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The use of Argon-37 measurements as a coincident signature with radioxenon and in high background environments in the International Monitoring System

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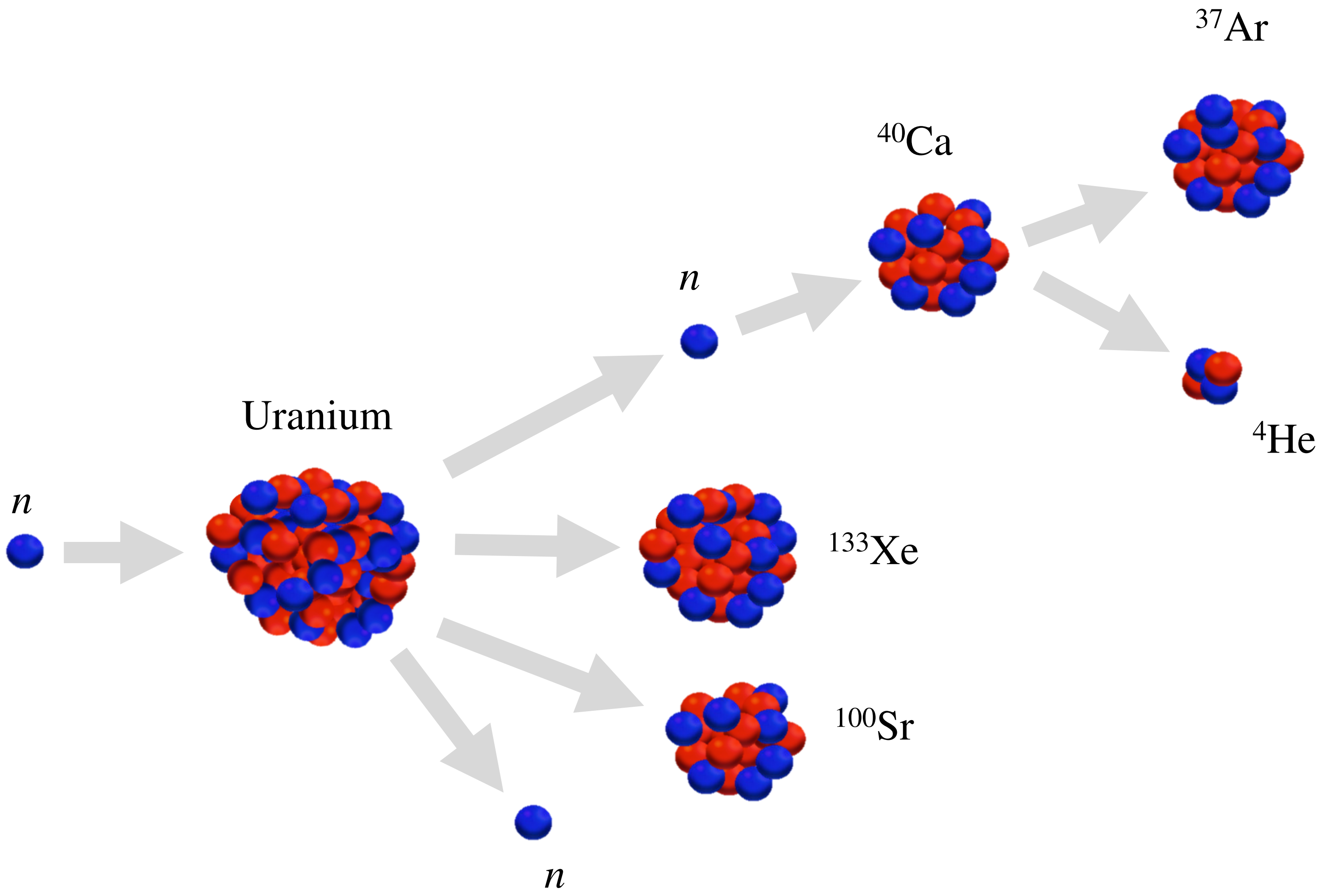
Production of radioactive gases from a nuclear explosion



