



## Department of Energy

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87-GTB-162

DEC 1, 1987

Those on Attached List

Ladies and Gentlemen:

### TRANSMITTAL OF INTERNAL IODINE-129 SUMMARY DOCUMENT

At your request, I am providing a copy of an internal Westinghouse Hanford Company (WHC) paper entitled "A Proposal to Change the Iodine-129 Concentration Evaluation Criterion in the Hydrology Option Paper," by R. E. Gephart, S. H. Hall, and E. I. Wallick. This paper contains (1) a summary of Iodine-129 data from the region around Hanford and from the Reservation proper, (2) information on the theoretically expected Iodine-129 natural abundance in basalt, (3) information on sources of sample contamination, and (4) information on Iodine-129 analytical sensitivity.

This paper is being provided in draft format, it has not been transmitted by WHC management to DOE/RL for action, and is not endorsed by DOE/RL. It is provided to you because it contains a summary of information germane to the Iodine-129 program discussions. It specifically does not represent project plan direction.

Sincerely,

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A PROPOSAL TO CHANGE THE IODINE-129 CONCENTRATION  
EVALUATION CRITERION GIVEN IN THE HYDROLOGY OPTION PAPER

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SEPTEMBER 8, 1987

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## Summary

This paper proposes that the Iodine-129 (I-129) concentration of  $10^{-8}$  picocurie per liter (pCi/L) given in the option D hydrology strategy report (GWG, 1987) is three orders of magnitude too stringent because:

- o Standard drilling and groundwater sampling procedures are not adequate to assure collection of such sufficiently pure water samples,
- o Natural flow system interconnection and or man-made mechanisms by which I-129 was introduced into the confined aquifers from Hanford waste water disposal operations are not understood,
- o Naturally occurring I-129 background levels in a basalt-groundwater environment are uncertain, and
- o Above background levels of I-129 are already present in portions of the confined aquifers in the vicinity of the new boreholes identified in GWG (1987) and in the pump test area at the RRL-2B site. This iodine was introduced from past drilling operations when hundreds of thousands of gallons of mud and Columbia River water were lost into basalt flow tops. The I-129 concentration in river water is  $10^{-5}$  pCi/L.

It is likely that I-129 concentrations above the  $10^{-8}$  pCi/L evaluation criterion listed in GWG (1987) for groundwater samples from the first large scale pump tests already exist in the RRL-2B area. Even if this contamination did not exist, there is still no method to detect I-129 at the  $10^{-8}$  pCi/L level against the probable levels of surface-source contamination introduced through drilling fluids. If an I-129 evaluation criterion is to remain in the option D report, then a new value of equal to or less than  $10^{-5}$  pCi/L is proposed for water discharged during the first pump tests. This value would be identified as the "likely background concentration" for I-129. The previous value of  $10^{-8}$  pCi/L would remain in the option report but be qualified as "best analytical sensitivity" for I-129 detection.

The option D report states that if any evaluation criterion is found to be exceeded, then additional testing and analyses would be needed to determine if said condition was "pervasive" across the site. In other words, any I-129 findings must be examined in the context of all hydrochemical, hydraulic, and geologic data useful in defining flow system dynamics. Nevertheless, any evaluation criterion used in the option paper must be a meaningful number. Presently, one can not distinguish, using I-129 values alone, between iodine introduced into the basalts from natural flow system leakage and that artificially introduced by borehole drilling or past waste disposal operations. Nor is there a supportable basis for establishing a true background concentration for I-129 in a basalt-groundwater environment.

## 1.0 Introduction

The objective of this paper is to propose a change in the iodine-129 (I-129) concentration given as an evaluation criterion in the option D hydrology strategy report (GWG, 1987). That report outlines the geohydrologic testing program to be undertaken before construction of the first exploratory shaft. The evaluation criteria contained in GWG (1987) apply to the test results of and groundwater samples collected during the first large scale pump tests. The collection of drill and test information (i.e. hydraulic testing and or groundwater sampling during borehole drilling) is not planned for boreholes DC-24, -25, -32, and -33.

