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Title: RADIOLOGICAL AIR QUALITY IN A DEPLETED URANIUM
(DU) STORAGE VAULT

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RADIOLOGICAL AIR QUALITY IN A DEPLETED URANIUM STORAGE VAULT

Tom Robinson and Al Cucchiara

**Operational Health Physics
Los Alamos National Laboratory**

Abstract

The radiological air quality of two storage vaults, one with depleted uranium (DU) and one without, was evaluated and compared. The intent of the study was to determine if the presence of stored DU would significantly contribute to the gaseous/airborne radiation level compared to natural background.

Both vaults are constructed out of concrete and are dimensionally similar. The vaults are located on the first floor of the same building. Neither vault has air supply or air exhaust. The doors to both vaults remained closed during the evaluation period, except for brief and infrequent access by the operational group. One vault contained 700 KG of depleted uranium, and the other vault contained documents inside of file cabinets. Radon detectors and giraffe air samplers were used to gather data on the quantity of gaseous/airborne radionuclides in both vaults.

The results of this study indicated that there was no significant difference in the quantity of gaseous/airborne radionuclides in the two vaults. This paper gives a discussion of the effects of the stored DU on the air quality, and poses several theories supporting the results.

Introduction

DU is stored in various vaults and buildings at the Los Alamos National Laboratory. DU consists primarily of U-238 (ref 1). The decay scheme of U-238 is given in Figure 1. U-238 has eighteen daughter products, of which Rn-222 is an alpha-emitting gas. Rn-222 is also present in the environment as a naturally occurring gas formed from the decay of uranium in the earth.

The health effects of exposure to radon and radon progeny have been extensively studied, and, the carcinogenicity of radon is documented via epidemiological studies of underground miners. Because of the carcinogenicity identified with radon and its progeny, it is imperative to the health of those who enter into DU storage vaults to determine if the presence of DU contributes to the quantity of radon and its progeny in the vault air, thus the quantity inhaled.

The purpose of this experiment was to determine if the presence of DU in a closed environment significantly contributes to the presence of radon and radon progeny. In particular, the experimenters wanted to ascertain if Rn-222 was present in measurable quantities. An interesting aspect to this experiment is the presence of natural radon, as Rn-222 and as Rn-220, which would have to be taken into account when interpreting the results of any radon measuring devices.

Other experimenters have measured the quantity of radon in DU storage vaults, in particular, a study conducted by Milton McLain and Gerald Schlapper of Los Alamos (ref. 2). Studies, such as the one conducted by McLain, were based on the EPA Guidelines described in the EERF Standard Operating Procedure for Radon-222 Measurement Using Charcoal Canisters by David Gray and Sam Windam (ref 3). Drawbacks to this procedure include various potential variations such as the quantity of moisture in the atmosphere which can significantly reduce the amount of radon that can be absorbed into the charcoal. In addition, the EPA procedure does not require a background measurement to be made in a similar environment, without the DU, nor does it specify air flow restrictions, access by people, and other potential movement of air. These conditions could result in a false high reading, false low reading and a higher margin of error.

As described in the Materials and Methods Section, this experiment was conducted in a manner that should eliminate or minimize these potential sources of error.

Materials and Methods

Two concrete storage vaults on the first floor in the same building were selected for this study, both of similar dimensions (approximately 8'X20'X10' high). Neither vault has air supply or air exhaust, and the doors were kept closed during the study, except for several short term entries to check instruments and to record readings. There were no fans or other circulating devices in either vault. One vault had file cabinets containing documents, and was used as the control vault, and the other vault contained approximately seven hundred kilograms of DU in various containers. Most of the DU was wrapped in plastic, and was not airtight.

There were two phases to this experiment: 1) radon measurements and 2) measurements for total particulates. The radon measurements for both vaults were made utilizing a Femto Tech RS410F radon detector. The radon detector consists of an air ionization probe. The detector was used in a passive mode and averaged the radon level. The vault with the paper had the radon detector in place November 17 through November 20, 1997. The vault with the DU had the radon detector in place November 12 until November 17, 1997. The particulate measurements for both vaults were made utilizing a giraffe air sampler with a two inch diameter Hollingsworth & Vose Hovoglas Laminated Filter. The flow rate was two cubic feet per minute.