Man-made backgrounds

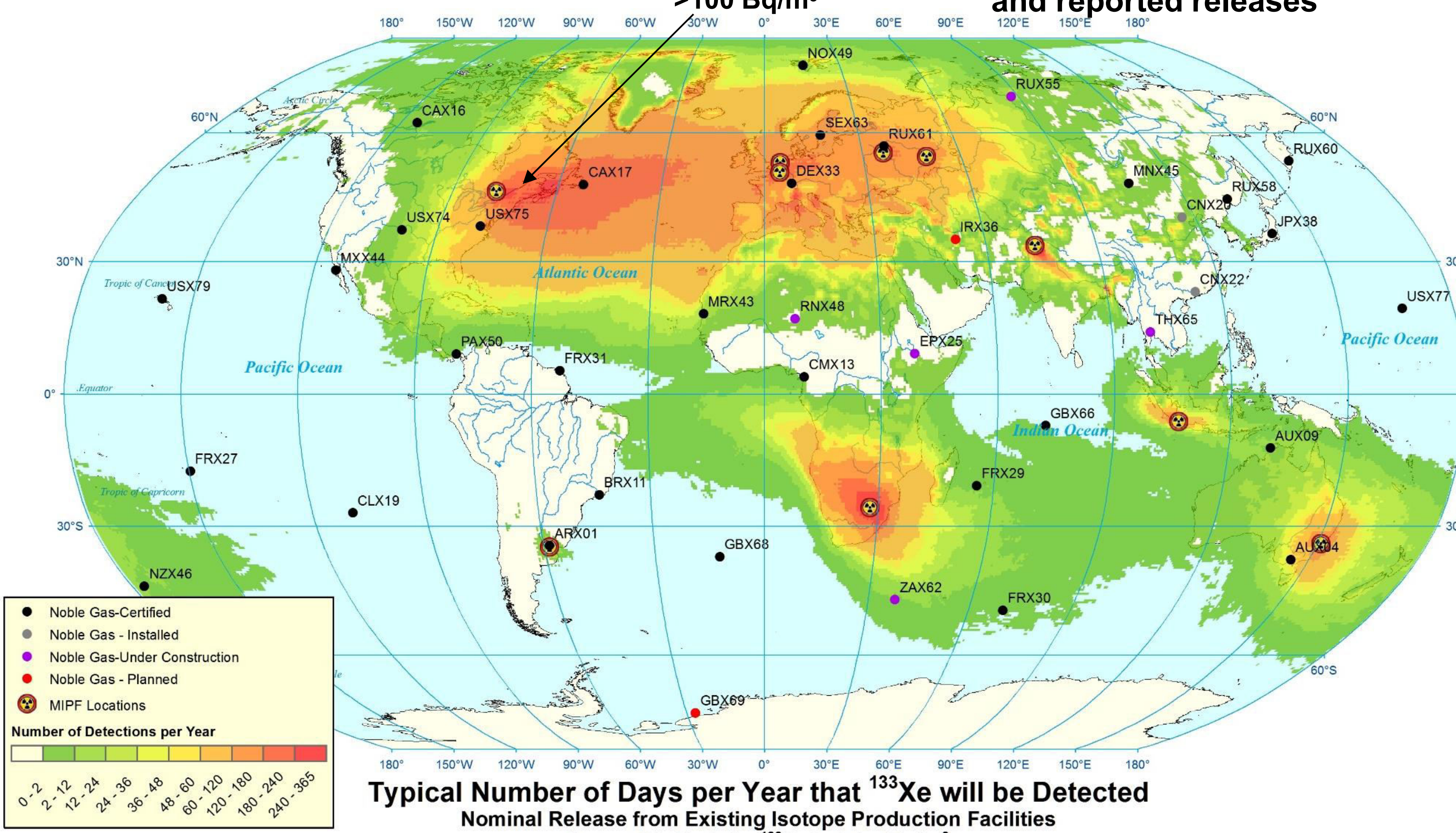
- Over the last several years, we have seen a number of cases in which radionuclide background caused by various man-made phenomena have caused problems with the understanding of radionuclide measurements in the IMS
  - Specifically ^{133}Xe detections have been frequent

- The situation is getting more frequent and we are in search of tools to try to improve the situation

- See presentations by Friese, et al.

Map of Xe backgrounds from ^{99}Mo production

Calculations based on estimated and reported releases



Typical Number of Days per Year that ^{133}Xe will be Detected
Nominal Release from Existing Isotope Production Facilities
Detectable limit for ^{133}Xe is 0.2 mBq/m^3

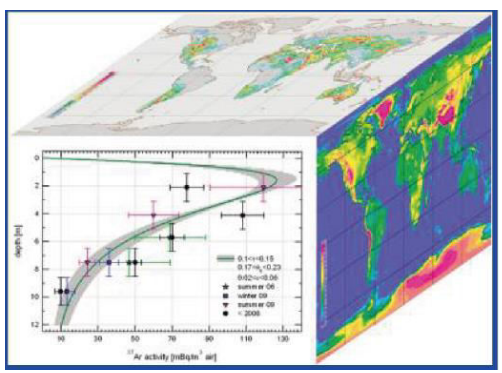
Argon-37 for OSI

- Argon-37 has been identified as a nuclear explosion signature for on-site inspection and was discussed at SnT2015 from the China Academy of Engineering Physics.
  - "…a definitive and unambiguous indicator of an underground nuclear explosion."

• ^{37}Ar signature was verified during the last Chinese UNEs. Subsoil gas sample in two months after the explosion showed that the activity was in the order of 10^2-10^7Bq/m^3.

中国工程物理研究院
CHINA ACADEMY OF ENGINEERING PHYSICS

  - Work by Riedmann and Purschert have also explored the signature, including a study of global backgrounds



Riedmann and Purschert, Env. Sci. Tech. 2011.

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A signature to consider for the IMS

- We should consider augmenting current signatures and bring something else to the table
  - Xe-133 is a good signature; I am not suggesting replacing it
- A good signature should have a reasonable chance to be detected in the IMS for at least a few weeks
- Would ideally be based on existing or modification of existing capabilities

Argon-37
Noble gas
Half-life: 35 days
Production: Approximately 10^{13}-10^{14} Bq/kt (see below)

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The basic advantages of ^{37}Ar for the IMS

- It has the perfect half-life
  - Long enough to be detectable for a long time, but short enough that it has not built up in the atmosphere

- The background of ^{37}Ar may be low many places on earth and anyway is most likely different than ^{133}Xe, so that where the ^{133}Xe background is high, ^{37}Ar might not be

- An explosion is expected to release both ^{37}Ar and ^{133}Xe and therefore, can be detected in coincidence at a station

- The production yield (Bq/kt) is 'pretty high', depending on soil conditions
  - ~10^{13} Bq/kt (Gasbuggy, 1971)
  - Up to 10^{14} Bq/kt, depending on rock type (Egnatuk, 2015)

Production yields – ^{37}Ar and ^{133}Xe

- Xe-133
  - Easy to calculate, based on fission yields
  - Cumulative yields maximum at about 3 days (10^{16} Bq)

- Ar-37
  - Estimates range of 10^{13} to 10^{14} Bq/kiloton, depends on
    - The amount of calcium in the soil (assumed a few percent)
      - By far the largest contributor to the variability
    - The n energy spectrum chosen (Watt spectrum assumed)
    - Details of the simulation (MCNP has been used)

While the cumulative radioxenon and radioargon production are near 10^{14} Bq (Ar-37) to 10^{16} Bq (Xe-133), the amount released can be much less

In principle, the amount of either gas could be "zero"

