



# Potential detection of low level aerosol isotopes from new civilian nuclear processes



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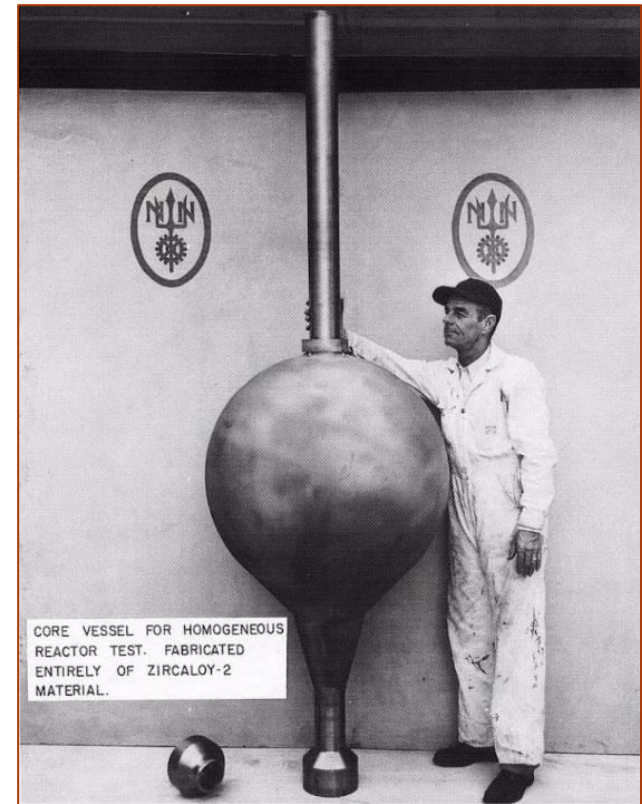
CTBT Science and Technology 2017

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# Potential New Sources of Low Level Activity in the Environment

- ▶ Aqueous Homogenous Reactor
  - Fuel – H<sub>2</sub>O mixture
- ▶ Direct extraction of fission products from liquid
- ▶ Motivation: Can this create ‘new’ environmental activity?

- ▶ Our goal: compare to common background sources, specifically, power reactors



## Aqueous Homogenous Reactor

[https://en.wikipedia.org/wiki/Aqueous\\_homogeneous\\_reactor](https://en.wikipedia.org/wiki/Aqueous_homogeneous_reactor)



