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TRITIUM IN THE BURIAL GROUND OF THE SAVANNAH RIVER SITE (U)

M. L. HYDER

Authorized Derivative Classifier

Michele J. Bullington 6/30/93
Date

Westinghouse Savannah River Company
Savannah River Technology Center
Aiken, SC 29803

SAVANNAH RIVER SITE

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M. L. Hyder, Author	M. J. Hitchler, Manager, STS
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Date	Date

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Introduction

This memorandum reviews the available information on tritium-contaminated material discarded to the burial grounds. It is the first report on a study of burial ground inventories undertaken by the author at the request of the Environmental Restoration Groundwater Section. Tritium was the first isotope studied, because it represents the most immediate concern with regard to release to the environment. Substantial amounts of tritium are known to be present in the ground water underneath the area, and outcropping of this ground water in springs and seeps has been observed. The response to this release of tritium from the burial ground is a current concern.

I have drawn on the following sources for information:

COBRA records of tritium in the burial grounds, supplied by J. R. Cook.

Interviews:

- W. C. Reinig, Health Protection Section, formerly in HP in the burial grounds;
- J. L. E. "Pete" Cheetham, manager of the burial grounds up to his retirement in 1972;
- D. E. "Red" Ward, engineer in tritium production since the late 1950's;
- Sara Davis, keeper and maintainer of burial ground records for many years;
- Bradley Wilson, engineer currently responsible for estimates of tritium sent to the burial ground.

Works Technical Monthly Reports. These are classified "Secret".

Burial ground log book.

Building 232-H log books.

HP periodic reports.¹⁻⁶

Recent summary reports on the condition of the burial ground.⁷⁻⁹

Other reports relating to tritium in the burial ground.¹⁰⁻¹⁴

Summary

The amount of tritium emplaced in the burial ground facilities is very uncertain. These uncertainties come from several fundamental factors:

- There is no record of specific tritium burials before 1961. For some time after that date, the data base has little information on the nature of material buried.
- Tritium burials associated with spent target melts are estimated from a very limited study, and have a large uncertainty associated with the estimates.
- Burials involving discarded equipment are very difficult to estimate, and so many of the estimates are little better than guesses.
- In many of the burials, especially Building 232-H in the early years, waste casks were simply filled with waste material of various types as it was generated, and shipped to the burial ground and buried without any attempt to specify the contents.

It is however possible to reach some general conclusions about tritiated waste burials. In particular, most of the tritium buried is associated with spent equipment and other waste, rather than spent melts. Correspondingly, most of the

tritium in the ground water seems to be associated with burials of this type, rather than the spent melts.

Maps are presented showing the location of burials of tritiated waste by type, and the location of the largest individual burials according to COBRA records.

Discussion

1. Tritium Production.

Tritium production began at the Savannah River Site in the 1950's, and has continued through the most recent reactor campaigns. Essentially all tritium produced in the United States for weapons, research, and commercial uses has been produced in the Savannah River reactors and processed in the tritium facilities on the site. These are presently all located in the "H" separations area, and include the following operations:

232-H: Recovery of tritium from irradiated targets.

234-H: Tritium purification, concentration, and loading for offsite shipment.

238-H: Tritium recycle operations.

A new tritium facility (RTF) that will assume many of these functions has recently been built and put into operation in H Area. It is designed to incorporate improved technology while minimizing offsite releases. A second tritium facility similar to 232-H was operated briefly in 232-F in the late 1950's and then shut down.

Security considerations prevent discussing all aspects of tritium burials. Both tritium production quantities and some of the process technology remain classified. The identity of some of the materials buried is classified, but the associated tritium content has been estimated. This memorandum has been prepared to be an unclassified document, and the only numbers quoted are those from the unclassified COBRA database and other unclassified reports. These are regarded as adequate for the intended purpose.

2. Tritium Properties.

Tritium is the only radioactive isotope of hydrogen. It has a relatively short half-life (12.3 y) and therefore a very high specific activity (9700 Ci/g). However, there is no radiation hazard to personnel working in tritium facilities so long as direct contact or ingestion are prevented. This is because tritium emits only beta particles of very low energy, averaging less than 6 keV. These are absorbed in minute thicknesses of virtually any kind of matter. Consequently, although tritium facilities must be well contained to avoid releases to the building air, radiation shielding is only necessary in those areas where the irradiated target material is handled. This contains gamma-emitting radioactivity from activation of impurities in target and cladding material.