Procedure

The two different phases of this experiment were conducted as follows: 1) measure and compare the concentration of radon in both vaults, and 2) measure and compare the concentration of radionuclide particulates in both vaults.

The radon detector was read and recorded each day. The filters were approximately four feet above the floor. Nine samples were collected of 168 hours each in the DU vault, and two samples, one of 168 hours and one of 144 hours were collected in the control vault. The vault with the documents (control vault) was started on October 2, 1997 and terminated on October 15, 1997. The vault with the DU was started on July 30, 1997 and terminated on October 2, 1997. The filters were allowed to decay for a minimum of 24 hours. The filters were then counted for three minutes on a Berthold LB780 Alpha/Beta Counter, which is a low level, gas proportional counter.

Results

The results of the samples for the total particulate measurements for both vaults are given in Table 1.

Table 1, Total Alpha and Beta Measurements of Filtered Particulates

DU Vault, Sampling Period	Alpha (Bq/M ³)	Beta (Bq/M ³)	Control Vault, Sampling Period	Alpha (Bq/M ³)	Beta (Bq/M ³)
1	NDA	5.0E-4	10	NDA	6.7E-4
2	1.7E-4	6.7E-4	11	NDA	3.3E-4
3	1.7E-4	8.3E-4			
4	NDA	4.0E-4			
5	NDA	1.2E-3			
6	NDA	8.3E-4			
7	2.2E-3	4.4E-3*			
8	NDA	5.0E-4			
9	NDA	6.7E-4			
Average	2.8E-4	1.1E-3*		NDA	5.0E-4
+ or - (2 s)	3.7E-5	1.9E-4		-	1.7E-4

*Average without sampling period #7 is 7.1E-4+ or - 1.7E-4.

The results of the measurements, + or - 10%, for the average radon levels observed by the radon detector in the two vaults are 22.2 Bq/M³ for the DU Vault, and 18.5 Bq/M³ for the control vault. These readings were barely within the dynamic range of the instrument as reported by the manufacturer's specification sheet of 0.5 to 2000 pCi/l. (18.5 Bq/M³ to 7.6E4 Bq/M³).

Discussion

Particulates were sampled for in both vaults because both RN-222 and Rn-220 decay into nongaseous isotopes. In order to minimize interference by the presence of radon which may have adhered to dust particles and thus may have been collected on the filter paper, the filters were allowed to decay. It is noted that for sampling period number seven, there is a significantly higher level of radioactivity measured than for the other sampling periods. This is due to not allowing the sample to decay long enough to minimize interference from radon gases adhering to dust particles.

The results of this experiment show that the concentration of radionuclides measured on the filter paper for both the DU and control vaults are not significantly different. This is evidence that the presence of radon must not be significantly different in both vaults, because the nongaseous isotopes are daughters of radon, and the vaults were not ventilated.

The results also show that there is no significant difference in the quantity of radon measured by the radon detector for both vaults. Variations in the weather, especially precipitation, atmospheric stability and wind, result in significant variations in concentrations of Rn-222. The concentration of radon inside of buildings; however, largely depends on the radium content and porosity of the building material and the degree of ventilation of the building or room in which the measurements are being made (ref. 4). Because the control vault is constructed similarly to the DU vault, the porosity and radium content of both vaults is the same. Additionally, neither vault was ventilated. Deviations in the quantities of radioactivity in the soil is a geological variable (ref. 4). This variation is minimized in this experiment because the vaults are within a distance of 50 yards. Any difference in the quantity of radon, i.e. Rn-220 plus Rn-222, we theorize would be due to an increase in the quantity of Rn-222 in the DU vault, since Rn-222 is a daughter of U-238.