The rationale for including I-129 as an evaluation criterion in the option D paper is discussed in section 2.0. This is followed in section 3.0 by a summary of what is known about I-129 concentrations in the local surface and groundwater flow system. This discussion is condensed from information contained in section 3.9.5.5 of the Site Characterization Plan (SCP). Sections 4.0 and 5.0 address background levels of I-129 and how it can be introduced into groundwater samples via borehole drilling and water sample collection. Section 5.0 also presents an evaluation of borehole cleanup needs to remove drilling fluids. Report conclusions are summarized in Section 6.0. These discussions are followed by section 7.0 which proposes a specific text change to the option D paper. This change reflects a more realistic role on how I-129 can be used to support the hydrology site characterization strategy.

## 2.0 Background

An I-129 concentration of  $10^{-8}$  picocurie per liter (pCi/L) was given in GWG (1987) as one of several criteria related to:

"... evaluating the presence of disqualifying conditions... Should the testing program provide data that exceed the evaluation criteria and thereby indicate the potential presence of a disqualifying condition, all available data related to that criterion will be evaluated and/or additional testing will be performed to confirm whether the data are representative of the site and the condition if pervasive across the site." (page 4).

The above I-129 concentration was selected because it was the understanding of the working group that levels above  $10^{-8}$  pCi/L ("background") of I-129 was not expected in the Grande Ronde Basalt. Therefore, this concentration was identified as an evaluation criterion in the GWG (1987) report. Any higher iodine values measured during pump

testing was thought to suggest vertical groundwater leakage. Such leakage might indicate the presence of a disqualifying condition (rapid groundwater travel time) for the basalt site. However, the authors of this white paper (Gephart, Hall and Wallick) believe that one should be cautious in using induced leakage (and any associated I-129) as a potential criterion for disqualification. Rather, one should use leakage under equilibrium conditions.

However, it appears that the following information was not adequately factored into the option D report: (a) information on known and suspected I-129 concentrations in the groundwater system, (b) recognized uncertainties in attempting to drill holes and sample waters for detecting low I-129 concentrations, (c) the extraordinary borehole cleanup needs for removing past drilling fluid losses containing above background I-129 levels, and (d) the uncertainty associated with what is a true I-129 background level.

### 3.0 Presence of I-129 in Surface Water Near and Groundwater Beneath the Hanford Site

Iodine-129 concentrations of  $6 \times 10^{-5}$  pCi/L and  $2 \times 10^{-2}$  to  $8 \times 10^{-3}$  pCi/L have been detected in the Columbia River and Hanford 300 Area rain water, respectively. Price et al. (1985) reported that I-129 concentrations in the Columbia River during 1984 ranged from  $1.2 \times 10^{-5}$  pCi/L upstream from the Hanford Site to  $7.4 \times 10^{-5}$  pCi/L downstream from the Hanford Site. Within the unconfined aquifer, I-129 concentrations range from  $1 \times 10^3$  pCi/L to  $1 \times 10^{-6}$  pCi/L. Concentrations are primarily influenced by the sampling site's closeness to present or past waste disposal facilities. The highest concentrations generally occur within and adjacent to the 200 Areas. Iodine-129 concentrations within the confined aquifers of the Saddle Mountains and Wanapum Basalts range from about  $10^{-2}$  pCi/L to  $10^{-6}$  pCi/L.

Most I-129 data for the confined aquifer system are from the Rattlesnake Ridge and Mabton interbeds. Fourteen boreholes completed in the Mabton interbed were periodically sampled in the 1970's. Most of these are located along or near the eastern extension of the Umtanum Ridge-Gable Mountain anticline. This is an area of known vertical groundwater leakage that is or has taken place along an area of folded and faulted basalt (Gephart, et al. 1976, Graham et al. 1984, DOE 1986).

The U.S. Department of Energy derived concentration guideline (draft) for I-129 is 500 pCi/L (Gerton, 1987). The U.S. Environmental Protection Agency drinking water standard is 1.0 pCi/L (EPA, 1976).