Because tritium emits no penetrating radiation, it can only be measured by internal counters, such as ionization chambers or internal gas proportional counters (for gaseous tritium), or liquid scintillation counters (for liquids or dissolved solids).

There is no practical non-destructive technique that permits an estimate of tritium concentrations in solid waste. Consequently, none of the values cited for tritium discards in the COBRA records are based on monitoring. Generally, they are based on estimates provided by operations personnel, who know the history of the material; often they are simply guesses. There are some measurements of tritium contents of spent melts, and this will be discussed later.

Once tritium is recovered from the irradiated targets, it is primarily handled as the elemental material, i. e. as hydrogen gas. Hydrogen, in the absence of oxidants, is chemically rather inert, and at ordinary temperatures reacts with few materials. This is also true for trace amounts of tritium in elemental form. More concentrated tritium, however, is quite reactive because of ionization and recoil phenomena. Thus high specific activity tritium reacts with air or oxygen and rapidly generates tritiated water vapor. Similarly, tritium exchanges rapidly with hydrogen atoms in organic materials such as oils and sealants. Solid oxides may acquire a surface layer of tritium-contaminated hydroxides. Consequently, during handling tritium must be kept from air and materials that will take up hydrogen under irradiation. A corollary to this is that since most materials have layers of oxide or organic materials on their surfaces, virtually anything exposed to substantial amounts of tritium will be contaminated to some extent. If the material is porous, the tritium can also accumulate in inaccessible internal regions.

Virtually all the tritium discarded to the burial ground is in the form of tritium oxide (tritiated water), or else is in a form (such as metal oxides or hydroxides) that converts to tritium oxide on contact with water. Experiments have shown that this is true even for spent melt material from which tritium gas was recovered. It is not surprising, therefore, that any tritium that has access to the ground water is incorporated into the ground water and moves along with it, for the properties of tritiated water are essentially the same as normal water.

3. Records Pertaining to Tritium Burials.

The records I have located that pertain to tritium burials include:

- A. 232-H log books.
- B. Burial ground log books.
- C. COBRA records.
- D. HP records.

Each of these is discussed in turn.

- A. Building 232-H log books are complete. They are stored in a vault in Building 232-H. Their principal purpose seems to have been to record information needed for shift turnover during operation. Thus the information in them on burial of material deals mainly with the status of the casks and trailer used to transport material to be buried: whether they are full or empty, where located, etc. The number of target components, spent melts, and other specific items discarded in these casks is not recorded in the log.

The main value of these log books is in giving a picture of how these aspects of waste handling operated.

- B. Burial ground log books were maintained by the manager of the burial ground. They are kept in a file in a storage cabinet in Building 724-7G. The contents of these logs vary greatly. For some brief periods enough information is provided to identify shipments from H area and their disposition. For other periods the information is much less complete and of little value. Because of this inconsistency these logs are also mainly useful in providing general operating information.
- C. The COBRA records were derived from HP burial slips. (These are also kept in Building 724-7G.) The COBRA records begin in 1961, which is several years after tritium production began. Hence this information is not available on the first few years of tritium production operations.

The COBRA description of buried items is very spotty, and in the early years generally non-existent. Inferences as to the nature of some of the material (such as spent melts or Mg beds) can be made from the types and amounts of radioactivity specified on the burial sheets, but it appears that this is not reliable, because some materials estimated as spent melts probably were not. In recent years the COBRA records are more reliable and informative.

- D. HP published a summary report between 1959 and 1965 that gives gross information on radioisotope burials.¹⁻⁶ For the years in which comparisons are possible, this report estimates much higher amounts of tritium emplaced in the burial grounds than the COBRA database. For example, the COBRA data for one year (1963) shows that 227,000 Ci of tritium were sent to the burial ground. In the corresponding annual HP report, the total material buried was estimated as 531,000 Ci of tritium.⁵ It appears that these HP values were estimated on the basis of the difference between calculations of the amounts of tritium produced and the tritium product actually recovered. The presumption would have been that anything not otherwise accounted for went into the burial ground. (This basis is not stated in the HP reports. It is inferred to be the method used, because it is the only method that would have been available for calculations after the fact.) If these numbers are correct, they imply that at least twice the tritium has entered the burial ground than the COBRA records account for.