When uranium is refined, all of the daughter products are essentially removed. Uranium had not been refined and separated in large quantities until the early 1940s. This would mean that depleted uranium is, at the most, between 50 and 60 years old. Because of the length of the halflife of uranium daughters, U-234, Th-230 and Rad-226, there has not been ample time for the Rn-222 to be produced. Based on the decay of these three daughters, one Ci (3.7×10^{10} Bq) of U-238 would produce an inventory of 2.31×10^{-10} Ci (8.55 Bq) of Rn-222 after 50 years (ref. 5). Based on the specific activity of U-238, which is 0.33 $\mu\text{Ci/g}$, and the total of 700 Kg of U-238 in the vault, there is a total of 0.231 Ci of U-238 in the vault. This would produce an inventory of 5.31×10^{-11} Ci (2.0 Bq) of Rn-222 after 50 years. If the total quantity of Rn-222 were available to the volume in the vault, there would be a concentration 4.4×10^{-2} Bq/ M^3 of Rn-222 dispersed in the air. This extremely small amount of Rn-222 would not be detected by the radon detector since the lower range of the radon detector is 18.5 Bq/ M^3 .

Due the various shapes of the DU in the vault, and the thinnest dimension being approximately one cm, it is safe to assume that the majority of the small amount of Rn-222 molecules that are produced is trapped within the matrixes of the DU metal as they are formed. In addition, the majority of radon daughters would also be trapped. Since the radon daughters are solids, the portion that is not trapped within the matrixes are adhered to the surface, lacking a mechanism to become airborne.

Conclusion

We conclude, from the results of this experiment, that the presence of stored DU did not result in a significant increase in gaseous/airborne radionuclide concentrations in the vault.

References

- 1) Health Physics Manual of Good Practices for Uranium Facilities, US DOE, Idaho National Engineering Lab., EG&G Idaho, Inc., Bryce Rich, Chairperson, 1988.
- 2) Los Alamos National Laboratory Memorandum ESH-12:95-986, "Radon Characterization, Storage Magazine," McLain, M., Schlapper, G., August 14, 1995.
- 3) Environmental Protection Agency, EERF Standard Operating Procedures for Rn-222 Measurement Using Charcoal Canisters, EPA520/5-87-005.
- 4) National Council on Radiation Protection and Measurements, Environmental Radiation Measurements, NCRP Report No. 50, 1976.
- 5) Los Alamos National Laboratory E-Mail, Excel Spreadsheet with U-238 Decay Products, Smale, R.F., Nov 13, 1998.

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Real Time Radon Stack Monitoring

Radon Generation

Radon can pose a health hazard in industrial settings. Mining operations (in particular, for uranium or thorium) often involve high levels of radon in tunnels. Handling of ores or other uranium process material may free substantial amounts of radon gas into the work environment. Radon (or more specifically, the isotope Rn-222) and thoron (the radon isotope Rn-220) are produced in the natural decay of natural uranium (mostly U-238) and thorium (Th-232) respectively. The two gases decay (with quite different half-lives) into non-gaseous isotopes. The daughter isotopes decay through both alpha-particle and beta-particle emission. It is the decays of the daughters which poses the greatest health hazard when the daughters have been deposited in the lungs.

The decay chains of radon and thoron.

Real-time Radon Monitoring

Both radon and thoron as well as several of their daughters, emit alpha particles upon decay. Monitoring for ionization in the air (often termed the Long-Range Alpha Detection technique) is ideally suited to detecting the radon or thoron in real time (that is, with immediate response).

Portable radon monitors are being developed by Los Alamos and industrial partners for both home use and spot usage in other facilities. In some cases, however, it is better to sample the air exhausted by a ventilation system. A radon stack monitor detects radon in the air passing through an emissions stack. A

