Ultrapure gases – from the production plant to the laboratory

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Outline

- Motivation
- Measurement techniques
- Gas purification techniques
  - Adsorption
  - Distillation
- Survey of gases from european suppliers
- Test of whole delivery chain
- Conclusion

Motivation

- Inert gases are used in low-level nuclear and particle physics:
  - for cleaning / as blankets
  - liquefied: for shielding / as scintillator
- Count-rates down to event/year-scale require highest purity!
- Usually orders of magnitude cleaner than commercial specifications (6.0, <1ppm)
- Task: Removal of dissolved radioactive impurities (e.g. ^{222}Rn, ^{85}Kr, ^{39}Ar)

Example: Purity requirements in nitrogen for BOREXINO

	Required purity	
^{222}Rn	<3 atoms/m^3	<7 μBq/m^3
Krypton	<0.14 ppt	^{85}Kr: <0.2 μBq/m^3
Argon	<0.36 ppm	^{39}Ar: <0.6 μBq/m^3

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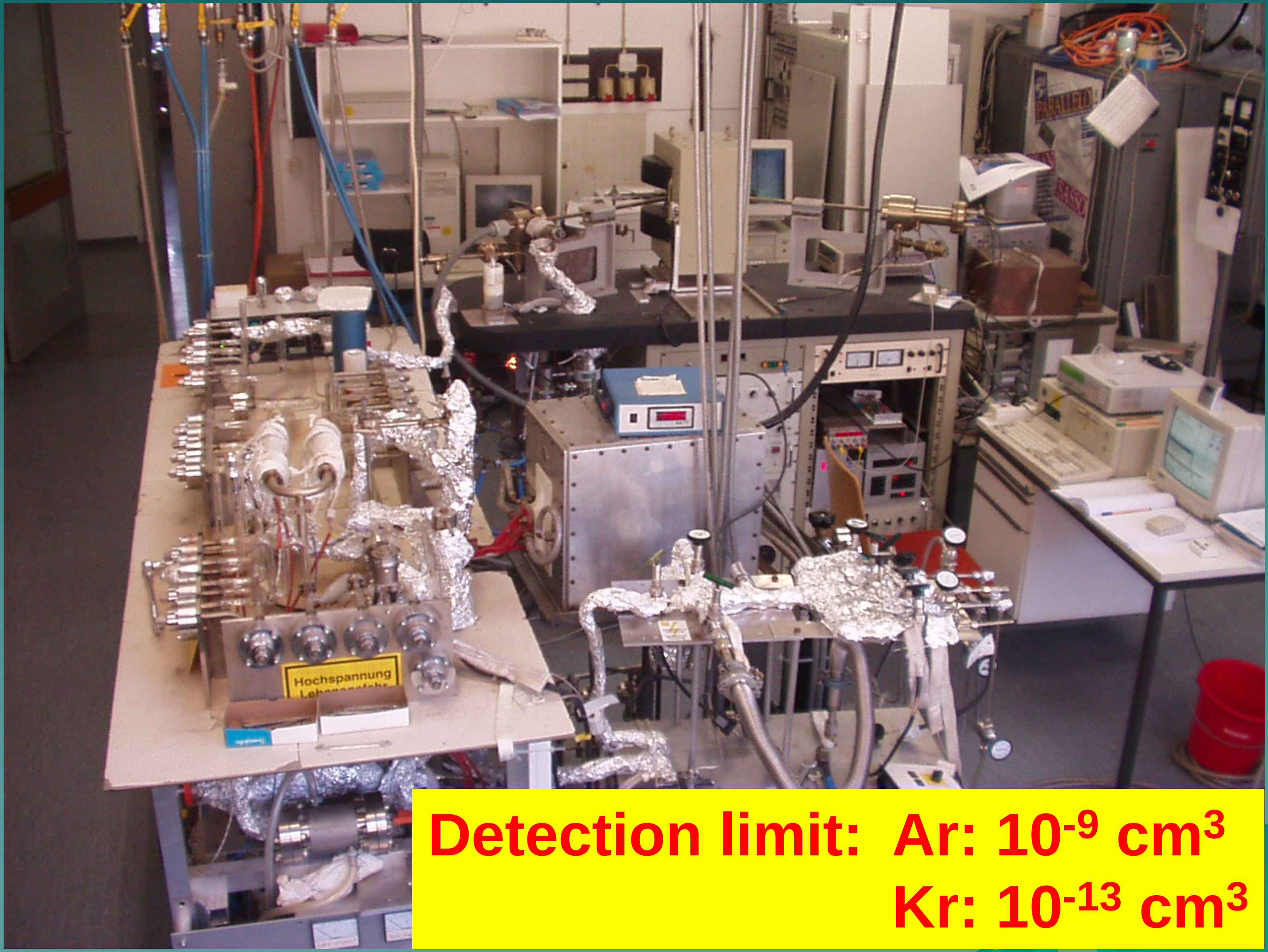
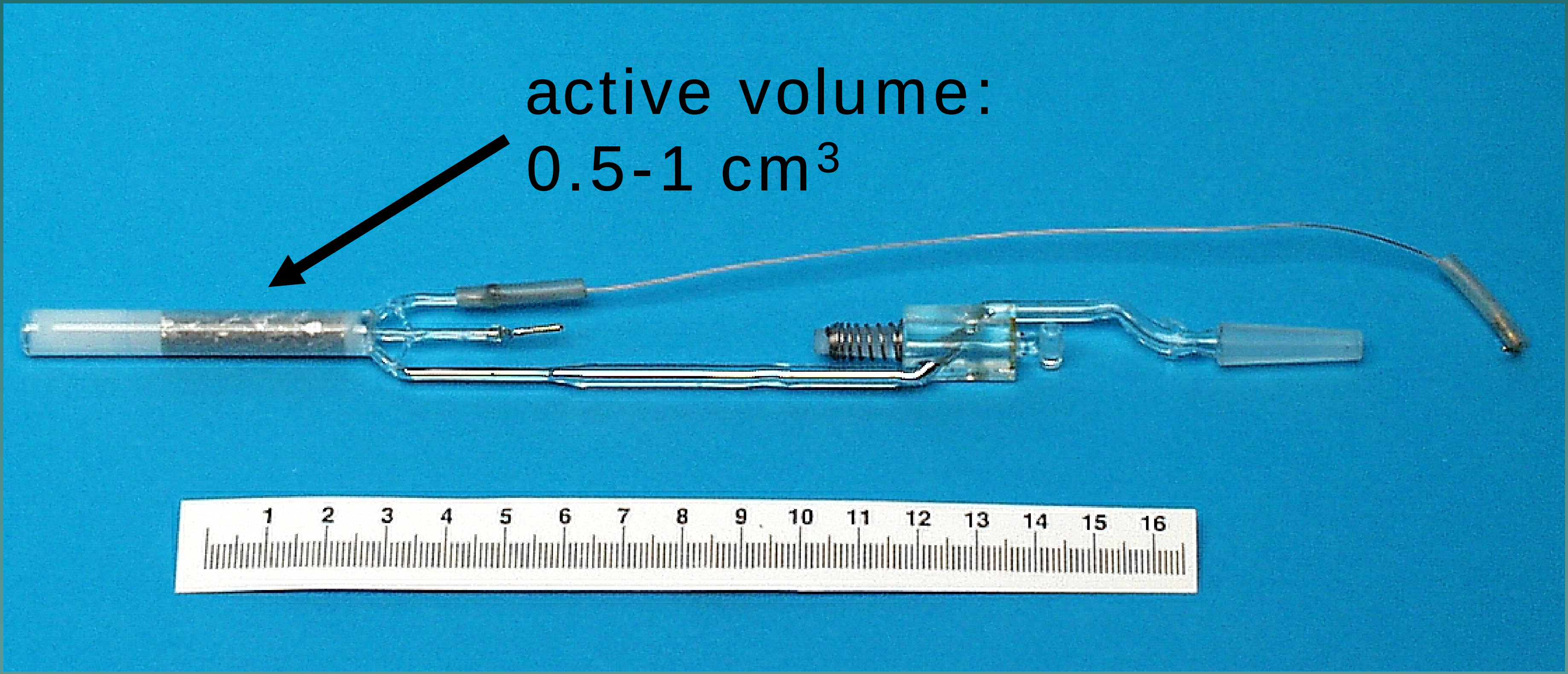
Noble gas mass spectrometer



Detection limit: Ar: 10^{-9} cm^3
Kr: 10^{-13} cm^3

Low background proportional counter



- ^{222}Rn detection limit (including background from counter-filling): ~15 atoms !

