U.S. DEPARTMENT OF ENERGY
OFFICE OF CIVILIAN RADIOACTIVE WASTE MANAGEMENT

PRESENTATION TO THE NUCLEAR WASTE TECHNICAL REVIEW BOARD

SUBJECT: GASEOUS AND SEMI-VOLATILE RADIONUCLIDES

PRESENTER: DR. U-SUN PARK

PRESENTER'S TITLE AND ORGANIZATION: SENIOR STAFF ENGINEER
SCIENCE APPLICATIONS INTERNATIONAL CORPORATION
LAS VEGAS, NEVADA

PRESENTER'S TELEPHONE NUMBER: (702) 794-7643

REGISTRY HOTEL, DENVER, COLORADO
JUNE 25-27, 1991

U.S. DEPARTMENT OF ENERGY
OFFICE OF CIVILIAN RADIOACTIVE WASTE MANAGEMENT

PRESENTATION TO THE NUCLEAR WASTE TECHNICAL REVIEW BOARD

SUBJECT: GASEOUS AND SEMI-VOLATILE RADIONUCLIDES

PRESENTER: DR. U-SUN PARK

PRESENTER'S TITLE AND ORGANIZATION: SENIOR STAFF ENGINEER
SCIENCE APPLICATIONS INTERNATIONAL CORPORATION
LAS VEGAS, NEVADA

PRESENTER'S TELEPHONE NUMBER: (702) 794-7643

REGISTRY HOTEL, DENVER, COLORADO
JUNE 25-27, 1991

WHY GASEOUS AND SEMI-VOLATILE RADIONUCLIDES ARE IMPORTANT

- YUCCA MOUNTAIN SITE IS IN THE UNSATURATED ZONE

- THE EPA AND NRC REGULATIONS DID NOT ADEQUATELY CONSIDER THE RELEASE OF GASEOUS RADIONUCLIDES

- TEST PRIORITIZATION TASK FORCE REPORT (MARCH, 1991) ASSUMES THAT COMPLIANCE WITH THE EPA AND NRC REGULATIONS RATHER THAN THE PUBLIC HEALTH AND SAFETY CONSIDERATIONS DICTATE THE PRIORITY OF TESTS TO BE PERFORMED

OBJECTIVES OF GASEOUS RADIONUCLIDE STUDIES

- TO ASSESS THE RELATIVE IMPORTANCE OF RELEASE OF GASEOUS AND SEMI-VOLATILE RADIONUCLIDES FROM ENGINEERED BARRIER SYSTEM TO THE NATURAL BARRIER AND ACCESSIBLE ENVIRONMENT

- TO IDENTIFY DATA NEEDS AND DEVELOP STUDY PLANS TO MODEL AND ASSESS THE EFFECT OF RELEASE OF GASEOUS AND SEMI-VOLATILE RADIONUCLIDES

- TO PROVIDE INPUT TO TEST PLANS, TEST PRIORITIZATION EVALUATION AND PERFORMANCE ASSESSMENT

PRESENTATION OUTLINE

- REVIEW OF GASEOUS AND SEMI-VOLATILE RADIONUCLIDES IN SPENT FUEL
- RELEASE POTENTIAL
- DATA NEEDS AND TEST PLANS
- CONCLUSIONS

REVIEW OF GASEOUS AND SEMI-VOLATILE RADIONUCLIDES IN SPENT FUEL

GASEOUS AND SEMI-VOLATILE RADIONUCLIDES IN SPENT FUEL THAT COULD POTENTIALLY UNDERGO GASEOUS TRANSPORT AT REPOSITORY TEMPERATURES

GASEOUS RADIONUCLIDES	HALF-LIFE	
^{3}H	12.3	y
*^{14}C	5,730	y
^{85}Kr	10.7	y
*^{129}I	1.57×10^{7}	y
^{222}Rn	3.82	d

SEMI-VOLATILE RADIONUCLIDES	HALF-LIFE	
*^{79}Se	$\leq 6.5 \times 10^{4}$	y
*^{99}Tc	2.14×10^{5}	y
*^{135}Cs	2.95×10^{6}	y
^{137}Cs	30.2	y