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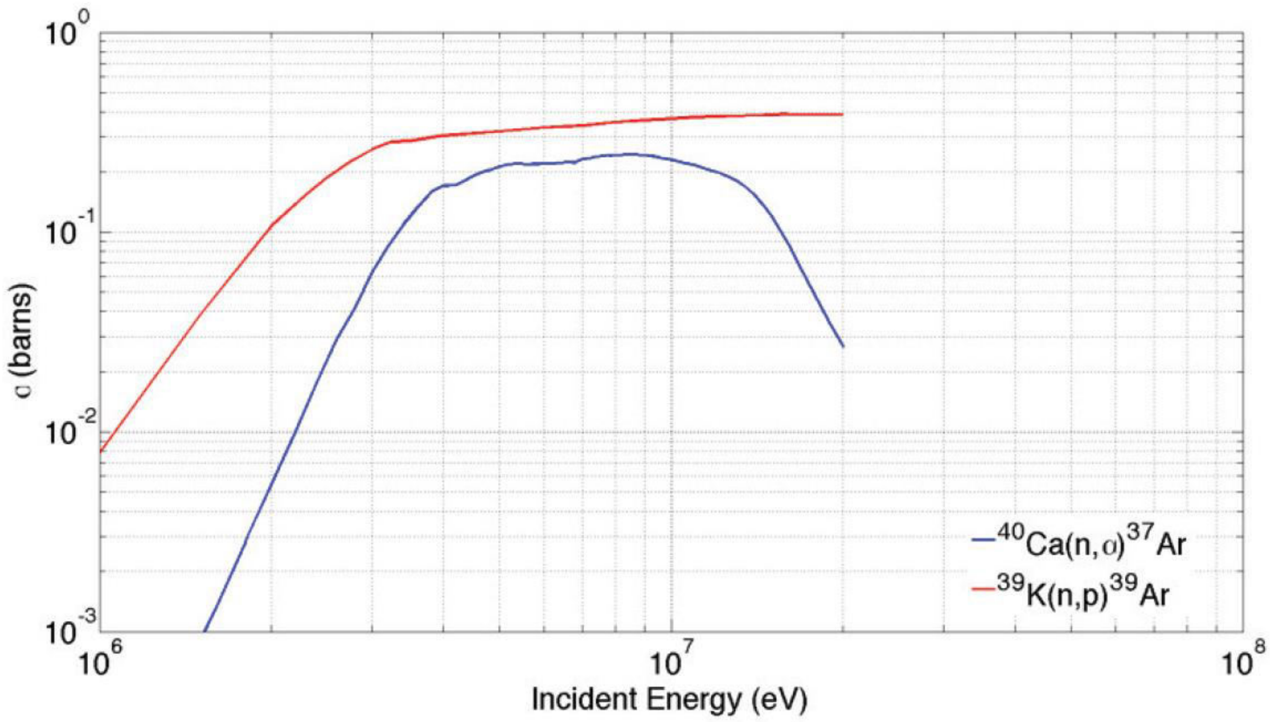
Source term

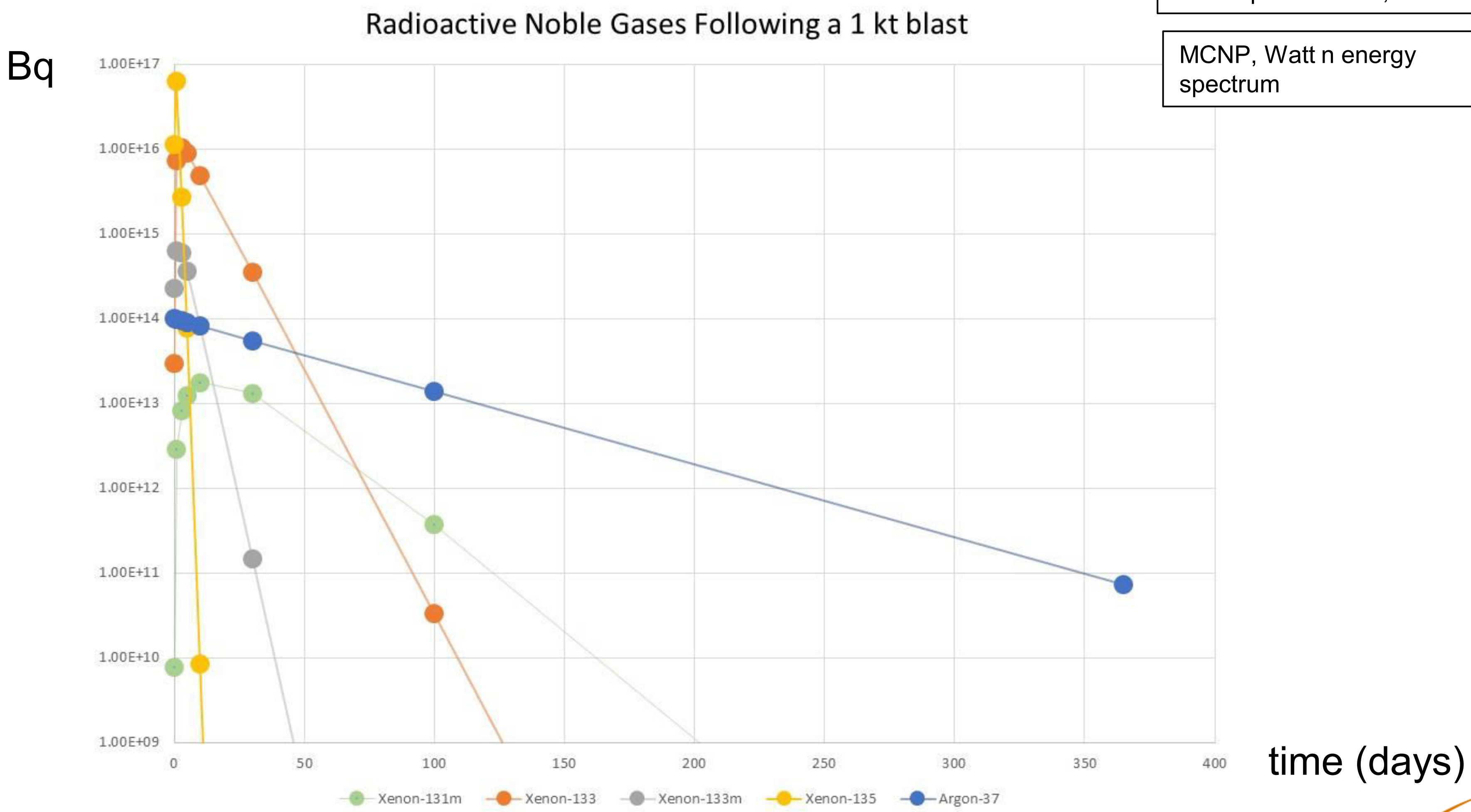
- $10^{13} – 10^{14}$ Bq/kiloton for most soils
  - Created through (n,α) on ^{40}Ca
  - Argon-39 also created via (n,p) on ^{39}K
- The ^{39}Ar background may be high due to its 269-year half-life
  - See poster by McIntyre