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N_2 purification from Ar/Kr/Rn by adsorption techniques

→ H. Simgen: „Adsorption techniques for gas purification“ LRT 2004, Sudbury

Conclusions:

- Ar/N_2 separation impossible
- Kr/N_2 separation difficult
- Rn/N_2 separation „easy“

Main issue: re-contamination

N_2 purification plant @ LNGS

^{222}Rn in N_2 before purification: ~50 μBq/m^3 (STP)

Adsorber mass: 2.1 kg
Flow-rate: up to 100 m^3/h (STP)

^{222}Rn in N_2 after purification: <0.5 μBq/m^3 (STP)

Argon purification from ^{222}Rn

- Argon: Cryogenic shield in GERDA
- ^{222}Rn purity requirement: $<0.5\ \mu Bq/m^3$
- Concerning adsorption:
  - N_2 and Ar behave similar (both very different from radon)
  - But: $T(LAr) = T(LN_2) + 10K$
- Technical difficulty: Argon can easily freeze!

Argon purification from ^{222}Rn

^{222}Rn in Ar before purification: ~200 $\mu Bq/m^3$ (STP)

Adsorber mass: 0.15 kg

Flow-rate: up to 20 m^3/h (STP)

^{222}Rn in Ar after purification: <0.5 $\mu Bq/m^3$ (STP)

Air separation plant

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Ar/Kr concentration in nitrogen from different sources

Description	Ar [ppm]	Kr [ppt]
Linde AG (Worms)	0.4	6
Westfalen AG (Hörstel)	0.03	7
SOL (Mantova)	0.2	9
Goal	**0.36**	**0.14**

Problems with sampling

- Gaseous samples (1-5 ccm) have bad volume/surface ratio

  $\Rightarrow$ high risk of contamination

- Thus: Liquid samples (240 l LN_2)

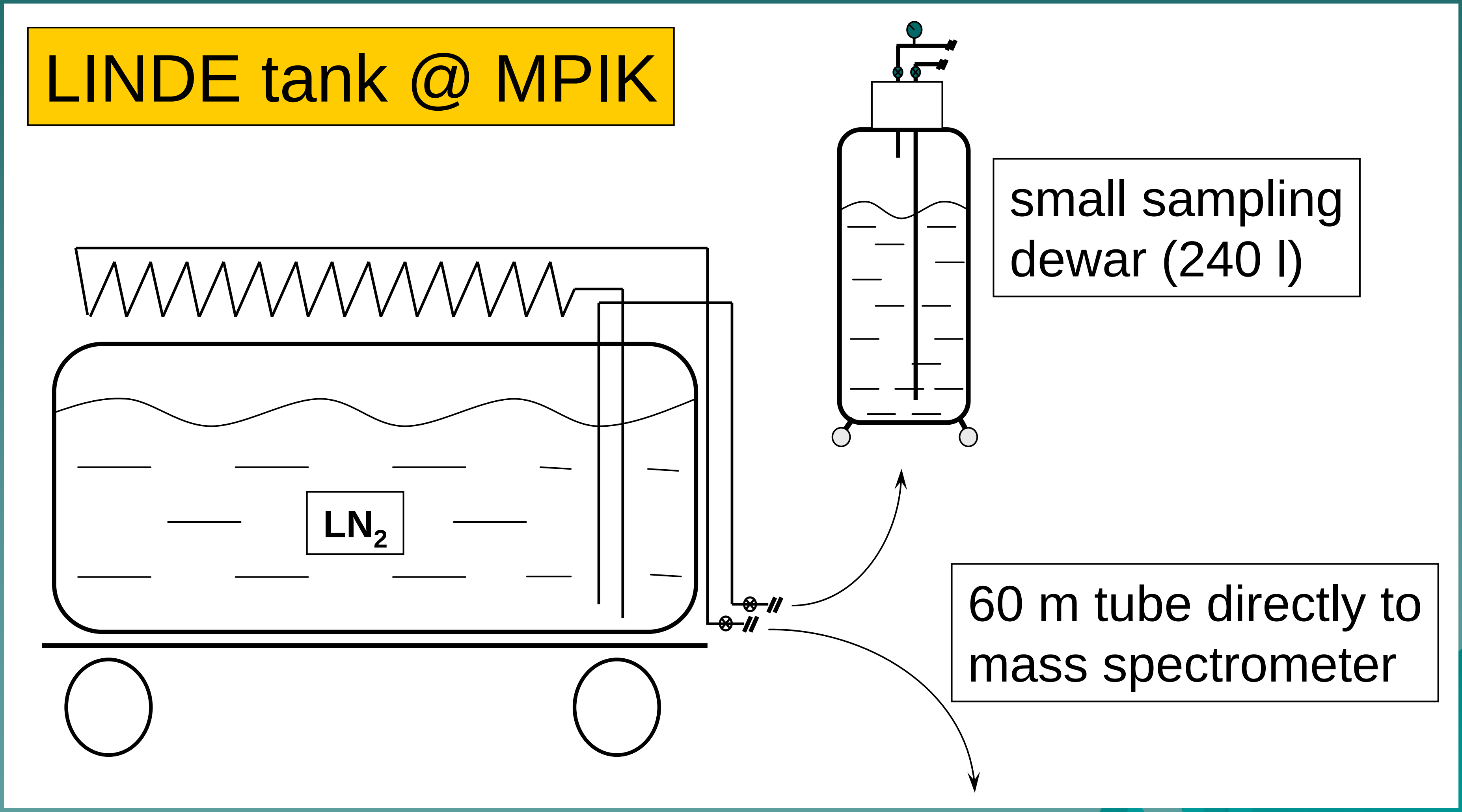
Ar/Kr concentration in nitrogen from different sources