*NUCLIDES OF SUFFICIENTLY LONG HALF-LIFE TO BE PRESENT IN SIGNIFICANT AMOUNTS AFTER THE 300 TO 1000 YEAR CONTAINMENT PERIOD

COMPARISON OF INVENTORY TO CURRENT EPA 10,000-YEAR CUMULATIVE RELEASE LIMIT AT ACCESSIBLE ENVIRONMENT AND NRC 10CFR60.113 MAXIMUM RELEASE RATES FROM THE ENGINEERED BARRIER SYSTEM*

GASEOUS RADIONUCLIDES	INVENTORY AT 1,000 YEARS (Ci)	EPA 10,000-YEAR CUMULATIVE RELEASE LIMIT, Ci (ANNUAL AVG. Ci/YR)	NRC POST-CONTAINMENT PERIOD RELEASE LIMIT FROM EBS (Ci/YR)
^{14}C	62,000	6,200 (0.62)	**1.07
^{129}I	1,950	6,200 (0.62)	**1.07
SEMI-VOLATILE RADIONUCLIDES			
^{79}Se	25,050	62,000 (6.2)	**1.07
^{99}Tc	806,000	620,000 (6.2)	8.06
^{135}Cs	21,390	62,000 (6.2)	**1.07

*BASED ON 62,000 MTIHM SPENT FUEL
**NUCLIDES FOR WHICH THE MAXIMUM RELEASE RATE IS GREATER THAN 1 X 10^{-5} PER YEAR INVENTORY BECAUSE OF THEIR SMALL INVENTORIES

RELEASE POTENTIAL

- RELEASE UNDER UNDISTURBED CONDITIONS IS INFLUENCED BY THE NEAR-FIELD ENVIRONMENT
- RELEASE BY DISTURBED SCENARIOS IS LIMITED TO A SMALL NUMBER OF WASTE PACKAGES
- RELEASE DUE TO DEFECTIVE WASTE PACAKGES IS LIMITED TO A SMALL, BUT FINITE NUMBER

WASTE PACKAGE EMPLACEMENT ENVIRONMENT

- THE REPOSITORY WILL BE LOCATED IN AN UNSATURATED ZONE

- THE WASTE PACKAGES WILL SEE THE PEAK TEMPERATURE LESS THAN 35 YEARS AFTER REPOSITORY EMPLACEMENT; THE TEMPERATURE IN THE EMPLACEMENT ENVIRONMENT DROPS SIGNIFICANTLY DURING THE FIRST 300 YEARS AND SLOWLY THEREAFTER

- THE TIME PERIOD OF 0 TO 300 YEARS WHEN THE RELEASE POTENTIAL OF GASEOUS AND SEMI-VOLATILE RADIONUCLIDES IS HIGH COINCIDES WITH THE PERIOD WHEN MOST OF THE WASTE PACKAGES ARE EXPECTED TO REMAIN INTACT (REFERENCE CONCEPTUAL DESIGN)

- MOST LIKELY ENVIRONMENT IN WHICH THE GASEOUS AND SEMI-VOLATILE RADIONUCLIDES ARE RELEASED IS BELOW 200^0 C (SEE ATTACHMENT 2)

WOULD THESE POTENTIALLY GASEOUS RADIONUCLIDES ACTUALLY BE PRESENT IN THEIR VOLATILE FORMS ?

NUCLIDE	PROBABLE LOCATION & FORM IN SPENT FUEL	HIGH VAPOR PRESSURE FORM	COULD HIGH VAPOR PRESSURE FORM BE PRESENT UNDER OXIDIZING CONDITIONS?
^{14}C	FUEL ROD SURFACES, BULK CLAD, BULK UO_2. ELEMENT, CARBIDE	CO_2	YES
^{79}Se	BULK UO_2	SeO_2	YES
^{99}Tc	BULK UO_2	Tc_2O_7	YES
^{129}I	FUEL-CLAD GAP AND BULK UO_2. CsI	I_2	ONLY IN SMALL AMOUNTS
^{135}Cs	FUEL-CLAD GAP & BULK UO_2. Cs_2O, Cs_2UO_4, Cs_2MoO_4, CsI, Cs	Cs	NO

