranges in the results of aquifer field tests in a single simple formation, let alone in one of the complexity of the regional aquifer at LANL.

Conclusions and Recommendations

There is little doubt that the use of drilling fluids has compromised the water quality in many of the LANL wells. This compromise is unfortunate but not necessarily avoidable in the first place or permanent in its effect. The conclusion by Gilkeson (2004) that the damage is irreparable is contrary to EPA guidance and the seeming consensus in the technical literature. The current approach by LANL appears prudent: that is, to monitor the wells over time, recognize that water chemistry in some wells is not currently representative of the actual aquifer, and observe trends to evaluate whether the wells are approaching an equilibrium representative of the aquifer. LANL's effort in this regard has focused on the effects of degradation of organic drilling muds and has been carefully documented in the well geochemistry reports by Longmire. This effort should be expanded to include an evaluation of potential effects of bentonite additives on chemical species and radionuclides subject to ion exchange.

My review has indicated uneven documentation of the drilling techniques and fluids used during the LANL well installations. Those information deficiencies should be corrected. A complete tabulation of the types and quantities of drilling fluids use by depth interval is essential. This tabulation should be compared with the screened intervals in the installed wells to identify which drilling fluids have the potential to affect which well screens.

Gilkeson's (2004) conclusion that radionuclides are present in the aquifer at some wells (and specifically R-7) but masked by altered water chemistry appears to be premature at best. He does not state his hypothesized mechanism behind this conclusion with great clarity, but it is clear that the available data are subject to alternative explanations that would lead to the conclusion that radionuclides are, in fact, not present. Those alternative mechanisms of water chemistry are presented with considerable clarity in the multiple reports by Longmire and are more persuasive in my opinion. Nonetheless, as recommended above, additional consideration should be given to Gilkeson's (2004) hypothesis as the well data continue to be evaluated over time.

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