Description	Ar [ppm]	Kr [ppt]
Linde AG (Worms)	0.4	6
Westfalen AG (Hörstel)	0.03	7
SOL (Mantova)	0.2	9
Linde AG (Worms)	0.04	4
Westfalen AG (Hörstel)	0.0005	0.06
SOL (Mantova)	0.005	0.04
Goal	**0.36**	**0.14**

Problems with sampling

- Even liquid samples must be prepared very carefully
- No appropiate sampling port for small-scale samples available at Linde plant
- Thus: Test-tank installed @ MPIK

Ultrapure nitrogen from LINDE



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Linde AG (Worms)	0.04	4
Westfalen AG (Hörstel)	0.0005	0.06
SOL (Mantova)	0.005	0.04
Linde AG (Worms)	0.013	0.1
Goal	**0.36**	**0.14**

Search for ultrapure N_2 on the market - Summary

- Air separation plants produce N_2 of very high purity
- Contaminations are brought in by transport / refilling / storage
- Conclusion (for BOREXINO):

  Full delivery chain (from the production plant to the laboratory) needs to be tested under realisitic conditions.

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SOL

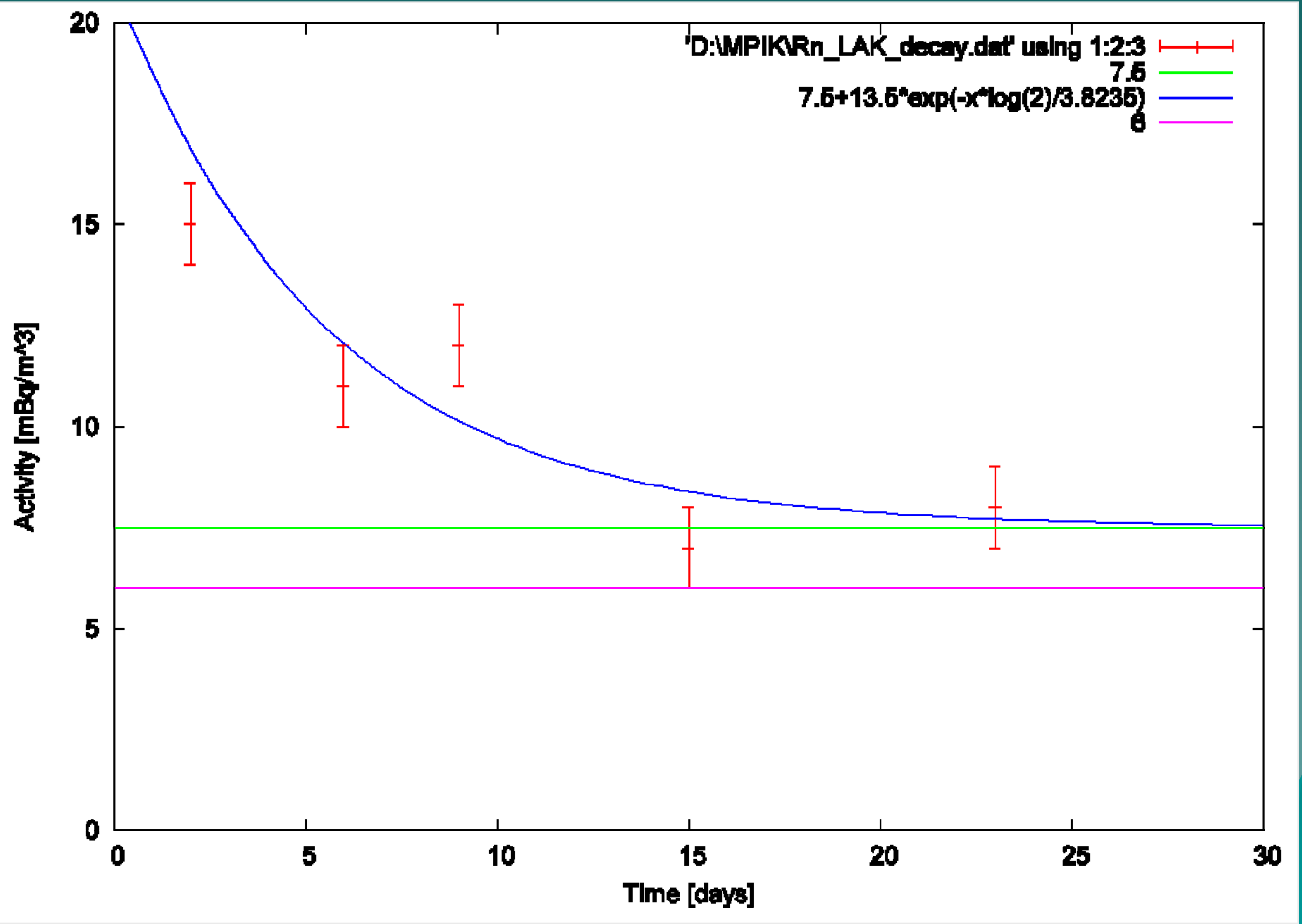

SOL

Total ^{222}Rn budget inside tank: 65 mBq

Converted in ^{222}Rn concentration: 6 μBq/m^3 (STP)


SOL
SOLGROUP

Time dependency of ^{222}Rn (tank 90% full)