SUMMARY - ^{135}Cs WOULD NOT. ^{129}I COULD BE TO ONLY A SMALL EXTENT. THE OTHERS COULD BE, WHEN THEY ESCAPE THE SPENT FUEL

VAPOR PRESSURES

SPECIES	VAPOR PRESSURE (ATMOSPHERES)	
	100°C	200°C
CO_2	> 2,000	> 12,000
I_2	6×10^{-2}	3.7
SeO_2	9.1×10^{-4}	5.4×10^{-2}
Tc_2O_7	1.2×10^{-4}	3.7×10^{-2}

(FROM LANGE'S HANDBOOK OF CHEMISTRY, 13TH EDITION, 1985)

RELEASE POTENTIAL OF GASEOUS AND SEMI-VOLATILE RADIONUCLIDES

- POTENTIAL GASEOUS RELEASE OF $^{14}CO_2$ FROM THE REFERENCE CONCEPTUAL DESIGN WASTE PACKAGE WOULD LIKELY EXCEED THE CURRENT NRC AND EPA RELEASE LIMITS. DOE IS CURRENTLY CONSIDERING WHAT APPROACH TO TAKE ON WASTE PACKAGE STRATEGY

- ^{129}I WOULD LIKELY UNDERGO SOME GASEOUS RELEASE FROM WASTE PACKAGES, BUT THE INVENTORY WILL BE LESS THAN THE CURRENT EPA 10,000-YEAR CUMULATIVE RELEASE LIMIT

- GASEOUS RELEASE OF ^{99}Tc AND ^{79}Se WOULD LIKELY BE LESS IMPORTANT THAN ^{14}C OR ^{129}I BECAUSE OF LOWER PURE-PHASE VAPOR PRESSURES AND DILUTION IN THE SOLID PHASES IN SPENT FUEL

- ^{135}Cs WOULD NOT BE PRESENT IN THE VOLATILE ELEMENTAL FORM IN ANY SIGNIFICANT AMOUNT

DATA NEEDS AND TEST PLANS

QUESTION # 1

CAN WE RELEASE GASEOUS RADIONUCLIDES C-14 AND I-129 WITHOUT VIOLATING THE EPA AND NRC REGULATIONS?

ANSWER #1

PROBABLY NOT FOR THE REFERENCE CONCEPTUAL DESIGN. THE INVENTORY AND RELEASE POTENTIAL FOR BOTH RADIONUCLIDES ARE TOO HIGH. FURTHER DATA AND ANALYSES MAY BE NEEDED.

QUESTION # 2

CAN WE SAFELY DISMISS THE RELEASE OF SEMI-VOLATILE RADIONUCLIDES WITHOUT VIOLATING THE EPA AND NRC REGULATIONS?

ANSWER #2

WE CAN DISMISS THE RELEASES AS RADIOLOGICALLY INSIGNIFICANT; HOWEVER, THEY MAY EXCEED CURRENT EPA AND NRC REGULATIONS.

RELEASE MECHANISMS AND DATA NEEDS

- WASTE CONTAINER AND CLADDING FAIL UNDER UNDISTURBED AND DISTURBED CONDITIONS
- C-14 IS RELEASED FROM CLADDING SURFACE AND SPENT FUEL MATRIX
- GASEOUS AND SEMI-VOLATILE RADIONUCLIDES ARE RELEASED FROM FUEL MATRIX AND GAP/GRAIN BOUNDARIES
- FURTHER RELEASE FOLLOWS WITH FUEL OXIDATION
- GASEOUS AND VAPOR TRANSPORT THROUGH THE NEAR- AND FAR-FIELD FOLLOWS VIA DIFFUSION AND ADVECTION
- TRANSPORT MAY BE RETARDED BY VAPOR CONDENSATION, SORPTION, GASEOUS ISOTOPIC EXCHANGE AND GEOCHEMICAL REACTIONS WITH PORE WATER AND HOST ROCK

SPENT FUEL WASTE FORM

DATA NEEDS:

- INVENTORY OF GASEOUS AND SEMI-VOLATILE RADIONUCLIDES
- DISTRIBUTION IN MATRIX, GAP/GRAIN BOUNDARY AND CLADDING
- RELEASE RATE OF GASEOUS RADIONUCLIDES
- VAPOR PRESSURES OF SEMI-VOLATILES IN OXIDIZING CONDITION
- SPENT FUEL OXIDATION RATE