Although the HP numbers appear to be based on sound logic, there is reason to question them. The reconciliation of calculated production and material accountability was a major problem in the early 1960's, and was only gradually resolved. The amount of product discarded to the burial ground was in any case only a small fraction of production, so that calculating the amount discarded depends on the accuracy of a small difference between two relatively large numbers. Inasmuch as the calculations generally overestimated production compared to the amount of material actually recovered, it is reasonable to accept the HP values as upper limits to the amount of tritium discarded for the periods in question. However, the documentable COBRA data are likely to be closer to true values.

4. Tritium Processing and Materials Discarded.

Tritium is produced by the irradiation of lithium-aluminum alloy targets, and is recovered by heating these to the melting temperature. The resulting melted alloy material is referred to as "spent melts". Spent melts are discarded in the stainless steel crucibles in which they were melted to recover tritium. Tritium residues in spent melts are a small fraction of the tritium product originally present, but still make up a significant part of the tritium discarded to waste. For many years the spent melts were discarded without packaging or other treatment. Subsequently, the practice of pouring an epoxy resin into the crucible containing the melts before it was buried was adopted. The effectiveness of this practice in limiting the release of tritium to ground water has not been experimentally evaluated.

Tritium as recovered following melting is partly elemental tritium and partly oxidized. After recovery it is converted to elemental tritium or to metal tritides for further processing. Tritium handling requires a tightly sealed system from which all atmospheric gases are rigorously excluded. Equipment used in handling tritium gas includes a variety of valves, vacuum pumps, and particulate beds that are used for sorption and purification. This equipment and associated materials such as pump oil and mercury were discarded to the burial grounds when they must be replaced. The problem in estimating the amount of contamination associated with these materials is obvious. Tritium on internal surfaces cannot be sampled, and this is particularly true for equipment containing porous particles.

After spent melts, the most commonly discarded items are the magnesium beds used to convert tritiated water to elemental tritium. These must be replaced when chemically exhausted. Pump oil is disposed of by sorbing it on "Oil-Dri" or some similar material, and discarding it in a sealed jar. Mercury was formerly discarded to the burial grounds in a sealed bottle, but this practice was stopped many years ago. Spent, contaminated mercury has since been retained in above-ground storage.

Items of discarded hardware such as pumps have generally been wrapped in plastic and boxed before discarding. While this provides protection to the handlers during transport and emplacement, it is not expected that this packaging will reliably retain tritium after burial. Backfilling will likely collapse the boxing, and the integrity of the plastic under these circumstances cannot be guaranteed. Many of the burials containing the largest amounts of tritium apparently were buried in this fashion; they are identified in the burial records simply as "carton".

A substantial amount of tritium-contaminated waste material from off-site has been buried at SRS. Most of this originated at Mound Laboratory or the Pinellas plant. This material has generally been buried in the shipping containers in which it was received, without repackaging. Steel drums are typical shipping containers. The nature of the waste material, and its internal packaging, are generally not known. The largest single burial in the burial grounds originated at Mound Laboratory and was packed in drums.

Table 1

(From WSRC Manual 5Q1.1, Procedure 501, Rev. 0 (Obsolete)
Section 5.4.3, "Tritium")

<u>Curies</u>		
Type Shipment	Tritium	Induced Activity
Waste Dumpsters	Estimate based on Content (Usually 0.1)	Estimate based on known content (usually 0)
Spent Melts:		
Lithium aluminum (Li-Al)	100/melt 200/double melt	20/melt 40/double melt
Lithium Aluminate (LiAlO ₂)	1000/melt	20/melt
Mg, U, and Z beds	500/bed	0.1/bed

Notes to Table 1: COBRA data appear to differ from these recommendations by a few per cent for reasons that are not known. Melts and double melts cannot be further defined here. Mg, U, and Z beds are used to purify and concentrate tritium. They have a finite life and are eventually replaced. Lithium aluminate has been irradiated only in a few special experimental irradiations.