U-238 Decay

1.0 Curie Initial Activity

FOR TIME T =	.01 YEARS	.02 YEARS	.03 YEARS	.05 YEARS	.07 YEARS
ACTIVITY OF U -238 IS:	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00
ACTIVITY OF Th-234 IS:	9.972271E-02	1.895008E-01	2.703260E-01	4.085998E-01	5.206706E-01
ACTIVITY OF Pa-234m IS:	9.972607E-02	1.895072E-01	2.703351E-01	4.086136E-01	5.206882E-01
ACTIVITY OF Pa-234 IS:	1.614317E-04	3.067650E-04	4.376052E-04	6.614437E-04	8.428645E-04
ACTIVITY OF U -234 IS:	1.432510E-09	5.537894E-09	1.204961E-08	3.135609E-08	5.770530E-08
ACTIVITY OF Th-230 IS:	4.428988E-17	3.453460E-16	1.136480E-15	5.008213E-15	1.310082E-14
ACTIVITY OF Ra-226 IS:	4.821753E-23	7.557686E-22	3.749221E-21	2.780169E-20	1.027508E-19
ACTIVITY OF Ra-222 IS:	5.760787E-24	1.641101E-22	1.117000E-21	1.174812E-20	5.272640E-20
ACTIVITY OF Po-218 IS:	5.764032E-24	1.642025E-22	1.117629E-21	1.175473E-20	5.275610E-20
ACTIVITY OF Pb-214 IS:	5.791068E-24	1.649727E-22	1.122872E-21	1.180987E-20	5.300355E-20
ACTIVITY OF Bi-214 IS:	1.376098E-27	3.920152E-26	2.668216E-25	2.806311E-24	1.259493E-23
ACTIVITY OF Po-214 IS:	1.375699E-27	3.919015E-26	2.667442E-25	2.805498E-24	1.259128E-23
ACTIVITY OF Pb-210 IS:	3.063948E-28	1.773305E-26	1.836468E-25	3.300171E-24	2.117363E-23
ACTIVITY OF Bi-210 IS:	2.104544E-29	2.320717E-27	3.437146E-26	9.384695E-25	7.717668E-24
ACTIVITY OF Po-210 IS:	4.886161E-32	1.092721E-29	2.459033E-28	1.144867E-26	1.343967E-25
ACTIVITY OF At-218 IS:	1.152812E-27	3.284067E-26	2.235270E-25	2.350958E-24	1.055127E-23
ACTIVITY OF Tl-210 IS:	2.890489E-30	8.234264E-29	5.604576E-28	5.894646E-27	2.645560E-26
ACTIVITY OF Tl-206 IS:	2.779615E-35	3.065129E-33	4.539673E-32	1.239501E-30	1.019325E-29
Atoms of Stable Pb-206					
Mass of Stable Pb-206					

U-238 Decay

1.0 Curie Initial Activity

	.1 YEARS	.2 YEARS	.3 YEARS	.5 YEARS	.7 YEARS	1 YEARS
U-238	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00
	6.502458E-01	8.776720E-01	9.572153E-01	9.947662E-01	9.993598E-01	9.999726E-01
	6.502677E-01	8.777016E-01	9.572475E-01	9.947998E-01	9.993935E-01	1.000006E+00
	1.052622E-03	1.420780E-03	1.549545E-03	1.610333E-03	1.617769E-03	1.618761E-03
	1.075853E-07	3.288144E-07	5.897908E-07	1.144409E-06	1.707885E-06	2.554786E-06
	3.564754E-14	2.314579E-13	6.521131E-13	2.243739E-12	4.866152E-12	1.074556E-11
	4.046073E-19	5.452199E-18	2.372291E-17	1.417965E-16	4.423021E-16	1.431374E-15
Rn-222	4.464855E-19	4.203551E-18	1.995568E-17	1.281189E-16	4.119810E-16	1.363362E-15
	2.466244E-19	4.205919E-18	1.996692E-17	1.281911E-16	4.122131E-16	1.364131E-15
	2.477811E-19	4.225647E-18	2.006057E-17	1.287924E-16	4.141465E-16	1.370529E-15
	5.887881E-23	1.004116E-21	4.766879E-21	3.060419E-20	9.841127E-20	3.256709E-19
	5.886174E-23	1.003825E-21	4.765496E-21	3.059532E-20	9.838274E-20	3.255765E-19
	1.450977E-22	5.243818E-21	3.871957E-20	4.334356E-19	2.005488E-18	9.722716E-18
	6.674684E-23	3.412850E-21	2.893005E-20	3.642185E-19	1.773864E-18	8.934551E-18
	1.700511E-24	1.824154E-22	2.366379E-21	4.982457E-20	3.335336E-19	2.296087E-18
	4.932511E-23	8.411879E-22	3.993403E-21	2.563834E-20	8.244302E-20	2.728274E-19
	1.236747E-25	2.109142E-24	1.001281E-23	6.428398E-23	2.067125E-22	6.840705E-22
	8.815710E-29	4.507584E-27	3.820990E-26	4.810482E-25	2.342863E-24	1.180047E-23