TEST PLANS:

WBS 1.2.2.3.1.1
SPENT FUEL WASTE FORM TESTING

WBS 1.2.2.2
WASTE PACKAGE ENVIRONMENT

WASTE CONTAINER

DATA NEEDS:

- CONTAINER BREACH RATE
- CLADDING BREACH RATE

TEST PLANS:

WBS 1.2.2.3.2
WASTE PACKAGE METAL BARRIER TESTING

WBS 1.2.2.2
WASTE PACKAGE ENVIRONMENT

RELEASE AND TRANSPORT MODELING

MODEL INPUT:

- CONVECTIVE GAS (AIR) FLOW BY THERMAL BUOYANCY, EXPANSION AND BREATHING OF THE MOUNTAIN
- DIFFUSION OF GASEOUS AND SEMI-VOLATILE RADIONUCLIDES THROUGH BREACHED CONTAINER
- GASEOUS TRANSPORT MODELING

STUDY PLANS:

8.3.1.2.2.6
CHARACTERIZATION OF GASEOUS PHASE MOVEMENT IN THE UNSATURATED ZONE

WBS 1.2.1.4.2
WASTE PACKAGE PERFORMANCE ASSESSMENT

WBS 1.2.1.4.7
DEVELOPMENT AND VALIDATION OF FLOW AND TRANSPORT MODELS

WBS 1.2.1.4.9
DEVELOPMENT AND VERIFICATION OF FLOW AND TRANSPORT CODES

RETARDATION DATA NEEDS

DATA NEEDS:

- ANALYSIS OF RETARDATION MECHANISM
- KINETIC EQUILIBRIUM CONSTANTS OF DISSOLVED H_2CO_3, HCO_3^- AND CO_3^{-2} TO GASEOUS CO_2
- RETARDATION MAY BE CONSERVATIVELY IGNORED FOR ^{129}I AND SEMI-VOLATILE RADIONUCLIDES AND USE BOUNDING CALCULATIONS

STUDY PLANS:

8.3.1.2.2.7
HYDROCHEMICAL CHARACTERIZATION
OF THE UNSATURATED ZONE

CONCLUSIONS

CONCLUSIONS

- C-14 IS THE MOST SIGNIFICANT GASEOUS RADIONUCLIDE FROM A REGULATORY COMPLIANCE (BUT NOT HEALTH AND SAFETY) POINT OF VIEW UNDER THE CURRENT EPA AND NRC REGULATIONS. THE DOE IS CURRENTLY CONSIDERING ALTERNATIVE STRATEGIES TO RESOLVE THE C-14 ISSUE

- AMOUNT OF RELEASE AND THE RESULTING HEALTH EFFECTS TO THE POPULATION FROM THE GASEOUS AND SEMI-VOLATILE RADIONUCLIDES ARE EXPECTED TO BE INSIGNIFICANT

CONCLUSIONS

(CONTINUED)

- THE CURRENT REGULATIONS FOR THE RELEASE OF GASEOUS RADIONUCLIDES FROM A GEOLOGIC REPOSITORY ARE TOO RESTRICTIVE. A REGULATORY RELIEF THROUGH THE REPROMULGATION OF THE 40CFR191 RELEASE LIMIT IN TABLE 1 OF THE APPENDIX WOULD BE THE MOST COST-EFFECTIVE WAY TO AVOID COSTLY SOLUTIONS THAT PROVIDE NO MEASURABLE BENEFITS TO THE HEALTH AND SAFETY OF THE PUBLIC

RELATIVE PERSPECTIVES IN SAFE RELEASE OF GASEOUS RADIONUCLIDES FROM NUCLEAR INDUSTRY*

	POWER REACTORS** (1980-1984) (Ci/GWeY)	(Ci/GWeY) FUEL PROCESSING PLANT M (1980 - 1985) U.K.	LaHAGUE	MARCOULE
^3H (AIRBORNE)	160 PWR 92 BWR 146 GCR	3,240	86	1,535
^3H (LIQUID)	730 PWR 57 BWR 260 GCR	15,650	7,720	7,950
^{14}C	9.3 PWR 8.9 BWR 35 LWGR	95		
^{131}I	0.05 PWR 0.25 BWR 0.04 GCR 2.16 LWGR	0.5		
^{131}I	0.77 PWR (U.S. 1982) 0.09 BWR (U.S. 1982)			
$^{131-135}I$	1.25 PWR 0.14 BWR			