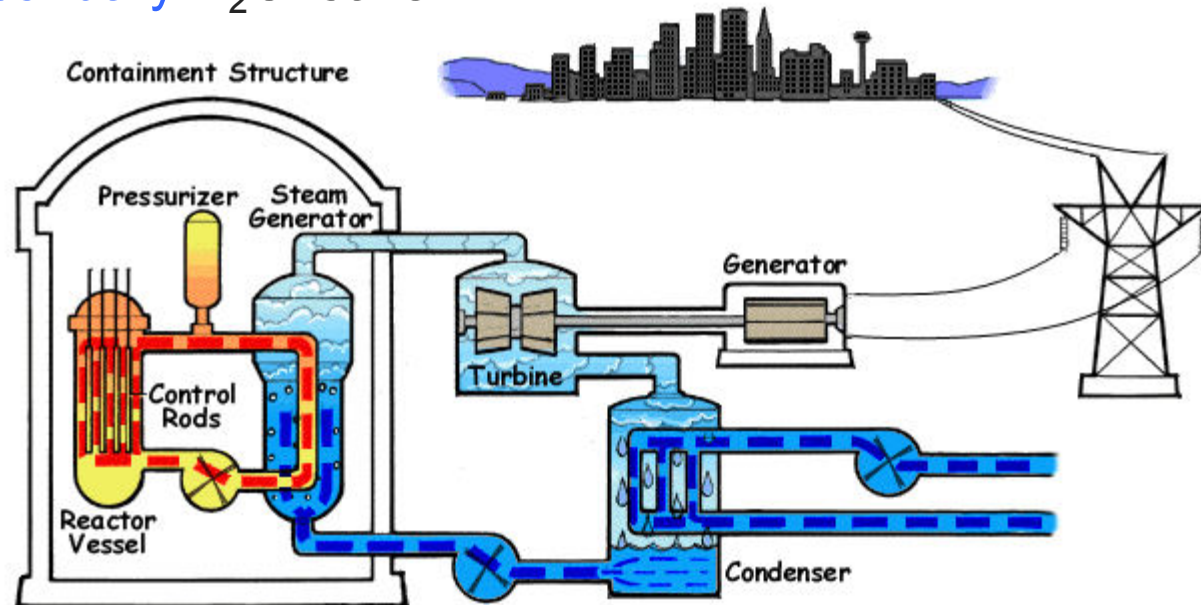
# Comparing Old and New Sources

## Power Reactor

- ▶ Fuel inside pressure vessel
  - Several cubic meters
- ▶ Zirconium clad oxide fuel
- ▶ Low-medium-high MWD/T
- ▶ Potential for 'tramp uranium'
- ▶ **Primary** to **secondary** H<sub>2</sub>O leaks?

## New Methods In Development

- ▶ Liquid target with <sup>235</sup>U in solution
- ▶ Irradiation via reactor or accelerator neutrons -> Fission
- ▶ Continuous or batch
  - flow + separation + recycling