Large scale pump testing will begin at the RRL-2B site located in the central portion of the controlled area study zone (G&G, 1987). Borehole drilling in this area over some 8 years has resulted in the loss of hundreds of thousands of gallons of drilling fluid into the basalts. Specifically, at the RRL-2 site alone, over a hundred thousand gallons of drilling fluid (water and muds) were lost into the same basalt flow tops scheduled for large scale pump testing. Since Columbia River water

was used for drilling some of these holes, local I-129 concentrations of at least  $10^{-5}$  pCi/L are presumed to already exist in the groundwater to be withdrawn during pump testing.

#### 4.0 Expected I-129 Background Concentrations

This section addresses natural background levels of I-129 in a basalt groundwater system, theoretically calculated limits on I-129 concentrations, and groundwater sample size dependence for measuring low iodine concentrations.

#### 4.1 Natural Background Levels Evaluated for I-129 in a Basalt Groundwater System

Iodine-129 (a beta emitter with a half life of 16 million years) is a naturally occurring radioactive tracer that is useful for study of hydrogeologic processes on time scales up to 100 million years. Iodine-129 is constantly produced in the upper atmosphere by the interaction of cosmic rays with stable elements in the atmosphere and by fission of heavy elements in the crust of the earth. Man-made sources include gaseous emissions from nuclear reactors and reprocessing plants, leaching of high-level radioactive wastes, and fallout from atmospheric testing of nuclear weapons.

Fabryka-Martin et al (1985) calculated that the steady state inventory of I-129 in the hydrosphere is  $1.2 \times 10^{27}$  atoms, neglecting production by neutron-induced U-235 decay. The global average of I-129/total Iodine (I-129/I) in the hydrosphere ranges from  $3 \times 10^{-13}$  to  $3 \times 10^{-12}$ , depending on the extent of mixing between iodine in recent marine sediments with iodine in sea water. If this mixing is assumed to be 50% efficient, then the predicted average hydro-spheric ratio is  $5.5 \times 10^{-13}$ . The I-129/I ratio in deep groundwater was given as  $4.5 \times 10^{-13}$ , a somewhat lower value, because of removal from active exchange with the atmospheric source reservoir.

Fabryka-Martin et al (1985) also presented calculated data for I-129 production rates and steady-state concentrations in various rock types, depending upon the typical concentrations of uranium and iodine. For example, the highest I-129 content would be found in a marine black shale, while the lowest would be found in a halite. The ratio of I-129/I for basalt was given as  $16 \times 10^{-13}$ .

Comparing the ratio for deep groundwater with that of basalt, in theory at least, shows that leaching of iodine from basalt into groundwater can significantly raise the background ratio.

The background activity of Iodine-129 in deep basalt groundwater is, at a minimum, the activity that corresponds to the deep groundwater ratio. According to Fabryka-Martin et al (1985), a minimum of 10 mg of iodine must be available for analysis. This information can be used to arrive at a specific background activity for iodine in groundwater.



Consider the decay equation:  $-dN/dt = N \text{ Lambda}$

Given that:

- o I-129/I ratio in deep groundwater is  $4.5 \times 10^{-13}$ ;
- o Lambda (decay constant for I-129) =  $1.3724 \times 10^{-16} \text{ sec}^{-1}$ ;
- o Minimum of 10 mg total iodine for analysis. Assumed to be present in one liter;
- o Avogadro's number =  $6.02 \times 10^{23}$  atoms per mole;
- o One curie =  $3.7 \times 10^{10}$  disintegrations per second,

The equation may be solved to yield a background activity of  $7.8 \times 10^{-9}$  pCi/L. This is approximately the value of  $10^{-8}$  pCi/L that is given in GWG (1987).

$$\left[ \frac{(4.5 \times 10^{-13}) (10^{-3} \text{ g/L}) (6.02 \times 10^{23} \text{ atoms/mole})}{(1.3724 \times 10^{-16} \text{ sec}^{-1})} \right] \cdot \left[ \frac{1}{(129 \text{ g atoms/mole}) (3.7 \times 10^{10} \text{ sec}^{-1}/\text{Ci})} \right] \dots$$

As mentioned earlier, basalt rock can have a ratio of I-129/I that is about four times larger than the value is for deep groundwater. The contribution of this I-129 to the groundwater resulting from leaching of iodine along the flow path in a basalt groundwater flow system is unknown. Leaching would have the effect of increasing the background to a level somewhat higher than the stated  $10^{-8}$  pCi/L. A discrete value could only be obtained by:

- o Determining the ratio of I-129/I for Columbia River Basalts,
- o Studying the abundance and distribution of iodine in the basalts, and
- o Studying the change in total iodine concentration in groundwater as it evolved chemically along a flow path.