Relative size of the signal from all NGs

^{239}Pu explosion
10^{14} Bq/kiloton ^{37}Ar, 1% Ca

MCNP, Watt n energy spectrum



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Figures-of-merit; comparison to ^{133}Xe

- Detectability
  - Compare expected signal size to the average background
- Selectivity
  - Compare expected signal size to background sources and the variability of the background
- Costs
  - Existing IMS technologies are approximately $1M+ capital plus $100K+ annual costs

Figure of Merit (Detectability Ratio)

Expected Concentration / Expected Background

Figures of merit (FOM)

- Define a figure of merit (FOM)
  - Expected concentration of an analyte (expected unrelated background) at the measurement point from other processes

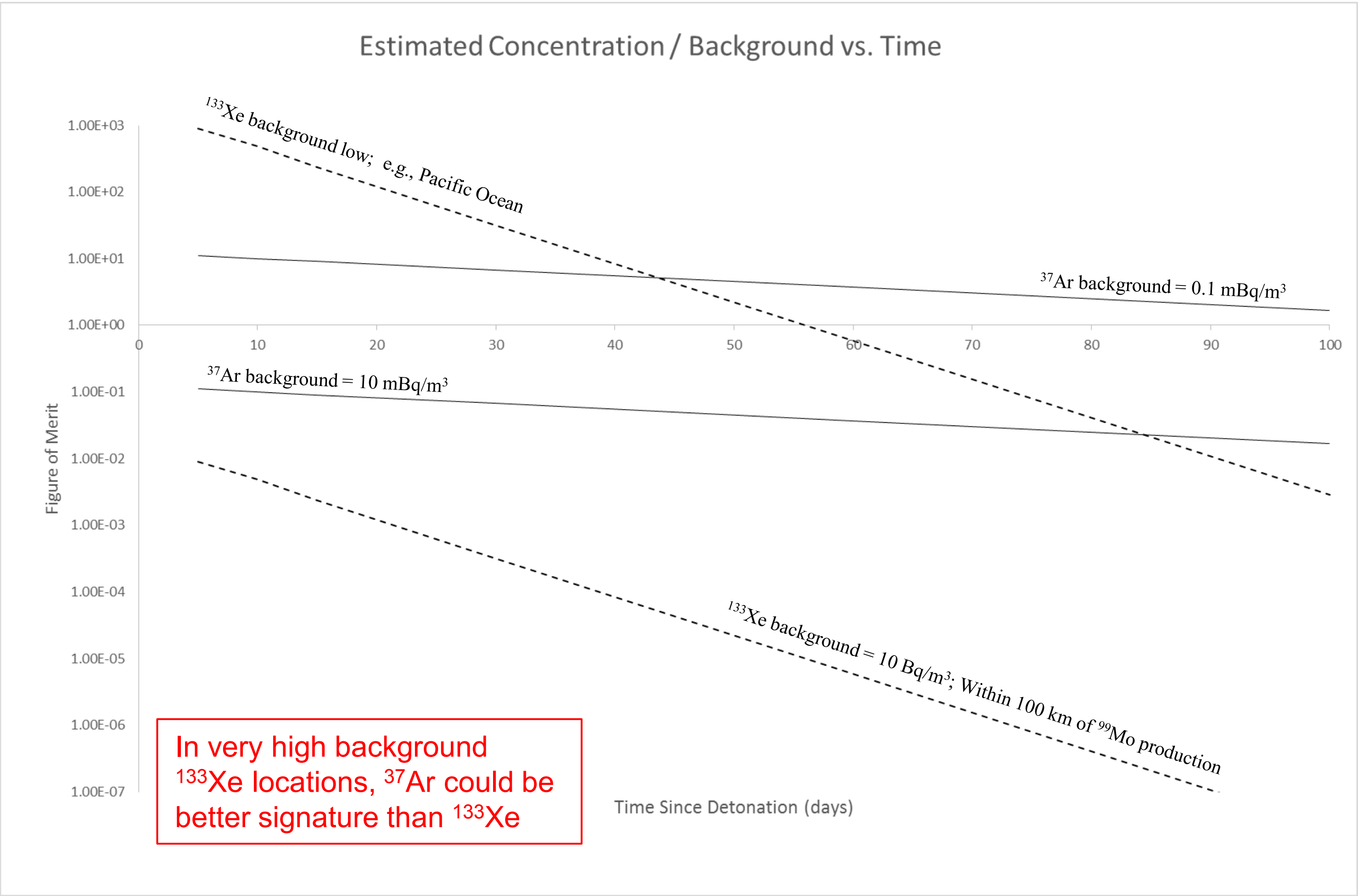
Isotope	Half-life (d)	Yield (10 days)	Concentration*	Lowest expected**	Highest expected***	Average background	10 days		
		Bq	(mBq/m3)	(mBq/m3)	(mBq/m3)	(mBq/m3)	FOM-	<FOM>	FOM+
Ar-37	35	1.0E+14	1.000	0.1	10	2	10	0.5	0.1
Ar-39	9.8E+04	3.6E+10	3.6E-04	10	10	10	3.6E-05	3.6E-05	3.6E-05
Kr-85	4.0E+03	4.0E+11	4.0E-03	1500	1.5E+06	1.5E+03	2.7E-06	2.7E-06	2.7E-09
Xe-131m	11.9	1.8E+13	0.177	0.1	100	0.2	1.77	0.885	1.8E-03
Xe-133	5.2	4.9E+15	48.500	0.1	1.0E+04	2	485	24.25	4.9E-03
Xe-133m	2.2	8.1E+13	0.813	0.1	100	0.2	8.13	4.065	8.1E-03
Xe-135	0.38	8.5E+09	8.5E-05	0.1	100	0.01	8.5E-04	8.5E-03	8.5E-07