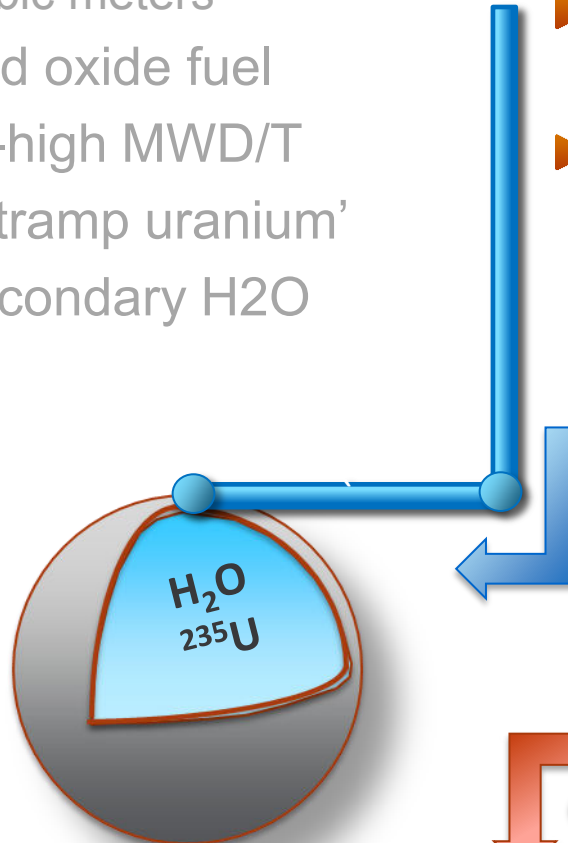




# Comparing Old and New Sources

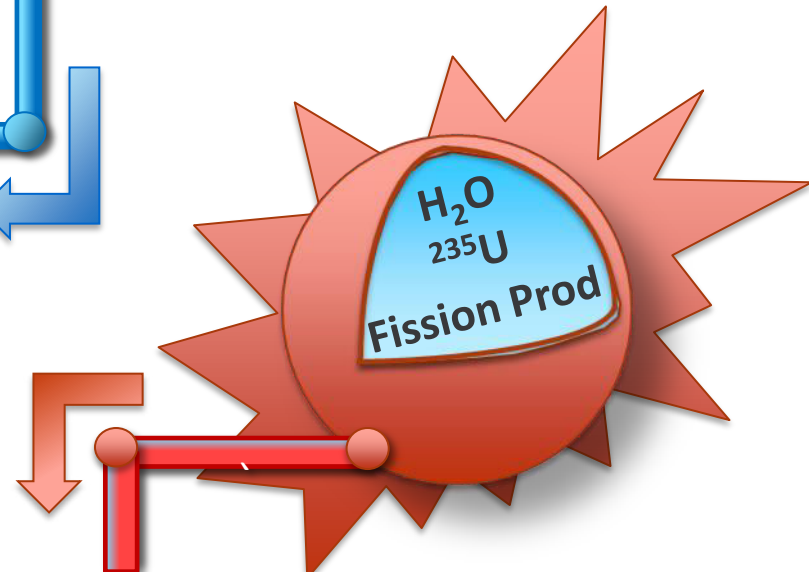
## Power Reactor

- ▶ Fuel inside pressure vessel
  - Several cubic meters
- ▶ Zirconium clad oxide fuel
- ▶ Low-medium-high MWD/T
- ▶ Potential for 'tramp uranium'
- ▶ Primary to secondary H<sub>2</sub>O leaks?



## New Methods In Development

- ▶ Liquid with  $^{235}U$  in solution
- ▶ Irradiation via reactor or external accelerator neutrons -> Fission
- ▶ Continuous or batch
  - flow + separation + recycling