Fabryka-Martin et al (1985) pointed out that total iodine concentrations increase with groundwater age due to leaching of iodine from the host rock. The isotope signature of this second I-129 source will be a function of lithology and formation age. In cases where significant leaching of iodine takes place, the initial isotope ratio of I-129/I in the groundwater would be the same as that of the recharge water. However, then the overall groundwater ratio would increasingly reflect the age of the host rock rather than that of the water.

Another problem in defining the background specific-activity of I-129 in Hanford groundwaters is variable contamination of the system with Columbia River water that has a specific activity of about  $10^{-5}$  pCi/L. Contamination has been variable due to differing volumes of fluid lost into basalt flow tops as a result of drilling and testing on the Hanford Site over the years.

Based upon data from the literature, the uncertainties of iodine leaching, and variable contamination, a background range of  $10^{-8}$  to  $10^{-3}$  pCi/L appears reasonable until additional geochemical data become available. For the present, this range is expressed as equal to or less than  $10^{-3}$  pCi/L.

#### 4.2 Theoretically Calculated Limits and Sample Size Dependency Summarized for Measuring Low I-129 Concentrations

Determining I-129 concentrations in water is complicated by two factors:

- o Low concentrations of iodine in natural groundwater, and
- o Low isotopic abundances of I-129 in natural iodine.

The measurement of I-129 in water requires the isolation of about 10 mg of iodine. Considering that the concentration of iodine in Hanford groundwaters is on the order of 2 parts per billion (ppb), large quantities of water must be collected to obtain the necessary concentrate.

According to Fabryka-Martin et al (1985), tandem accelerator mass spectrometry (TAMS) is the instrumental method of choice for determination of I-129/I ratios as low as  $1 \times 10^{-13}$ . At the 10% level of significance, a sample containing  $10^7$  atoms of I-129 in 20 mg of silver iodide gave a ratio of I-129/I of  $8 \times 10^{-13}$ . This information indicates that current analytical sensitivity is barely sufficient to resolve levels of I-129 at the  $10^{-8}$  pCi/L level. Fabryka-Martin et al (1985) also state that resolution improvements of about one order of magnitude are expected and may already be available.

For I-129 analysis of Hanford groundwaters, the following realities must be considered at this time:

- o Detection of an I-129 concentration of  $10^{-6}$  pCi/L requires a groundwater sample of about 50 L (13 gallons).
- o Detection of an I-129 concentration of  $10^{-8}$  pCi/L requires a groundwater sample of about 5000 L (1300 gallons).

Large quantities of groundwater should not be collected in an attempt to reach background levels that overlap with instrumental detection limits. Not only is the possibility of contamination from large sampling containers a danger, but there is also the increased uncertainty as to which stratigraphic section is actually supplying the water.

## 5.0 Sources of Groundwater Sample Contamination and Borehole Cleanup Needs

The laboratory measurement of I-129 in a groundwater sample represents the weighted average of the concentration of the isotope in the groundwater and that of the residual drilling fluid (which is also part of the collected sample) plus any I-129 introduced into the sample during collection, handling, and analysis. This latter contribution can be minimized and compensated by using scrupulously clean and consistent technique, and by using analytical blanks.

Residual drilling fluid in the sample presents a difficult problem in attempting to collect representative groundwater samples. The following hypothetical example shows how drilling fluid can add enough I-129 to the collected sample to completely mask the true I-129 concentration. Assume that the drilling fluid used to penetrate a basalt aquifer is Columbia River water, taken from the northern side of the Hanford Site. This water has an I-129 concentration of  $10^{-9}$  pCi/L (see Section 3.0). Also, make the unlikely assumption that the drilling fluid has not been further contaminated by contact with air, soil, or groundwater from the unconfined aquifer. Finally, assume that the I-129 concentration in the groundwater is a natural background level of  $10^{-8}$  pCi/L.