* Assumes 0.1% emission rate
** Lowest concentrations maybe lower, but set at instrumentally-constrained levels (MDCs)
*** Estimate

Low background — Average background — High background

- Note – some of the inputs to the FOMs are estimates; further work is needed to determine appropriate values

Range of FOMs between Ar and Xe



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Interpretation

- At t > 80 days (delayed releases) ^{37}Ar is a better signature than any radioxenon because of its half-life
  - The IMS detected a delayed release from DPRK of ~55 days
- When ^{133}Xe backgrounds are large (>1 mBq/m^3), ^{37}Ar is probably better than any radioxenon because of its half-life at times >25 days
  - There are some locations, even distant from man-made sources in which the background concentrations of ^{133}Xe exceed 1 mBq/m^3
  - We expect more examples of large background in the future
- When ^{133}Xe levels are very high (>10 mBq/m^3), ^{37}Ar is always better than any radioxenon because of its lower background in the atmosphere
  - There are a few locations, close to man-made sources in which the background concentrations of ^{133}Xe exceed 10 mBq/m^3
  - We expect more examples of large background in the future

So, why hasn't this been done?

- Xenon-133 was already being measured and is still an excellent signature

- Argon-37 is a difficult isotope to measure
  - The only decay mode measurable is a low-energy (2.8 keV) Auger electron
    - This means that a gaseous internal proportional counter is the only likely means for its measurement
      - A very pure sample is required --- <1 ppm of impurities

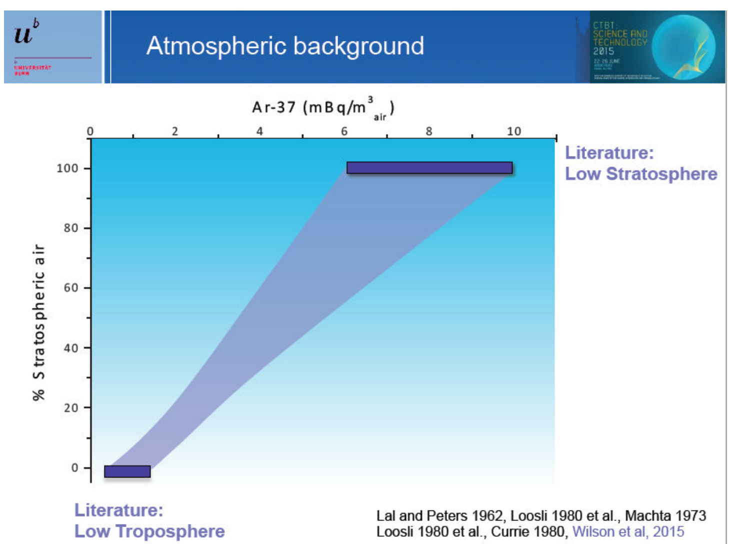
  - The gaseous separation between Ar and N_2 & O_2 isn't as simple as heavy noble gases (i.e., Xe)

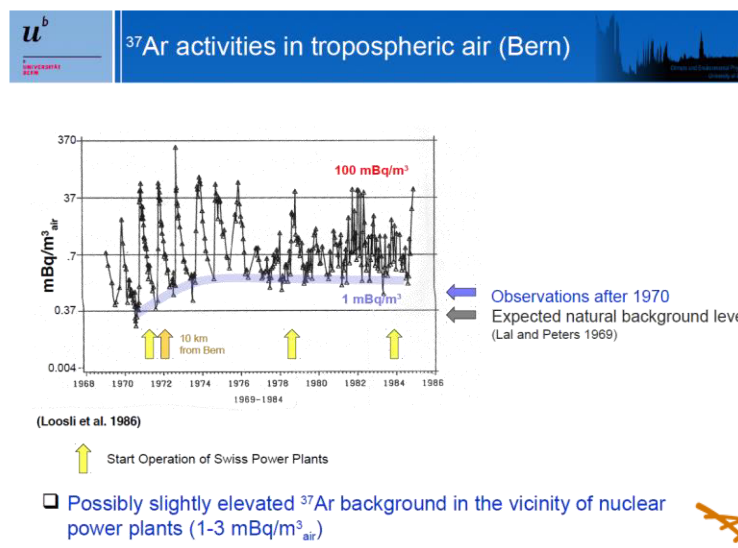
PRINCIPAL RADIATIONS PRODUCED IN THE DECAY OF ^{37}Ar

Decay Mode	Percent of All Decays	Energy of Auger Electrons (keV)	X-ray (keV)
K	81.5	2.823	0.0
L	8.9	0.270	0.0
K	2.7	0.202	2.621
K	5.5	0.201	2.622
M	0.9	0.018	0.0
K	0.5	0.007	2.816

Information needed

- Purtschert, et al. have measured ^{37}Ar concentrations at some locations, even some close to reactors and established a relatively low (few mBq/m^3 range) background (but higher in the subsurface)
  - However, a larger data set and map would be beneficial, as well as investigations into release details