# Detectable Aerosol Species with Gas or Volatile Precursors



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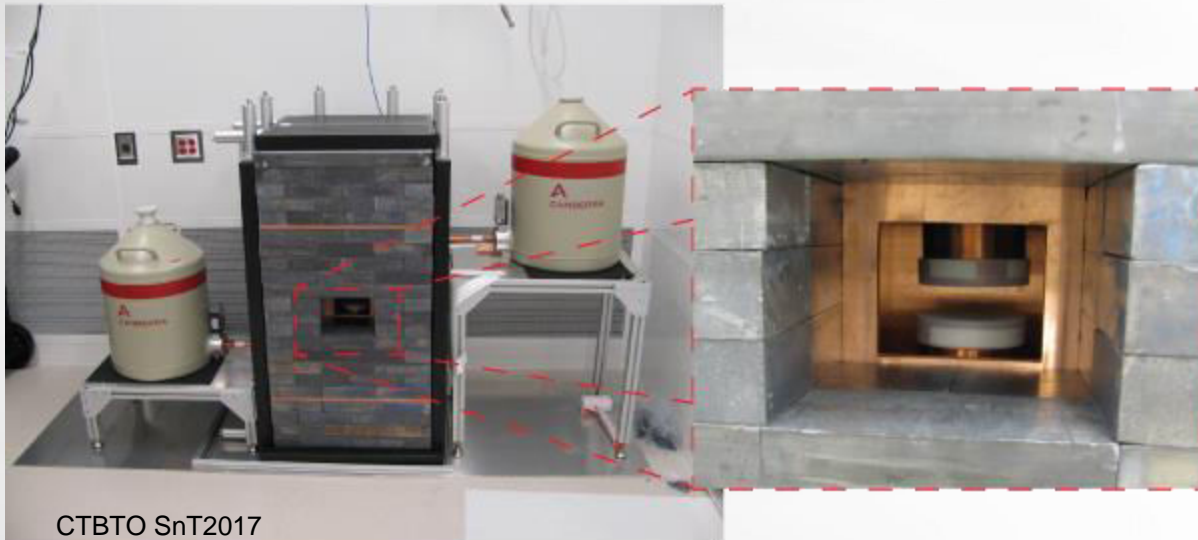
Volatile Parent	Half-life	Decay Product	Half-life	Decay Product	Half-life	Decay Product	Half-life	Decay Product	Half-life	Decay Product	Half-life
<sup>88</sup> Br	17 s	<sup>88</sup> Kr	2.8 h	<sup>88</sup> Rb	18 m	<sup>88</sup> Sr	Stable				
<sup>89</sup> Br	4.4 s	<sup>89</sup> Kr	3.2 m	<sup>89</sup> Rb	15 m	<sup>89</sup> Sr	51 d	<sup>89</sup> Y	Stable		
<sup>91</sup> Br	0.54 s	<sup>91</sup> Kr	8.6 s	<sup>91</sup> Rb	58 s	<sup>91</sup> Sr	9.7 h	<sup>91m</sup> Y	50 m	<sup>91</sup> Y	59 d
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<sup>91</sup> Br	0.31 s	<sup>92</sup> Kr	1.8 s	<sup>92</sup> Rb	4.5 s	<sup>92</sup> Sr	2.7 h	<sup>92</sup> Y	3.5 h	<sup>92</sup> Zr	Stable
<sup>92</sup> Br	0.31 s	<sup>92</sup> Kr	1.8 s	<sup>92</sup> Rb	4.5 s	<sup>92</sup> Sr	2.7 h	<sup>92</sup> Y	3.5 h	<sup>92</sup> Zr	Stable
<sup>93</sup> Kr	1.29 s	<sup>93</sup> Rb	5.8 s	<sup>93</sup> Sr	7.4 m	<sup>93</sup> Y	10 h	<sup>93</sup> Zr	1.6E6 y	<sup>93</sup> Nb	Stable
<sup>94</sup> Kr	210 ms	<sup>94</sup> Rb	2.7 s	<sup>94</sup> Sr	75 s	<sup>94</sup> Y	19 m	<sup>94</sup> Zr	Stable		
<sup>95</sup> Kr	0.11 s	<sup>95</sup> Rb	380 ms	<sup>95</sup> Sr	24 s	<sup>95</sup> Y	10 m	<sup>95</sup> Zr	64 d	<sup>95</sup> Nb	35 d
<sup>95</sup> Kr	0.11 s	<sup>95</sup> Rb	380 ms	<sup>95</sup> Sr	24 s	<sup>95</sup> Y	10 m	<sup>95</sup> Zr	64 d	<sup>95</sup> Nb	35 d
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<sup>137</sup> I	25 s	<sup>137</sup> Xe	3.8 m	<sup>137</sup> Cs	30 y	<sup>137m</sup> Ba	2.5 m	<sup>137</sup> Ba	Stable		