Table 2

Tritium Burials by Year, from HP Reports¹⁻⁶

Year	Tritium Buried, kCi
Through 1959	1201
1960	159
1961	615
1962	531
1963	531
1964	559

5. Tritium Estimation.

The problem of estimating the tritium content of discarded material is very difficult, because no penetrating radiation is emitted. Estimates have traditionally been made from values established early in the program, but documentation supporting these estimates has not been found. (See Table 1.) The estimates may simply be educated guesses. An exception to this is the tritium content of spent melts. These are melted targets from which virtually all tritium has been extracted by heating; the lithium and aluminum metal is melted and accumulated in a steel crucible. The whole melt-crucible package is discarded to the burial ground as a unit. As will be discussed, the tritium contents of some such packages have been measured.

In light of the inconsistencies between sources of data on tritium in the burial ground, a selection had to be made regarding which data to use. The COBRA data were selected, because they have a documentable basis. For comparison, the HP summary data are given in Table 2.

The tritium content of spent melts can be estimated from a number of experimental measurements:

- A study was made in which cores were drilled from spent melts, and their tritium content was determined by total dissolution and analysis.¹⁵ The numerical results of this study remain classified. However, it can be noted that the results showed considerable inhomogeneity. It appeared that a significant part of the activity was associated with the exterior surface of the melt.
- Two experiments have been performed in which spent melts were subjected to water leaching,^{11,12} and one in which spent melts were buried in a lysimeter apparatus in the burial ground.¹² (A lysimeter is a device that allows all the percolating water from a test area to be collected and analyzed.) The results of the two types of experiment differed substantially. A typical spent melt immersed in stagnant water released from 20 to 40 Ci of tritium activity into the water as HTO. (Release of tritium as insoluble hydrogen gas was negligible.) This release was very fast at first, but quickly decreased to a low level, and was essentially complete within a few weeks. However, the lysimeter test gave a very different result. In this test three melts were buried together, and the natural rainwater percolated through the apparatus and was collected. While there were variations associated with variations in rainfall, the tritium releases continued steadily for more than 90 weeks, the period of the study. At the end of the study, a total of 177 Ci of tritium had been leached from the three melts. It appears that the continual replenishment of fresh water is more effective in removing tritium from the melts.

These results are most easily explained by the assumption that the tritium released is associated with the surface oxides on the aluminum alloy ingot that constitutes the spent melt. Stagnant water is quickly saturated with aluminum and does not dissolve these oxides further. However, rainwater is continuously replenished and is acidic because it contains carbon dioxide and other dissolved gases. It continues to erode the melt surface, and to release tritium.

The corollary to this is that the tritium contained in the bulk of the melt is inaccessible, and will not be released before it decays. Aluminum encapsulation is an excellent method for retaining tritium! For environmental considerations, it is only necessary to consider the tritium near the surface of the melt which is accessible to the ground water. The lysimeter data suggest that this may be on the order of 100 Ci/melt, or about half of the amount estimated (for "double" melts) in Table 1. This result could be combined with spent melt burial data to estimate the activity available from this source.

However, for the early years of operation, the COBRA database cannot be used to identify the number of spent melts discarded to the burial ground. In these early records spent melts are not identified as such. They might be identified with the number of waste shipments from Building 232-H containing both tritium and induced activity. The actual number of spent melts can be determined from classified production records, and these were found in some month-by-month comparisons to be fewer than the number of such burials in COBRA records. It appears that other materials sent from 232-H at the same time as spent melts may have been estimated as if they were spent melts. In later burial records spent melts are specifically identified.

Consequently, for burials of the type that appear to be spent melts, it is reasonable to reduce the estimate of buried tritium by about a factor of two. The bases for doing this are:

- Only about 100 Ci of activity per melt is actually accessible to the ground water; and,
- The waste material in this category that is not spent melt probably has less activity than spent melt material.

Note that there is still an uncertainty of leachable tritium content because of differences among melts. The data suggest that this is of the order of $\pm 50\%$.

For burials other than spent melts, including burials of offsite material, there is no better basis for the amount of buried tritium than is provided by the COBRA record. However rough the guess, it is as good as any guess, and has the advantage of having been made by the people most directly concerned. In individual burials of this sort, where it is desired to estimate uncertainties, an uncertainty factor of two or three should probably be applied.