Some of the drilling fluid will effectively be injected into the aquifer during borehole penetration. Most of that fluid will be removed by development (cleanup) pumping. Simple calculations show that if the sample collected after development is 99% groundwater and 1% residual drilling fluid, the I-129 concentration of the sample will be  $1.1 \times 10^{-7}$  pCi/L, which is eleven times higher than the assumed true groundwater value. Similarly, if the sample represented 99.9% groundwater and 0.1% residual drilling fluid, the result of laboratory analysis would show an I-129 concentration of  $2 \times 10^{-8}$  pCi/L or twice the actual groundwater value. For this example, the collected water sample must be 99.99% free of residual drilling fluid before the I-129 level is representative within 10% of the actual groundwater concentration of I-129. It is clear that residual drilling fluid contamination is a major problem because it creates an unfavorable signal-to-noise ratio. Data analysis techniques, developed to monitor hydrochemistry at boreholes DC-18 and DC-23, can be used to quantify borehole development. These techniques (Hall, 1986) depend upon time-series sampling and field analysis during development pumping. Results show the following approximate relationship between residual contamination and pumped water volume during development of Hanford basalt aquifers:

Pumped volume/injected volume\*    % residual drilling fluid in samples

1	33%
10	2%
100	0.2%
1000**	0.02%

\* "Injected volume" is the amount of drilling fluid lost to the aquifer during borehole penetration.

\*\* Extrapolated. This value has not been reached in practice.

These results show that to reach the 99.99% cleanup cited in the example above, the water volume pumped during development must be over 1000 times the fluid loss during drilling. This figure is about 10 times greater than has been achieved to date during the best drill-and-test mode sampling conducted on site. At borehole DC-18, a pumped volume-to-injected volume ratio of over 100 was reached in the Rosalia flow top of the upper Wanapum Basalt during a three-week pumping campaign. This large ratio was achieved only because an unusually low volume of drilling fluid (3160 liters or 836 gallons) was lost during penetration. No other drill-and-test horizons have been developed to this degree. It would have required about 30 weeks of pumping (about a million gallons pumped) to reach 99.99% cleanup. Moreover, the pumped water has to come from somewhere. After such a massive pumping campaign, the final collected sample would not represent water from the original borehole site. Rather, it would represent a composite of groundwater induced to flow from the surrounding flow systems. Crossover of contaminants from any nearby boreholes that have penetrated the same aquifer then becomes a very real sample contamination concern, especially if such boreholes were not originally developed to a high degree (this is the usual case with most boreholes at the Hanford Site).

In the hypothetical example, it was assumed that there was no I-129 contamination in the drilling fluid beyond the  $10^{-3}$  pCi/L already measured in the Columbia River water. In reality, there are several sources of contamination. Airborne I-129 from processing facilities at Hanford can contaminate drilling fluids in the mud pits. Similarly, the soils are contaminated, so soil dust is a potential problem. Finally, water from the unconfined aquifer can contaminate the drill string and fluid recirculation system during drilling. This contamination can persist throughout the drilling campaign, i.e., throughout the drilling of the lower confined aquifers.

In the vicinity of RRL-2B, the pumping well designated for the forthcoming pump tests, literally hundreds of thousands of gallons of river water in drilling fluids have already been lost to aquifers, including over 380,000 liters (100,000 gallons) to the Birkett flow top alone. This is one of the flow tops to be pump tested and sampled for comparison against the evaluation criteria given in GWG (1987). Though the I-129 concentrations of these fluids are unrecorded, and the degree of borehole development is uncertain, an I-129 concentration of  $10^{-3}$

pCi/L may be assumed as a minimum. Consequently, the degree of existing contamination of aquifers due solely to drilling activities cannot be evaluated. In other words, I-129 measurements for samples taken during large-scale pump testing may be meaningless. Thus, there is no technical basis for assigning a supportable evaluation criterion ("yellow flag") value for the I-129 measurements.