*SOURCES, EFFECTS AND RISKS OF IONIZING RADIATION, UNITED NATIONS SCIENTIFIC COMMITTEE ON THE EFFECTS OF ATOMIC RADIATION (UNSCEAR) 1988 REPORT

**ELECTRICITY GENERATED BY NUCLEAR POWER IN 1987 = 189 GWeY

Attachment 1b

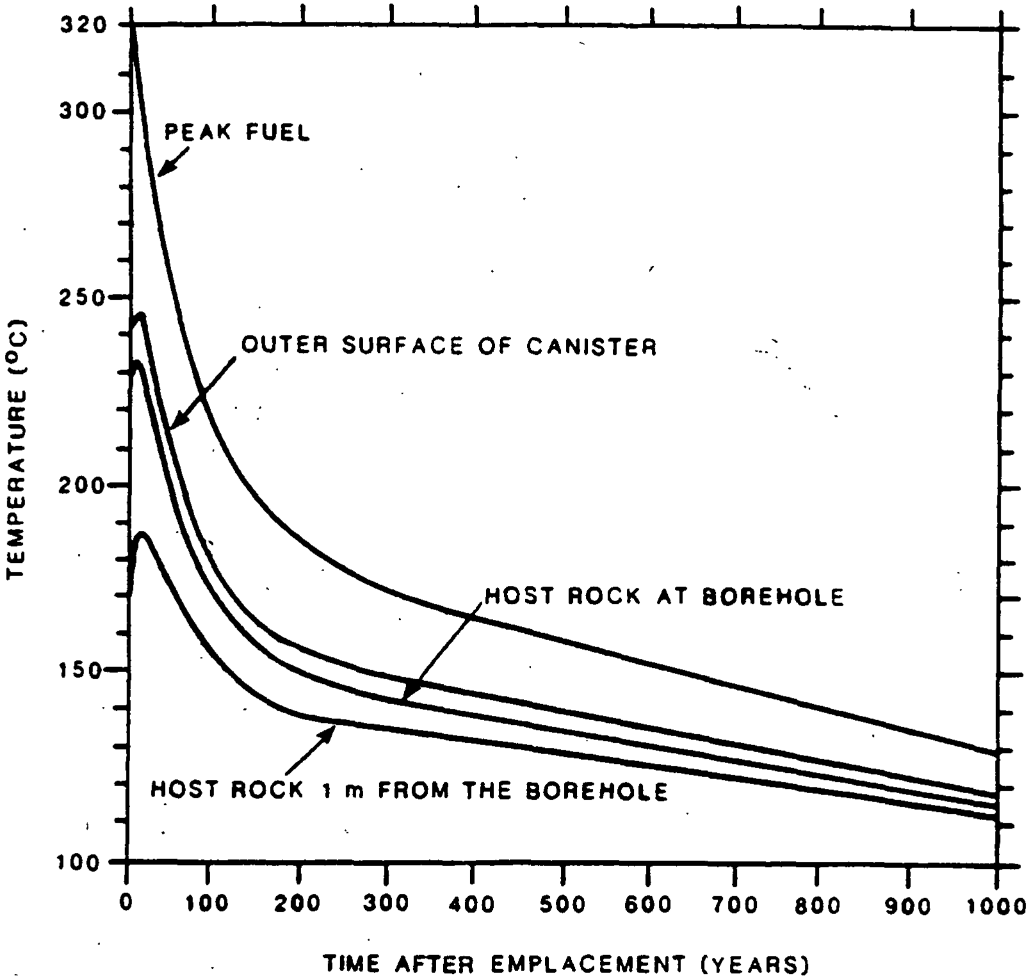
RELATIVE PERSPECTIVES IN SAFE RELEASE OF SEMI-VOLATILE RADIONUCLIDES FROM NUCLEAR INDUSTRY*