<sup>138</sup> I	6.2 s	<sup>138</sup> Xe	14 m	<sup>138</sup> Cs	33 m	<sup>138</sup> Ba	Stable				
<sup>139</sup> I	2.3 s	<sup>139</sup> Xe	40 s	<sup>139</sup> Cs	9.3 m	<sup>139</sup> Ba	83 m	<sup>139</sup> La	Stable		
<sup>140</sup> I	0.86 s	<sup>140</sup> Xe	14 s	<sup>140</sup> Cs	64 s	<sup>140</sup> Ba	13 d	<sup>140</sup> La	1.7 d	<sup>140</sup> Ce	Stable
<sup>140</sup> I	0.86 s	<sup>140</sup> X	14 s	<sup>140</sup> Cs	64 s	<sup>140</sup> Ba	13 d	<sup>140</sup> La	1.7 d	<sup>140</sup> Ce	Stable
<sup>141</sup> I	0.43 s	<sup>141</sup> Xe	1.7 s	<sup>141</sup> Cs	25 s	<sup>141</sup> Ba	18 m	<sup>141</sup> La	3.9 h	<sup>141</sup> Ce	33 d
<sup>141</sup> I	0.43 s	<sup>141</sup> Xe	1.7 s	<sup>141</sup> Cs	25 s	<sup>141</sup> Ba	18 m	<sup>141</sup> La	3.9 h	<sup>141</sup> Ce	33 d
<sup>142</sup> Xe	1.2 s	<sup>142</sup> Cs	1.7 s	<sup>142</sup> Ba	11 m	<sup>142</sup> La	91 m	<sup>142</sup> Ce	Stable		
<sup>143</sup> Xe	0.51 s	<sup>143</sup> Cs	1.8 s	<sup>143</sup> Ba	15 s	<sup>143</sup> La	14 m	<sup>143</sup> Ce	33 h	<sup>143</sup> Pr	14 d
<sup>143</sup> Xe	0.51 s	<sup>143</sup> Cs	1.8 s	<sup>143</sup> Ba	15 s	<sup>143</sup> La	14 m	<sup>143</sup> Ce	33 h	<sup>143</sup> Pr	14 d
<sup>145</sup> Xe	190 ms	<sup>145</sup> Cs	0.59 s	<sup>145</sup> Ba	4.3 s	<sup>145</sup> La	41 s	<sup>145</sup> Ce	3.0 m	<sup>145</sup> Pr	6.0 h

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# Minimum Detectable Activities

- ▶ Background, efficiency, geometry of a dual HPGe system in the PNNL Shallow Underground Laboratory were used
  - 30 meters water equivalent
  - 2x 50% BEGe ultra low background
  - Cosmic veto
- ▶ Assume best case count time, geometry per isotope.