Using the above information, another calculation can show how inadequate the planned total discharge volume for the first large-scale pump tests will be to remove "above background" levels of I-129 from some test horizons. For example, assume a discharge rate of 19 liters per minute (5 gpm) for 30 days. This gives a total discharge of 819,000 liters (216,000 gallons). Next, make the optimistic assumption that only 380,000 liters (100,000 gallons) of drilling fluid was originally lost into the RRL-2 test horizon. (This is the case for the Birkett flow top.) To reach a 99.99% cleanup, then over 380 million liters (100 million gallons) of water must be removed. At 19 liters per minute (5 gpm), this would take nearly 14,000 days or 38 years of pumping. Clearly, this is an unreasonable expectation to meet an I-129 evaluation criterion that itself has no solid basis.

A fundamental question in reviewing data for any groundwater sample and any laboratory analysis of that sample is, "What does the sample itself represent?" The above discussion has shown that actual groundwater samples collected at the Hanford Site represent part groundwater and part drilling fluid. For I-129 measurements, it has been shown that, within reasonable limits of development pumping, the drilling fluid contribution to a groundwater sample would completely mask the contribution from any uncontaminated deep groundwater.

## 6.0 Conclusions

The following conclusions are drawn from the information presented or referenced in the previous sections:

- o I-129 appears to be present in the confined aquifers near the 200 Areas particularly along the Umtanum Ridge-Gable Mountain anticline. Vertical groundwater leakage takes place within this geologic structure.
- o The mechanism(s) by which Hanford derived I-129 is introduced into the confined aquifers is not fully identified. Explanations include both natural and man-made mechanisms.
- o The true background level of I-129 naturally occurring in a basalt-groundwater environment is uncertain.

- o I-129 at concentrations of at least  $10^{-5}$  pCi/L has been introduced into the groundwater beneath the area planned for the first large-scale pump test. This I-129 is associated with the thousands of gallons of drilling fluids lost into the basalt rock since the late 1970's. These groundwaters are the same as those to be withdrawn during pump tests planned in the option D hydrology strategy report.
- o Based upon the facts that (a) analytical precision is poor in the  $10^{-8}$  pCi/L range, (b) large water sample volumes are needed to approach this range, and (c) there already is large uncertainty in the theoretical natural background for I-129 in basalt groundwater, it is necessary to seriously reconsider what is thought to be a realistic background value for I-129 in the option D strategy paper.
- o Massive borehole development is necessary to remove I-129 already introduced into the basalt-groundwater environment by drilling. The required cleanup exceeds that historically accomplished in past basalt drilling operations. In addition, the planned discharge volumes from the first large-scale stress tests appear to be inadequate to fully clean out the test horizons for low level I-129 measurements.
- o To separate the signal (true background) from noise (drilling fluid contamination) level of I-129, the in situ I-129 concentrations must be no less than about one-hundredth of the concentration in the drilling fluid itself. Otherwise, signal to noise separation is not possible.
- o Until analytical improvements and further geochemical studies are available, the background value given on I-129 in the option D strategy paper should be no more stringent than equal to or less than  $10^{-5}$  pCi/L.

## 7.0 Recommended Text Change to the Option D Report

The only reference to I-129 in the option D paper is in the first page of the table entitled "Strategies to Investigate Disqualifying Conditions". There an evaluation criterion of  $10^{-8}$  pCi/L is given. It is proposed that a footnote labeled "a" be placed next to this criterion. At the bottom of the page, the footnote would read "a=best analytical sensitivity". In addition, it is proposed that a second I-129 value of equal to or less than  $10^{-5}$  pCi/L be listed below the  $10^{-8}$  pCi/L value. Next to this new value, a footnote "b" would be placed. This footnote would be explained at the bottom of the page as "b=likely background concentration".

In proposing these changes, it must be recognized that even a  $10^{-5}$  pCi/L value may be meaningless because of the numerous unknowns discussed in previous sections. This is why section 2.0 emphasized the importance of not taking any single test value (whether it's a hydraulic or hydrochemical value) out of the context of all other known geohydrologic data. That is simple and sound scientific practice.

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