(Ci/GWeY)

	PWR	BWR	GCR	FRP U.K.	FRP FRANCE
^{99}Tc	0.002	0.016	-	127.1	105
$^{103,106}Ru$	0.012	0.004	1.16	0.3 (PARTICULATES) 3,760 (LIQUID)	2,680
$^{134,136,137}Cs$	0.77	0.55	2.11	1.0 (PARTICULATES) 15,320 (LIQUID)	230
^{129}I				0.1 (PARTICULATES)	0.13
PARTICULATES	0.12	1.17	0.04		

*SOURCES, EFFECTS AND RISKS OF IONIZING RADIATION, UNSCEAR 1988 REPORT

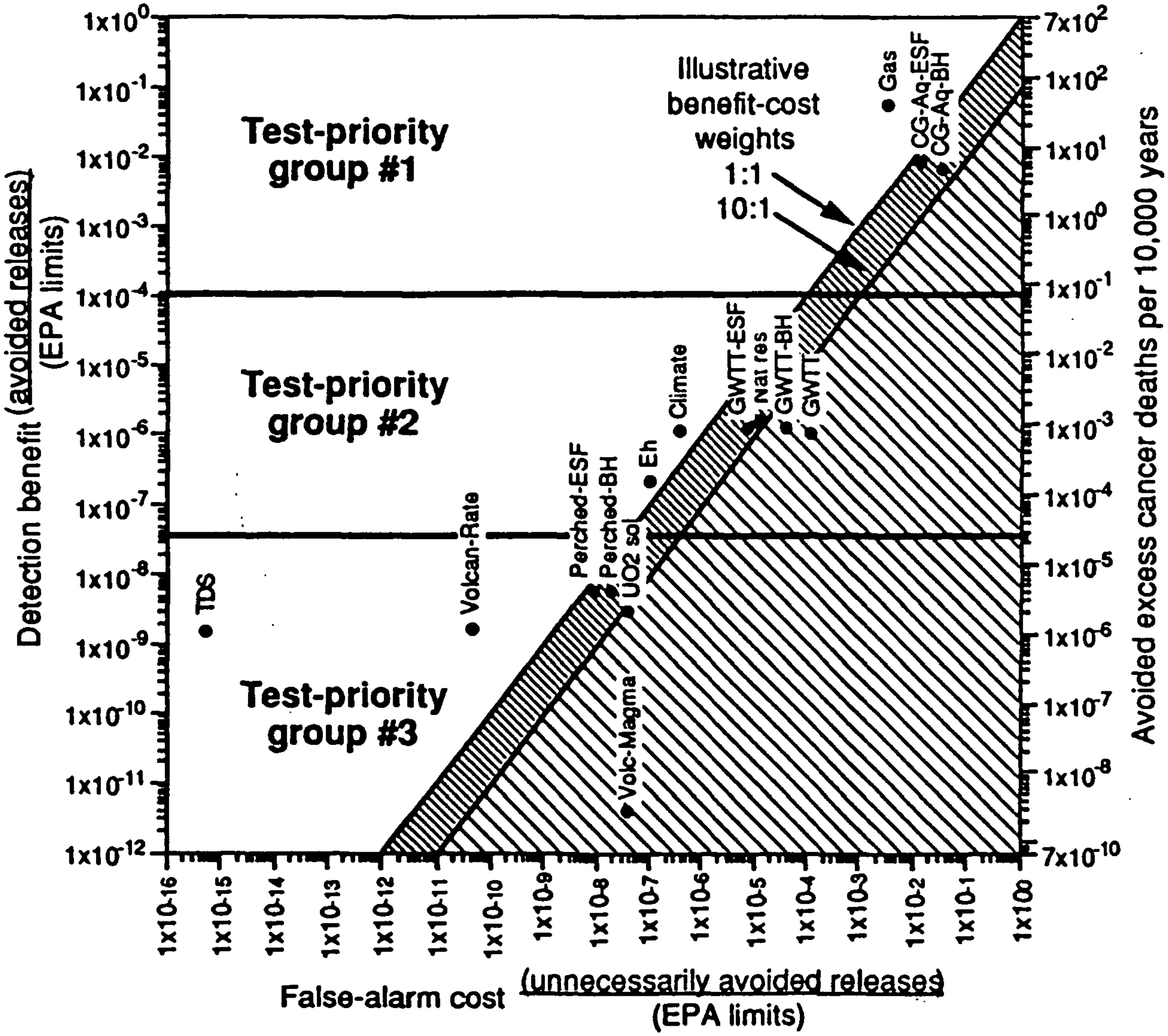
EXAMPLE OF TEMPERATURE HISTORIES OF SPENT FUEL WASTE PACKAGE AND HOST ROCK (VERTICAL EMPLACEMENT)