6. Quantities of Buried Tritium.

The total quantity of buried tritium, uncorrected for decay, was reported by Cook in 1991 as being 172 g (1,670,000 Ci) in the old burial ground, and 240 g (2,330,000 Ci) in the new burial ground.¹⁶ This is based on COBRA records, which give lower values than the HP estimates in Table 2. COBRA records also do not include material emplaced in the old burial ground prior to 1961. Table 2 suggests that approximately 1,800,000 Ci were buried during this time. However, the HP estimates for later years are roughly twice the values recorded in individual burial

records. Applying this factor reduces the unrecorded tritium burials to about 900,000 Ci (93g).

Radioactive decay of the buried tritium has substantially reduced these values. The material emplaced in the old burial ground prior to 1961 has decayed for 32 years, so that its activity has been reduced by at least 83.5%: of the original assumed 93 g, about 15 g remain. The remaining material in the old burial ground, buried between 1961 and 1972, has decayed at least 21 years. This corresponds to 69% decay; thus there is not over 53 g (520,000 Ci) of the original 172 g remaining. About half the material in the new burial ground is represented by the material from Mound buried in 1972, which is also decayed by 69%, and therefore represents only about 37 g (360,000 Ci). Some tritium has also been lost by groundwater transport to the environment, but the available data indicate that this amount is negligible compared to radioactive decay.

The great majority of the tritium in the old burial ground is burials other than spent melts. A review of COBRA data shows that 1,410,000 Ci (about 84%) of the tritium in the old burial ground was in the largest burials (>1000 Ci per item) which were associated with equipment and other process material from H Area and offsite. Spent melts, Mg beds, and other small routine items are all in the remainder. Thus the 50% factor previously derived for the fraction of spent melt tritium available to the ground water does not have a significant effect on the total available tritium. Similarly, in the new burial ground, 1,870,000 Ci (193 g, 80% of total burials) is in burials of 1000 Ci or more. The largest burials, of 5000 Ci or more, are located as shown in the maps in the following section.

7. Locations of Tritium Burials.

Location maps shown in this section are not drawn to accurate scale, because the computer software used does not allow accurate alignment of lengths with widths. Thus the vertical and horizontal scales are not exactly the same. However, the relative placement of points within the map should be accurate.

A. Old burial grounds

Figures 1 through 4 show the location of tritium buried in the old burial grounds. Each of the figures shows a different set of information. Figure 5 shows the approximate location of various types of material in the old burial grounds according to contemporary records. This can be compared with the other maps to show the nature of some of the burial areas in which tritium was emplaced.

The COBRA burial data used to generate this map are not completely accurate. In reviewing the data I found obvious typographical and recording errors, and corrected for them where possible. Generally this could be done by comparison with other materials of similar time buried at about the same time. However, there is a good chance that there are other errors of this type that are not obvious. There are certainly examples of burials that were made in places that appear to be inconsistent with the waste type. The question is whether the error was made in

recording the burial, or else the record really does represent an anomalous treatment of this particular burial.

The translation of coordinate data into graphical data required interpretation. For the old burial ground, coordinate data are given as coordinates labeled "NC" and "EC". Peculiarly, NC data do not represent a north-south measurement, but rather refer to measurement along a scale for which the westernmost point is zero. EC data refer to a coordinate grid in which the northernmost row is "A" and the southernmost is "K". It appears that each of the sections is divided into five subsections: thus the northwesternmost square of the burial ground is carries NC designations from 0.10 to 0.50 and EC designations A.10 to A.50. For graphing purposes these have been translated into a decimal system. Subdivisions 0.1, 0.2, 0.3, 0.4, and 0.5 are entered as 0.1, 0.3, 0.5, 0.7, 0.9. The letter designations have become negative numbers with A=0, B=-1, etc. Thus a coordinate of NC = 31.30, EC = B.40 is entered as (30.5, -1.7). The master burial ground map, Figure 5, retains the letter designations, but the other maps show the corrected coordinate grid.

There are a few records that are inconsistent with this system: for example, the subdivision is given as .00 or .60. These have been interpreted as representing points on the coordinate boundary, although they may simply be errors.