Purtschert, Riedmann, Raghoo, 2014

Fusion between Xe and Ar data

- The production of ^{37}Ar and ^{133}Xe come from the same root process (i.e., a nuclear explosion), there are similarities and differences (see presentations by Lowrey) and
  - Releases are similar since they are both noble gases, though some differences in release times might occur
  - The amount of calcium affects the signal size for ^{37}Ar
  - Xe-133 has significant other man-made (and large) sources
- Hence, if there are correlated detections of both isotopes, the confidence that an event is of a nuclear nature would increase significantly

Conclusions and next steps

- Argon-37 may be a better signature than Xe-133 in some cases, especially where backgrounds of Xe-133 are high or more than a few months have passed since an explosion

- Perform a background measurement campaign with defined scientific goals set by the community. Possible areas of concentration include:
  - Measurements at IMS stations
  - A model of nuclear reactors as a source
  - Continued source term models
  - Investigate ^{39}Ar as a background

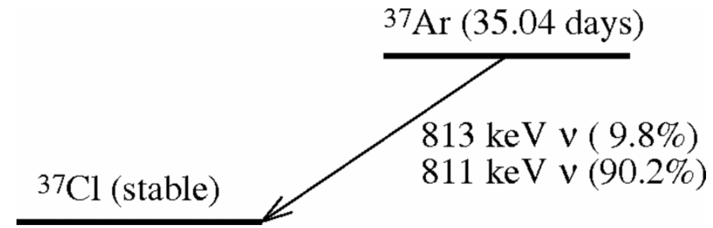
- Develop prototype field sensors and concentrate on the following issues (much can be leveraged from OSI systems)
  - Minimization of the use of consumables
  - Increasing sample size to x10 OSI systems
    - Possibly using IMS xenon systems “waste air”
  - Ensuring detector backgrounds are kept low

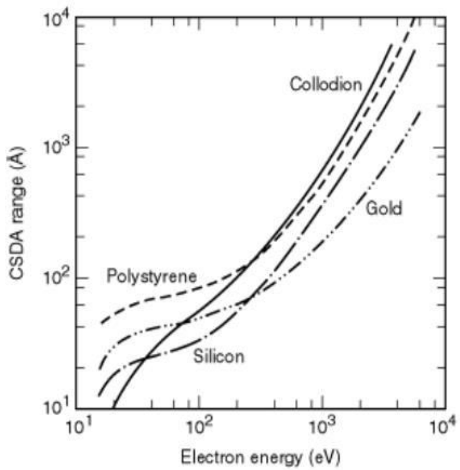
- Backup slides

Argon-37 for the IMS

- While Argon-37 is already a signature for OSI, there are many reasons to examine its use for the IMS
  - The production cross section and half-life makes the isotope perfect
    - Inert, and likely to be emitted from nuclear testing
    - Long half-life (35 d)
    - May help solve background issues associated with man-made processes
  - Correlation to OSI measurements may be needed

Decay of ^{37}Ar





Principal Radiations Produced in the Decay of ^{37}Ar

Decay Mode	Percent of All Decays	Energy of Auger Electrons (keV)	X-ray (keV)
K	81.5	2.823	0.0
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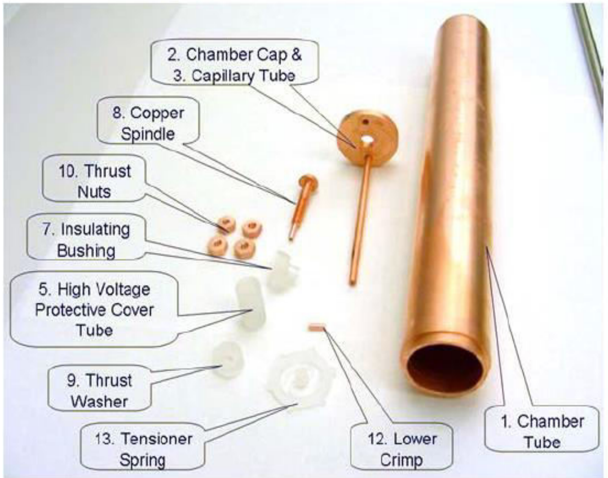
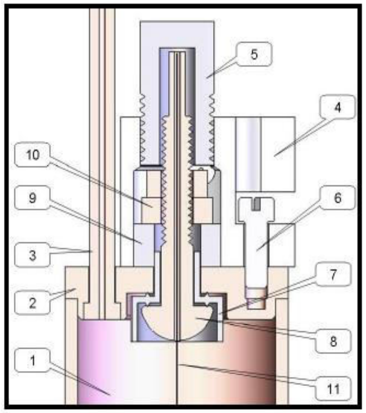
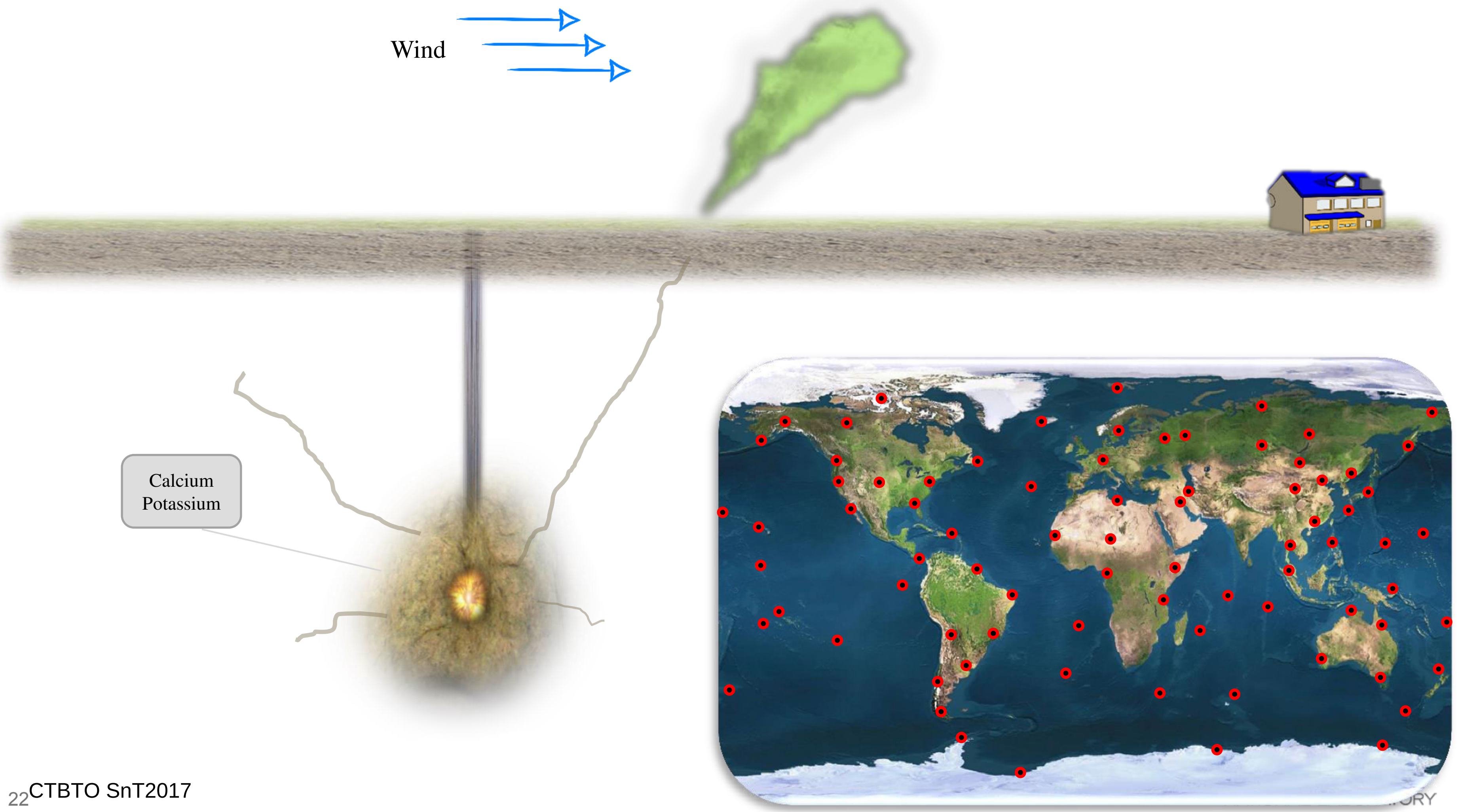
Attenuation length for
2.8 keV Auger electron
~0.01 cm in Ar