INITIAL CONDITIONS

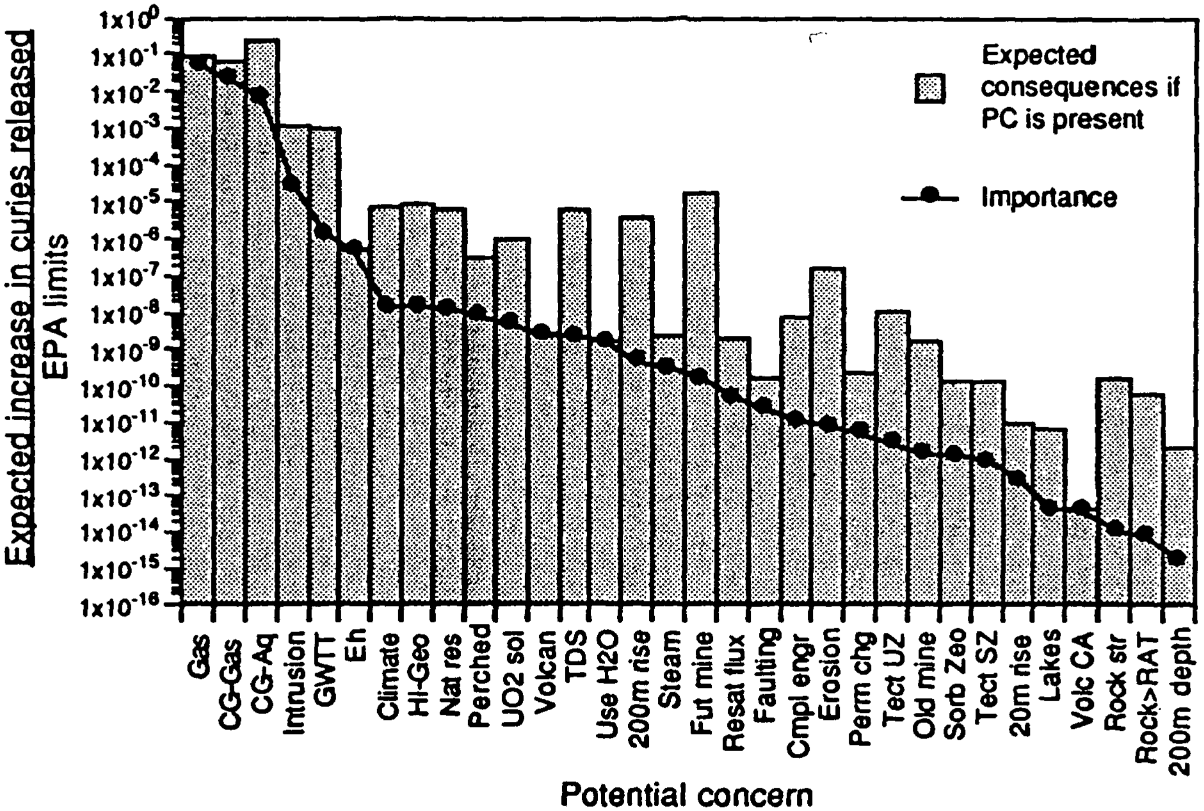
WASTE FORM . . .	SPENT FUEL
LOCAL POWER DENSITY . . .	57.0 kW/acre
AREAL POWER DENSITY . . .	48.4
AVERAGE 10-YR POWER . . .	3.3 kW
CONTAINER DIAMETER . . .	0.7 m
DISTANCE BETWEEN CONTAINERS . . .	5 m
DISTANCE BETWEEN DRIFTS . . .	47 m