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Nuclide	Half-life (s)	Energy (keV)	Yield (%)	MDA (Bq/sample)
<sup>88</sup> Rb	1066	1836	22.7	3E-01
<sup>89</sup> Rb	919	1031.9	62.9	8E-02
<sup>91</sup> Sr	34740	1024.3	33.5	5E-02
<sup>91</sup> Y	5E+06	1204.8	0.26	1E+00
<sup>91m</sup> Y	2983	555.6	95	1E-02
<sup>92</sup> Sr	9400	1383.9	90	8E-03
<sup>92</sup> Y	12744	934.5	13.9	1E-01
<sup>93</sup> Y	36648	266.9	7.3	1E-01
<sup>94</sup> Y	1122	918.7	56	7E-02
<sup>95</sup> Nb	3E+06	765.8	99.8	3E-03
<sup>95</sup> Y	618	954	15.8	4E-01
<sup>95</sup> Zr	6E+06	756.7	54.4	5E-03
<sup>137</sup> Cs	9E+08	661.7	89.9	3E-03
<sup>138</sup> Cs	2005	462.8	30.8	4E-02
<sup>139</sup> Ba	4984	165.9	23.8	1E-01
<sup>140</sup> Ba	1E+06	537.3	24.4	1E-02
<sup>140</sup> La	145027	1596.2	95.4	5E-03
<sup>141</sup> Ce	3E+06	145.4	48.3	2E-03
<sup>141</sup> La	14112	1354.5	1.6	1E+00
<sup>142</sup> La	5466	641.3	47.4	3E-02
<sup>143</sup> Ce	118940	293.3	42.8	1E-02
<sup>145</sup> Pr	21542	748.3	0.5	2E+00

# Assumptions

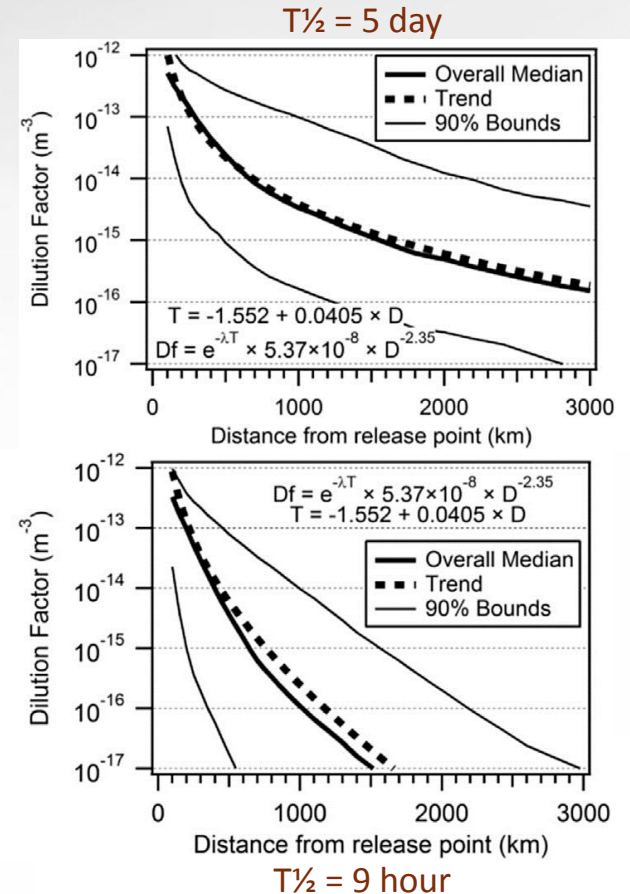
## Production, Release, Decay, Dilution



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- ▶ Creation, fission build-up and decay time in fission vessel
  - ORIGEN
- ▶ Residence time in fission vessel
  - Power Reactor: 3 hours
  - “AHR”: 1 second
- ▶ Decay and dilution after release
  - Dilution trend formulae from JER 148 (2015) pp123-129
  - Concentrations compared to best case low level radioactivity measurement technology