Gas release into the atmosphere and detection in the International Monitoring System (IMS)



Detection sensitivity

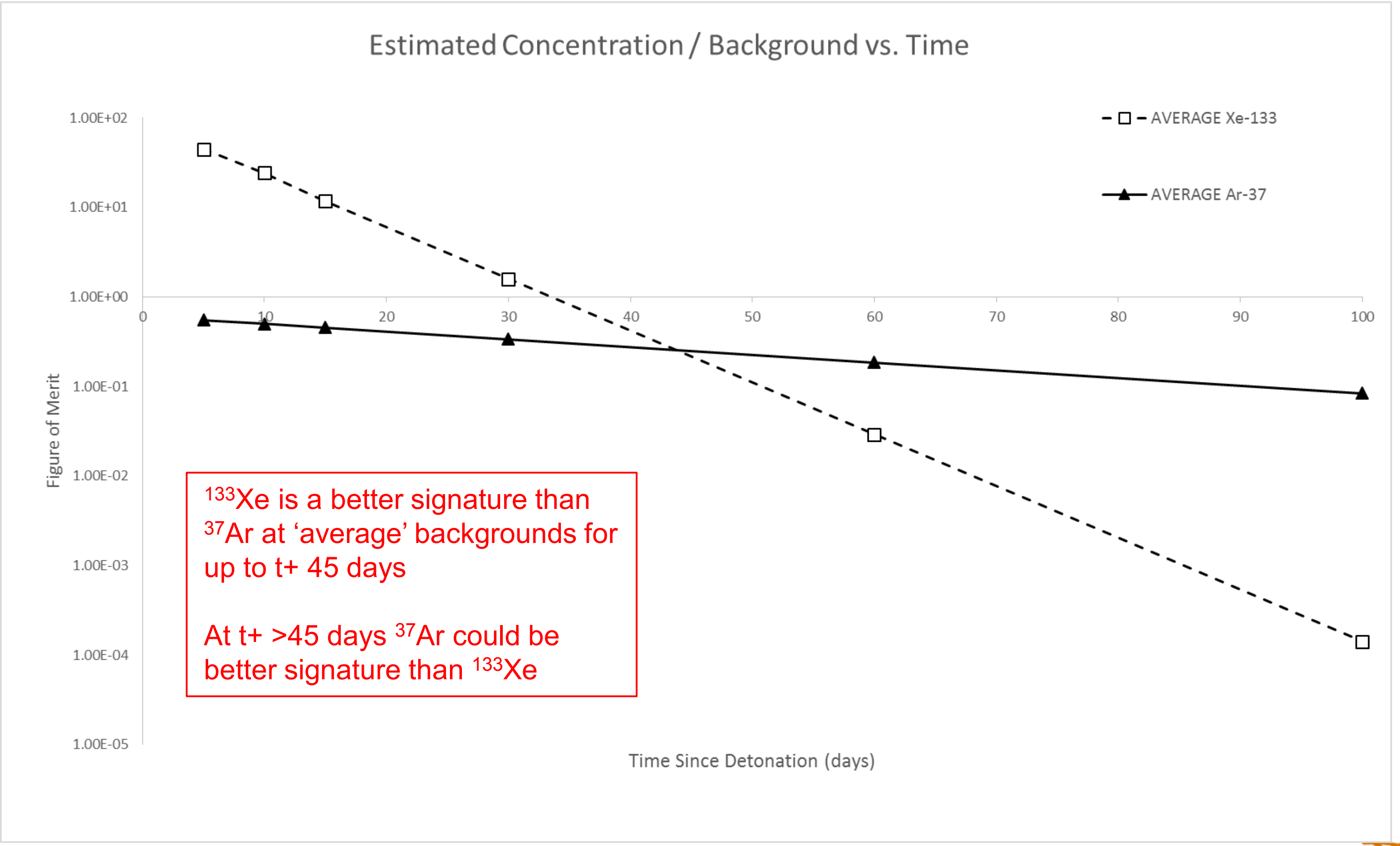
- Current goals of OSI systems reach MDCs in the range of 10 mBq/m^3 (12 hr count)
  - Detection levels depend on
    - The count time (t)
    - The amount of sample collected (V)
    - The background (n_b)