RESULTS OF PHASE I TEST PRIORITIZATION

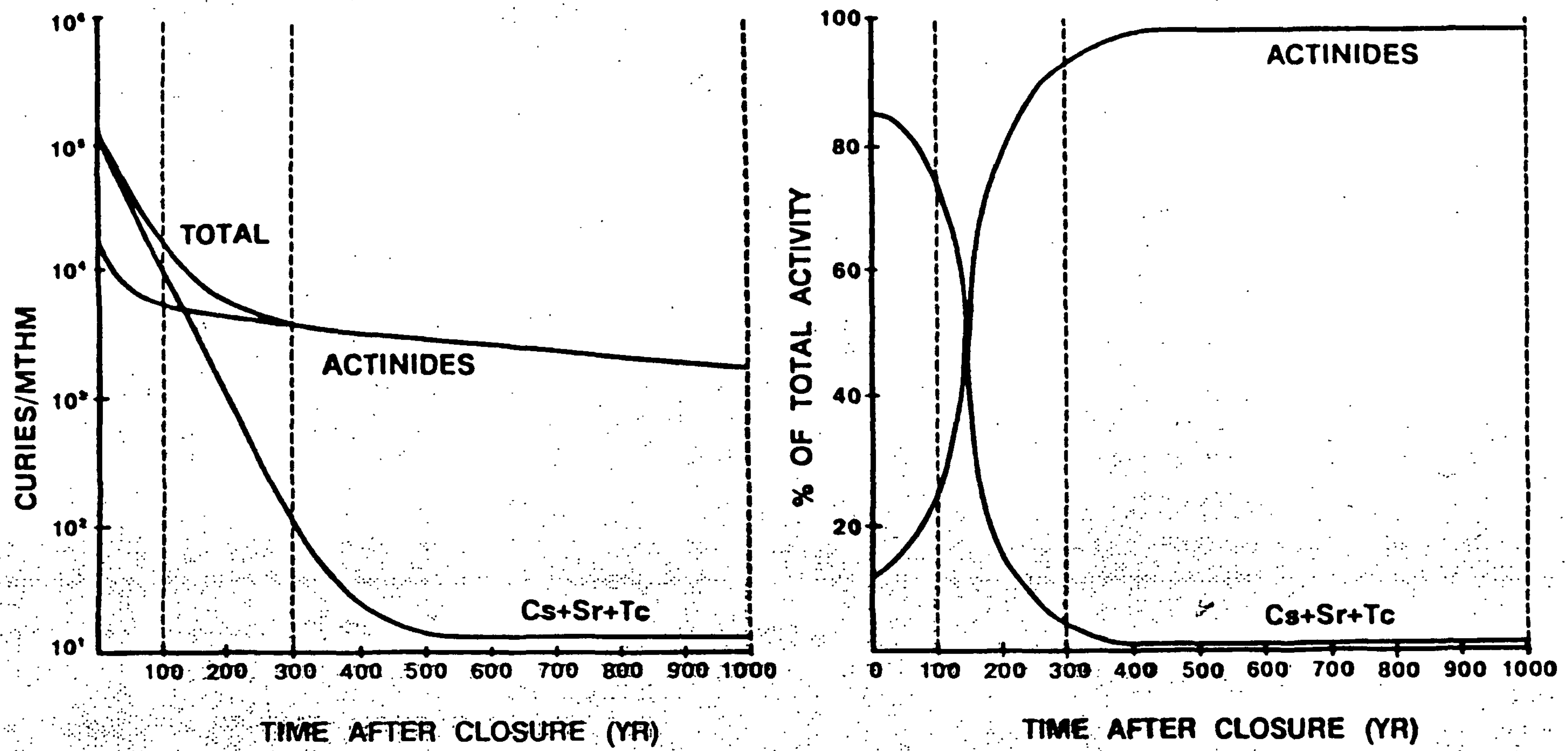


HGGRUP5P.125.NWTRB/6-25/27-91

IMPORTANCE AND EXPECTED CONSEQUENCES OF POTENTIAL CONCERNS



KEY RADIONUCLIDES ACTIVITIES AND RELATIVE CONTRIBUTIONS



ACNWT11P.A01 10/18/88