U-238 Decay 1.0 Curie Initial Activity

	2 YEARS	3 YEARS	5 YEARS	7 YEARS	10 YEARS	20 YEARS
^{238}U	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00
	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00
	1.000034E+00	1.000034E+00	1.000034E+00	1.000034E+00	1.000034E+00	1.000034E+00
	1.618806E-03	1.618806E-03	1.618806E-03	1.618806E-03	1.618806E-03	1.618806E-03
	5.378326E-06	8.201865E-06	1.384892E-05	1.949594E-05	2.796641E-05	5.620081E-05
	4.721925E-11	1.096560E-10	3.124175E-10	6.190276E-10	1.273653E-09	5.143129E-09
	1.304717E-14	4.607855E-14	2.213265E-13	6.170023E-13	1.820118E-12	1.475239E-11
^{234}Th	1.274317E-14	4.536884E-14	2.192965E-13	6.129740E-13	1.811822E-12	1.471889E-11
	1.275035E-14	4.539440E-14	2.194201E-13	6.133193E-13	1.812843E-12	1.472718E-11
	1.281015E-14	4.560731E-14	2.204492E-13	6.161960E-13	1.821346E-12	1.479626E-11
	3.044003E-18	1.083741E-17	5.238409E-17	1.464231E-16	4.327959E-16	3.515950E-15
	3.043120E-18	1.083426E-17	5.236890E-17	1.463807E-16	4.326704E-16	3.514930E-15
	1.879192E-16	1.012905E-15	8.168112E-15	3.176619E-14	1.323432E-13	2.039416E-12
	1.803591E-16	9.859072E-16	8.038515E-15	3.140812E-14	1.313056E-13	2.031553E-12
	7.737524E-17	5.333876E-16	5.417086E-15	2.351106E-14	1.068381E-13	1.830728E-12
	2.550082E-18	9.078923E-18	4.388423E-17	1.226645E-16	3.625703E-16	2.945450E-15
	6.393916E-21	2.276393E-20	1.100326E-19	3.075612E-19	9.090861E-19	7.385238E-18
	2.382125E-22	1.302155E-21	1.061702E-20	4.148285E-20	1.734243E-19	2.683211E-18
	30	325	5745	35782	237644	8446440
	1.04E-20	1.11E-19	1.97E-18	1.22E-17	8.13E-17	2.89E-15

*I have on the three
radiation type as per in the
value of Beta to be alpha on our instrument for a then decay.*

U-238 Decay

1.0 Curie Initial Activity

	30 YEARS	50 YEARS	70 YEARS	100 YEARS	200 YEARS	300 YEARS	500 YEARS
α U-238	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00	9.999999E-01
Th-234	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00	1.000000E+00	9.999999E-01
β Pa-234	1.000034E+00	1.000034E+00	1.000034E+00	1.000034E+00	1.000000E+00	1.000000E+00	9.999999E-01
	1.618806E-03	1.618806E-03	1.618806E-03	1.618806E-03	1.600000E-03	1.600000E-03	1.600000E-03
	8.443440E-05	1.408992E-04	1.973608E-04	2.820473E-04	5.645226E-04	8.466644E-04	1.410709E-03
	1.160835E-08	3.232478E-08	6.342046E-08	1.295196E-07	5.188303E-07	1.166901E-06	3.238796E-06
	4.997033E-11	2.317093E-10	6.354362E-10	1.848699E-09	1.466828E-08	4.896583E-08	2.218236E-07
β Rn-222	4.989479E-11	2.314995E-10	6.350256E-10	1.847864E-09	1.466837E-08	4.896615E-08	2.218251E-07
	4.992290E-11	2.316299E-10	6.353833E-10	1.848905E-09	1.466837E-08	4.896615E-08	2.218251E-07
	5.015706E-11	2.327163E-10	6.383635E-10	1.857577E-09	1.466544E-08	4.895636E-08	2.217807E-07
	1.191853E-14	5.529905E-14	1.516907E-13	4.414053E-13	1.466837E-08	4.896615E-08	2.218251E-07
	1.191508E-14	5.528301E-14	1.516467E-13	4.412773E-13	1.466531E-08	4.895592E-08	2.217787E-07
	9.823083E-12	6.845015E-11	2.385325E-10	8.677542E-10	9.522336E-09	3.632151E-08	1.846892E-07
	9.798190E-12	6.834862E-11	2.382852E-10	8.671410E-10	9.528200E-09	3.634388E-08	1.848030E-07
	9.145070E-12	6.562709E-11	2.315937E-10	8.504429E-10	9.441251E-09	3.612973E-08	1.841729E-07
	9.984628E-15	4.632620E-14	1.270773E-13	3.697827E-13	2.933674E-12	9.793230E-12	4.436501E-11
	2.503483E-17	1.161554E-16	3.186256E-16	9.271701E-16	3.080358E-12	1.028289E-11	4.658327E-11
	1.294114E-17	9.027269E-17	3.147195E-16	1.145292E-15	1.257723E-14	4.797393E-14	2.439400E-13
	64500496	790813831	3.98E+09	2.13E+10	4.97E+11	2.93E+12	2.57E+13
	2.21E-14	2.70E-13	1.36E-12	7.29E-12	1.70E-10	1.00E-09	8.79E-09