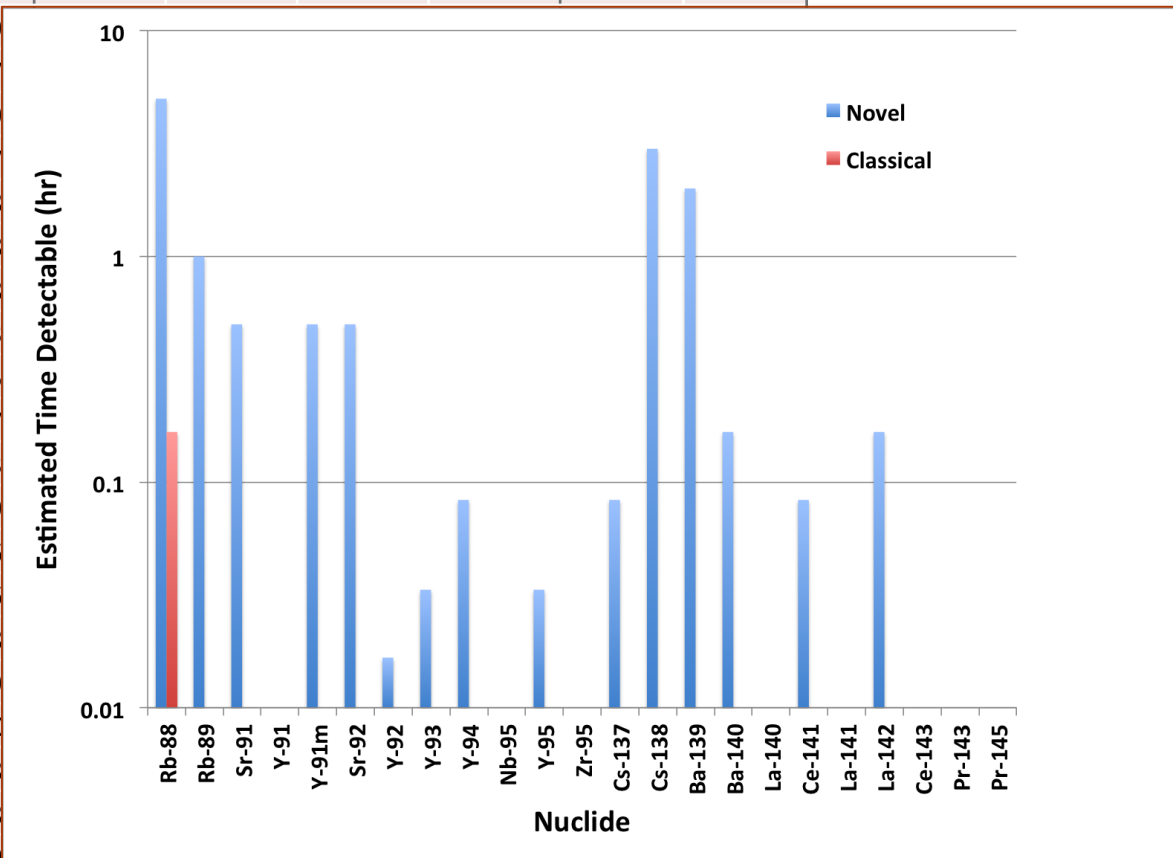
# Detectability on a Low Level System

Nuclide	Novel Production Notional Concentrations (Bq/m <sup>3</sup> )				Reactor Emissions Notional Concentration (Bq/m <sup>3</sup> )				MDC MDA (Bq) (Bq/m <sup>3</sup> )	
	1 min	30 min	1 hr	5 hrs	1 min	30 min	1 hr	5 hrs	MDA (Bq)	(Bq/m <sup>3</sup> )
<sup>88</sup> Rb	3E+01	3E-01	9E-02	1E-03	1E-02	1E-04	4E-05	5E-07	3E-01	3E-04
<sup>89</sup> Rb	5E+01	5E-02	3E-03	1E-09	3E-19	3E-22	2E-23	0E+00	8E-02	8E-05
<sup>91</sup> Sr	9E-02	1E-04	3E-05	6E-07	0E+00	0E+00	0E+00	0E+00	5E-02	5E-05
<sup>91</sup> Y	1E-07	2E-08	8E-09	1E-09	0E+00	0E+00	0E+00	0E+00	1E+00	1E-03
<sup>91m</sup> Y	3E-04	2E-05	9E-06	4E-07	0E+00	0E+00	0E+00	0E+00	1E-02	1E-05
<sup>92</sup> Sr	7E-02	4E-05	7E-06	8E-08	0E+00	0E+00	0E+00	0E+00	8E-03	8E-06
<sup>92</sup> Y	2E-04	4E-06	1E-06	9E-08	0E+00	0E+00	0E+00	0E+00	1E-01	1E-04
<sup>93</sup> Y	4E-04	2E-06	5E-07	1E-08	0E+00	0E+00	0E+00	0E+00	1E-01	1E-04
<sup>94</sup> Y	4E-03	2E-06	1E-07	7E-13	0E+00	0E+00	0E+00	0E+00	7E-02	7E-05
<sup>95</sup> Nb	4E-14	3E-14	2E-14	4E-15	0E+00	0E+00	0E+00	0E+00	3E-03	3E-06
<sup>95</sup> Y	2E-03	2E-07	5E-09	2E-17	0E+00	0E+00	0E+00	0E+00	4E-01	4E-04
<sup>95</sup> Zr	8E-09	1E-10	3E-11	9E-13	0E+00	0E+00	0E+00	0E+00	5E-03	5E-06
<sup>137</sup> Cs	6E-05	2E-07	5E-08	1E-09	4E-22	1E-24	3E-25	9E-27	3E-03	3E-06
<sup>138</sup> Cs	3E+01	2E-01	4E-02	7E-06	4E-06	3E-08	5E-09	9E-13	4E-02	4E-05
<sup>139</sup> Ba	3E-01	5E-03	9E-04	4E-06	0E+00	0E+00	0E+00	0E+00	1E-01	1E-04
<sup>140</sup> Ba	4E-03	6E-06	1E-06	4E-08	0E+00	0E+00	0E+00	0E+00	1E-02	1E-05
<sup>140</sup> La	5E-07	5E-08	2E-08	3E-09	0E+00	0E+00	0E+00	0E+00	5E-03	5E-06
<sup>141</sup> Ce	4E+00	0E+00	0E+00	0E+00	0E+00	0E+00	0E+00	0E+00	2E-03	2E-06
<sup>141</sup> La	8E-04	1E-05	4E-06	7E-08	0E+00	0E+00	0E+00	0E+00	1E+00	1E-03
<sup>142</sup> La	1E-03	9E-06	2E-06	1E-08	0E+00	0E+00	0E+00	0E+00	3E-02	3E-05
<sup>143</sup> Ce	1E-06	2E-08	5E-09	1E-10	0E+00	0E+00	0E+00	0E+00	1E-02	1E-05
<sup>145</sup> Pr	2E-07	2E-09	3E-10	6E-12	0E+00	0E+00	0E+00	0E+00	2E+00	2E-03