Large Burials. Figure 1 shows all recorded burials of more than 5000 Ci (1/2 g) in the old burial grounds. A large fraction of these have been made in the "low level waste" areas at the west end of the burial ground; this is also the area in which the largest amounts of tritium have been found in the ground water. Other burials have been made in the "high level waste" section at the southeast, which is another region in which tritium is found in the ground water. There are a few burials in other areas. The correlation of the ground water tritium with these large burials is remarkably good.

Spent Melts. Nearly all the material identified as spent melts (either by designation, or by the observation that the buried material contains both tritium and induced radioactivity) is buried in the extreme eastern end of the old burial ground. The distribution shown in Figure 2 actually understates the fraction buried in this region, as most of the points in the eastern section represent several burials, while those scattered about elsewhere are generally single item burials.

Offsite Burials. This is primarily material received from Mound or Pinellas. Little is known about it other than an estimate of tritium content. Locations of these burials are shown in Figure 3.

234-H Burials. Burials of items believed to have a significant tritium content are shown in Figure 4. These include some identified material such as spent beds, mercury, etc., and a great deal of unspecified material. Although a substantial amount of this material is buried in the trenches on the northwest side of the region identified as SP-234, an even larger quantity is not.

These maps suggest an answer to the question of where waste generated between 1957 and 1961 might be buried. It is likely that it is in the same general region as

similar waste buried after that time. This conclusion rests on the observation that during the time in which records were kept, waste of a given type was buried in the same general area. Thus it is likely that nearly all spent melts are buried in the east end of the burial ground, probably in some of the areas that are incorporated in the "scrap metal" section but which show no tritium buried. Similarly, there are probably tritium burials in the "SP-234" trenches in parts that now show no tritium content, and other burials throughout the low level waste region in the west part of the burial grounds.

B. New Burial Grounds

Records for the new burial grounds are complete, and presentation of the data is straightforward. Burial locations are recorded on a decimal coordinate system. Figures 6 and 7 show the location of spent melt burials and of large burials (> 5 kCi per burial) respectively. There is one extremely large burial in the new burial ground; this is identified as the "special Mound burial" in Figure 7. This burial exceeded 1 MCi of tritium in a number of containers. It has been discussed in Ref. 10. Because this burial contained items considered highly secret, it was mingled with extremely radioactive material from other sources to make it difficult to retrieve.

Smaller amounts of tritium are buried widely through the new burial grounds, especially in the western section. Regions containing such burials are shown in Figure 8.

8. Buried Mercury.

Locations of mercury buried in the early years of operation in the old burial ground are shown in Figure 9. COBRA records do not show the amounts of mercury so buried.

Conclusions Regarding Buried Tritium

The COBRA records indicate that the large majority of buried tritium is associated with buried equipment and related waste from both onsite and offsite. An even higher fraction of the accessible tritium is of this type, as tritium in the interior of spent melts is essentially inaccessible to ground water. Thus it is not surprising to find that much of the tritium in the ground water is in areas where the tritium is associated with equipment and related waste. As it has been demonstrated that tritium is also leached from spent melts by ground water, this will also contribute to the ground water contamination.

COBRA data are a reasonable representation of the tritium that is likely to be present in the burial ground. Although a significant amount of tritium waste was buried before COBRA records were initiated, decay has reduced this to a small fraction of the total. Accountability records suggest that more tritium was buried than COBRA data would account for, but these records are of uncertain quality, and the differences from COBRA inventories may simply represent small errors in calculating production.

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Figure 1: Large T Burials, Old Burial Ground

Figure 2: Spent Melts in the Old BG

Figure 3: Tritium Burials from Offsite

Figure 4: Tritium from 234-H Operations

Figure 5

OLD BURIAL GROUND LAYOUT

Figure 6: New BG Spent Melts

Figure 7: Composite T Burials, New BG

**Figure 8: Other Tritium Burials,
New Burial Ground**

**Figure 9: Locations of Buried Mercury
Old Burial Ground**

Distribution:

M. J. Hitchler	992W-1
M. A. Ebra	992W-4
S. R. Salaymeh	992W-1
H. P. Holcomb	992W-4
D. Brett	992W-4
J. R. Cook	773-43A
E. L. Wilhite	773-43A
D. Wyatt	992W-4

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