$$MDC = \frac{4.66 \times \sqrt{n_b \times t}}{V \times \eta \times \varepsilon \times I \times t}$$

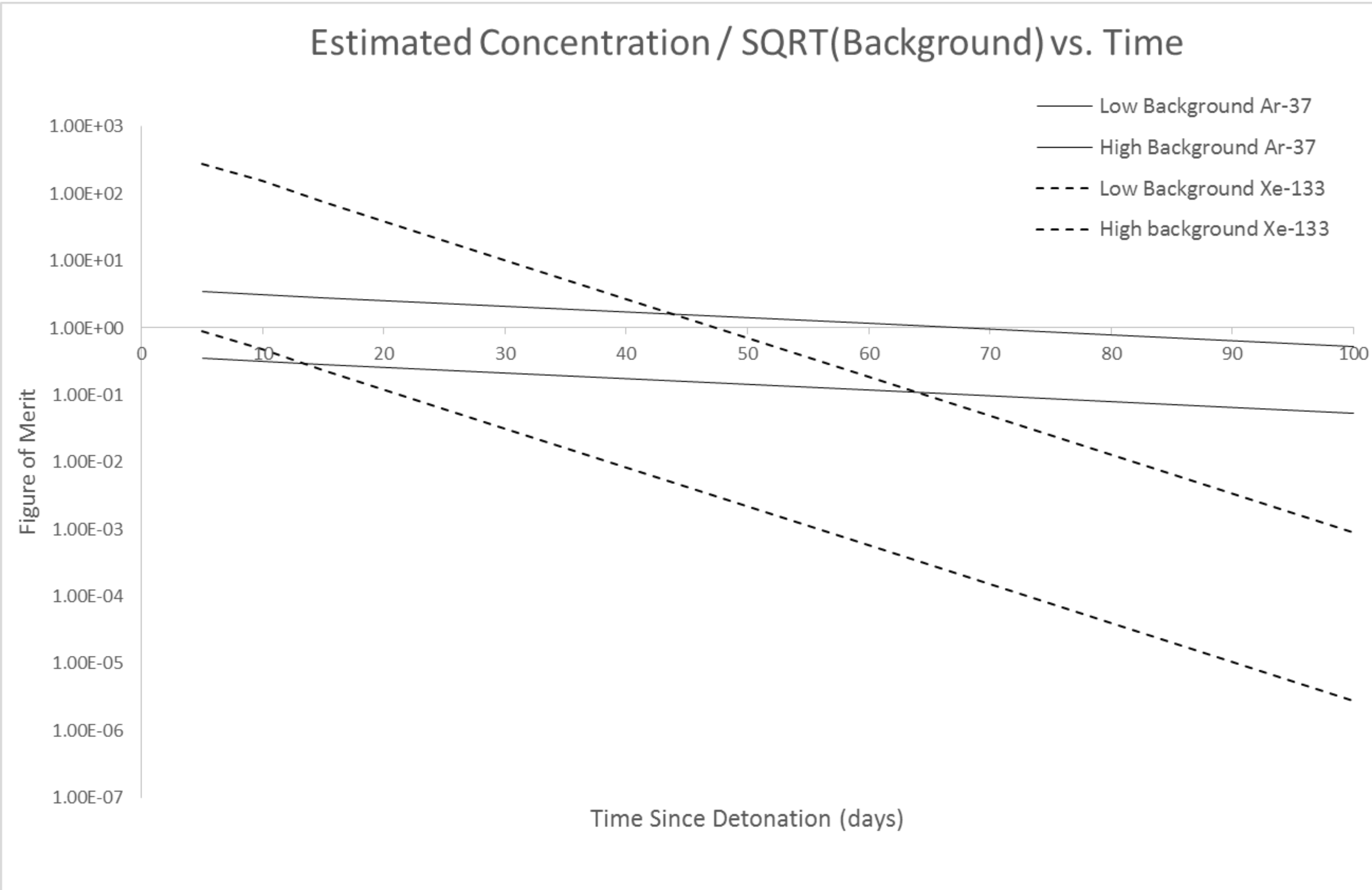
- Lab-based detection sensitivities for *14-day* counts are ~0.05 mBq/m^3, for a 1 liter Ar sample (!)

- For an IMS measurement, increasing the sample size (x4 or more?) and counting time (x4) over the OSI field system
  - These improvements for a 48-hr count could drive the MDC to ~1 mBq/m^3 for a field system or better?
  - Exploring the maximum sample size may be the best route

Average case FOM



Alternate FOM (using SQRT of background)



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History of ^{37}Ar measurement technology in the last 20 years

- A significant amount of R&D has been done by the University of Bern since the 1970s
  - The main applications have been for subsurface water tracer studies
  - More recent work by Purschert has been focused on OSI applications
- China NINT
  - MARDS field system (2008-Present)
- PNNL
  - ^{37}Ar field system (2015-Present)
  - ^{37}Ar lab capability (2009-Present)