1/10/99

4:54 PM

U-238 Decay

1.0 Curie Initial Activity

700 YEARS	1000 YEARS	2000 YEARS	3000 YEARS	5000 YEARS	7000 YEARS	10,000 YEARS
9.999999E-01	9.999998E-01	9.999997E-01	9.999995E-01	9.999992E-01	9.999989E-01	9.999984E-01
9.999999E-01	9.999998E-01	9.999997E-01	9.999995E-01	9.999992E-01	9.999989E-01	9.999984E-01
9.999999E-01	9.999998E-01	9.999997E-01	9.999995E-01	9.999992E-01	9.999989E-01	9.999984E-01
1.600000E-03	1.600000E-03	1.600000E-03	1.599999E-03	1.599999E-03	1.599998E-03	1.599998E-03
1.974435E-03	2.819428E-03	5.630906E-03	8.434457E-03	1.401787E-02	1.956984E-02	2.783922E-02
6.342958E-06	1.292927E-05	5.151061E-05	1.154365E-04	3.181074E-04	6.185420E-04	1.247367E-03
5.957665E-07	1.682758E-06	1.216255E-05	3.730723E-05	1.449742E-04	3.401844E-04	8.071258E-04
5.957704E-07	1.682769E-06	1.216263E-05	3.730748E-05	1.449751E-04	3.401866E-04	8.071311E-04
5.957704E-07	1.682769E-06	1.216263E-05	3.730748E-05	1.449751E-04	3.401866E-04	8.071311E-04
5.956513E-07	1.682432E-06	1.216019E-05	3.730001E-05	1.449461E-04	3.401186E-04	8.069697E-04
5.957704E-07	1.682769E-06	1.216263E-05	3.730748E-05	1.449751E-04	3.401866E-04	8.071312E-04
5.956459E-07	1.682417E-06	1.216008E-05	3.729968E-05	1.449448E-04	3.401155E-04	8.069625E-04
5.223895E-07	1.535165E-06	1.162934E-05	3.623733E-05	1.425839E-04	3.363288E-04	8.010141E-04
5.227112E-07	1.536111E-06	1.163650E-05	3.625965E-05	1.426718E-04	3.365359E-04	8.015074E-04
5.214651E-07	1.533602E-06	1.162029E-05	3.621916E-05	1.425433E-04	3.362633E-04	8.009103E-04
1.191541E-10	3.365538E-10	2.432525E-09	7.461495E-09	2.899502E-08	6.803732E-08	1.614262E-07
1.251118E-10	3.533815E-10	2.554152E-09	7.834570E-09	3.044478E-08	7.143919E-08	1.694975E-07
6.899791E-13	2.027667E-12	1.536019E-11	4.786275E-11	1.883268E-10	4.442276E-10	1.057990E-09
1.04E+14	4.43E+14					
3.55E-08	1.52E-07			3.93E-05	2.24E-04	8.85E-04