Results on Argon / Krypton

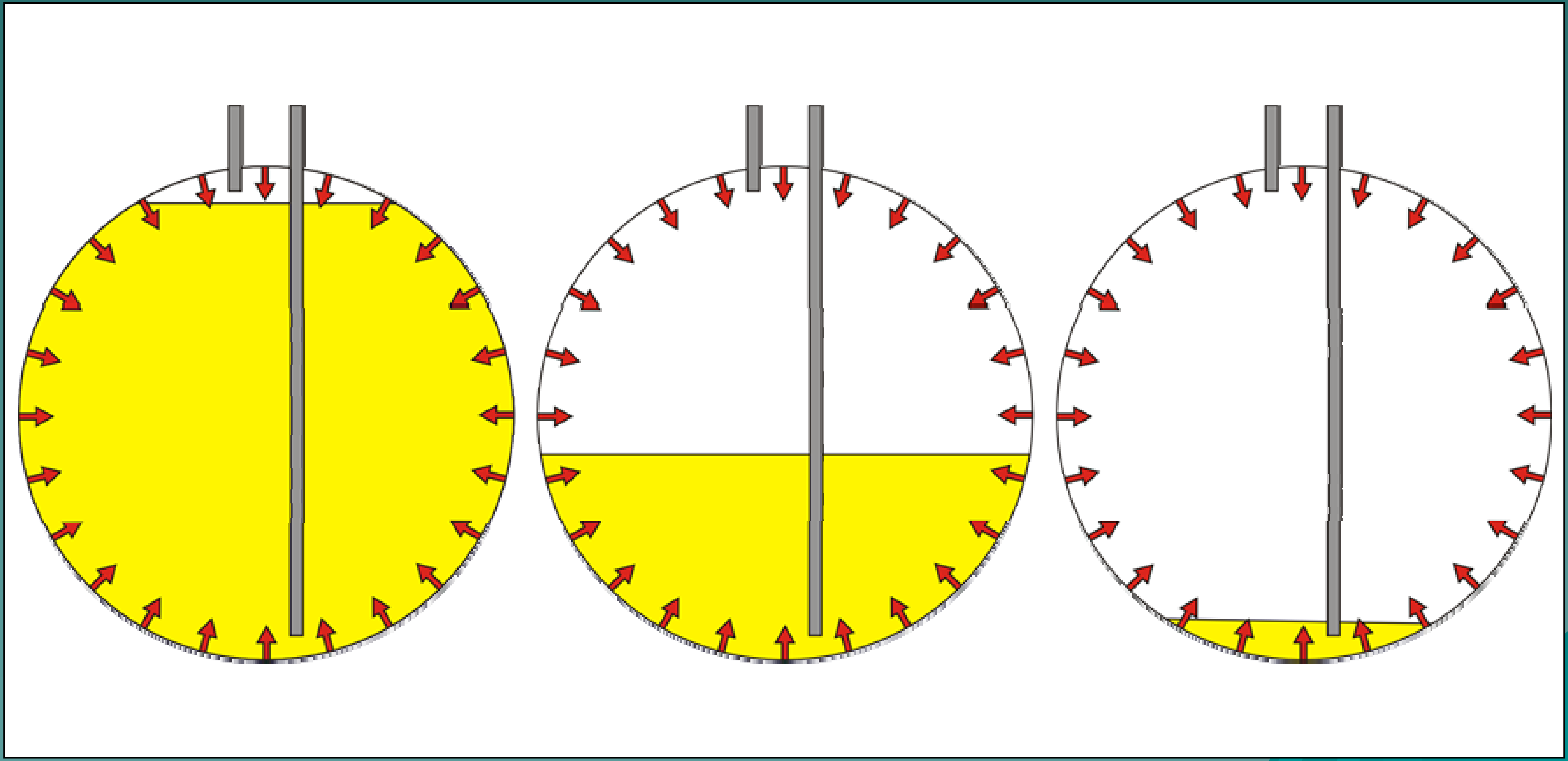
Conditions	C_{Ar} [ppb]	C_{Kr} [ppt]
In liquid phase	12 ± 2	0.02 ± 0.005
In gas phase	7 ± 1	0.006 ± 0.004
BOREXINO goal	**<360**	**<0.14**

- No dependency on filling level of tank
- Gas phase concentration always lower than liquid phase concentration
- Supply chain succesfully tested!

^{222}Rn dependency on filling level

Conditions	C_{Rn} [μBq/m^3] (STP)	
	Liquid phase	Gas phase
V_{LN2} ~14 m^3 (90 % filled)	8 ± 1	7 ± 1
V_{LN2} ~6 m^3 (38 % filled)	10 ± 1	26 ± 3
V_{LN2} ~3 m^3 (19 % filled)	11 ± 1	30 ± 3
V_{LN2} ~0.8 m^3 (5 % filled)	38 ± 5	42 ± 9
V_{LN2} ~200 l (almost empty)	237 ± 13	47 ± 3

^{222}Rn in gas phase / liquid phase
An attempt of interpretation



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Conclusion

- ^{222}Rn problem can be solved „in the lab“ by adsorption techniques
- Distillation is better approach for Ar/Kr purification
- Final Ar/Kr concentration strongly dependent on storage / refilling / transport
- Can be controlled (tank design / refilling procedure)
- Final ^{222}Rn concentration determined by storage tank