# Detectability on a Low Level System

Nuclide	Novel Production Notional Concentrations (Bq/m <sup>3</sup> )				Reactor Emissions Notional Concentration (Bq/m <sup>3</sup> )				MDA (Bq)	MDC (Bq/m <sup>3</sup> )
	1 min	30 min	1 hr	5 hrs	1 min	30 min	1 hr	5 hrs		
<sup>88</sup> Rb	3E+01	3E-01	9E-02	1E-03	1E-02	1E-04	4E-05	5E-07	3E-01	3E-04
<sup>89</sup> Rb	5E+01	5E-02	3E-03	1E-09						
<sup>91</sup> Sr	9E-02	1E-04	3E-05	6E-07						
<sup>91</sup> Y	1E-07	2E-08	8E-09	1E-09						
<sup>91m</sup> Y	3E-04	2E-05	9E-06	4E-07						
<sup>92</sup> Sr	7E-02	4E-05	7E-06	8E-08						
<sup>92</sup> Y	2E-04	4E-06	1E-06	9E-08						
<sup>93</sup> Y	4E-04	2E-06	5E-07	1E-08						
<sup>94</sup> Y	4E-03	2E-06	1E-07	7E-13						
<sup>95</sup> Nb	4E-14	3E-14	2E-14	4E-15						
<sup>95</sup> Y	2E-03	2E-07	5E-09	2E-17						
<sup>95</sup> Zr	8E-09	1E-10	3E-11	9E-13						
<sup>137</sup> Cs	6E-05	2E-07	5E-08	1E-09						
<sup>138</sup> Cs	3E+01	2E-01	4E-02	7E-06						
<sup>139</sup> Ba	3E-01	5E-03	9E-04	4E-06						
<sup>140</sup> Ba	4E-03	6E-06	1E-06	4E-08						
<sup>140</sup> La	5E-07	5E-08	2E-08	3E-09						
<sup>141</sup> Ce	4E+00	0E+00	0E+00	0E+00						
<sup>141</sup> La	8E-04	1E-05	4E-06	7E-08						
<sup>142</sup> La	1E-03	9E-06	2E-06	1E-08						
<sup>143</sup> Ce	1E-06	2E-08	5E-09	1E-10						
<sup>145</sup> Pr	2E-07	2E-09	3E-10	6E-12	0E+00	0E+00	0E+00	0E+00	2E+00	2E-03





# Conclusions

- ▶ This was one simple study
- ▶ It obtained results that some fission isotopes not normally observed from reactors could be seen substantially downwind in the environment under certain leakage assumptions.
- ▶ All trace environmental detections are of course far more prominent on low-level measurement systems
  - The subject of this study might **ONLY** be detectable on ultra-low level systems.
- ▶ H.S. Miley, J.L. Burnett, M.P. Foxe, D.A. Haas, M.E. Keillor, J.D. Lowrey, M.F. Mayer, J.I. McIntyre, J.S. Wood,
- ▶ The potential detection of low-level aerosol isotopes from new civilian nuclear processes,
- ▶ Applied Radiation and Isotopes, Available online 21 February 2017, ISSN 0969-8043, <https://doi.org/10.1016/j.apradiso.2017.02.033>.  
(<http://www.sciencedirect.com/science/article/pii/S0969804316306480>)



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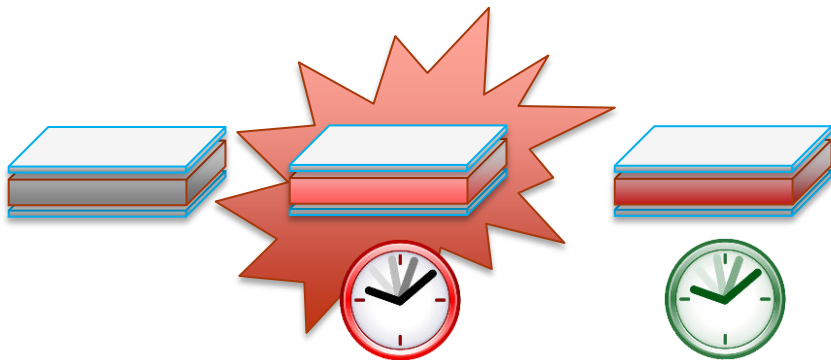




# Comparing Old and New Sources

## Classical Medical Isotope Production

- ▶  $^{235}\text{U}$  target inside metal sandwich
  - Worldwide switch to LEU
- ▶ Irradiation in reactor -> Fission
- ▶ Cooling Time
- ▶ Dissolution in filtered hood
  - Holdup and trapping for  $^{131}\text{I}$
- ▶ Eventual discharges through high volume stack



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## New Methods In Development

- ▶ Liquid target with  $^{235}\text{U}$  in solution
- ▶ Irradiation via reactor or accelerator neutrons -> Fission
- ▶ Continuous flow, separation, recycling

