Alpha energy adsorption in filters and its potential to degrade air monitoring accuracy.

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Alpha energy absorption in filters

1. Some basic principals
2. Investigation of spectra obtained from calibrated ^{239}Pu aerosols collection by filters
3. Performance with heavily degraded spectra
4. Information available in published papers and reports

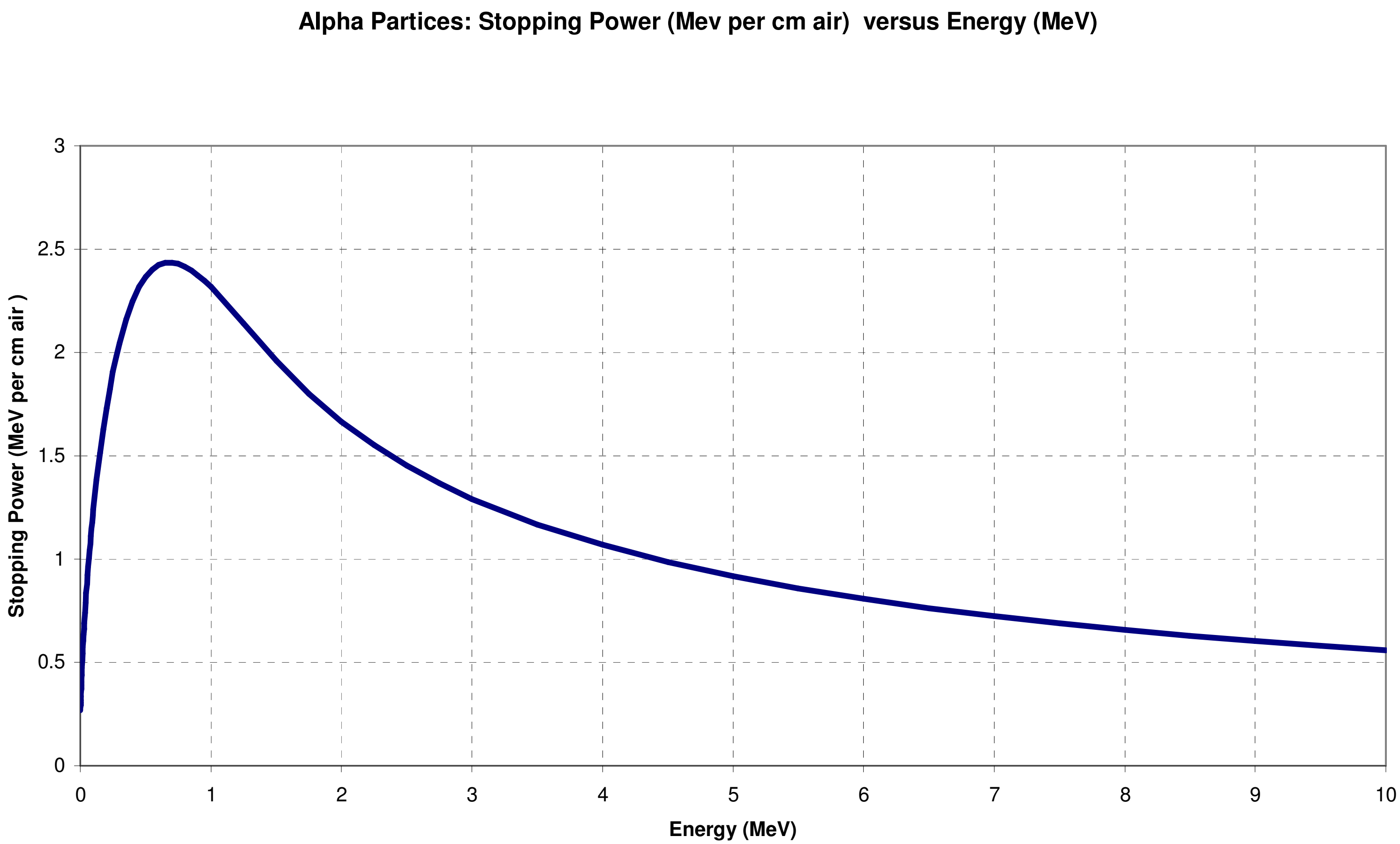
Alpha Energy Degradation can occur

1. In the active aerosol particle (diameters > 1 μm)
2. In the filter material
3. In other particulates collected by the filter
4. In the air gap between filter and detector
5. In the window of the detector

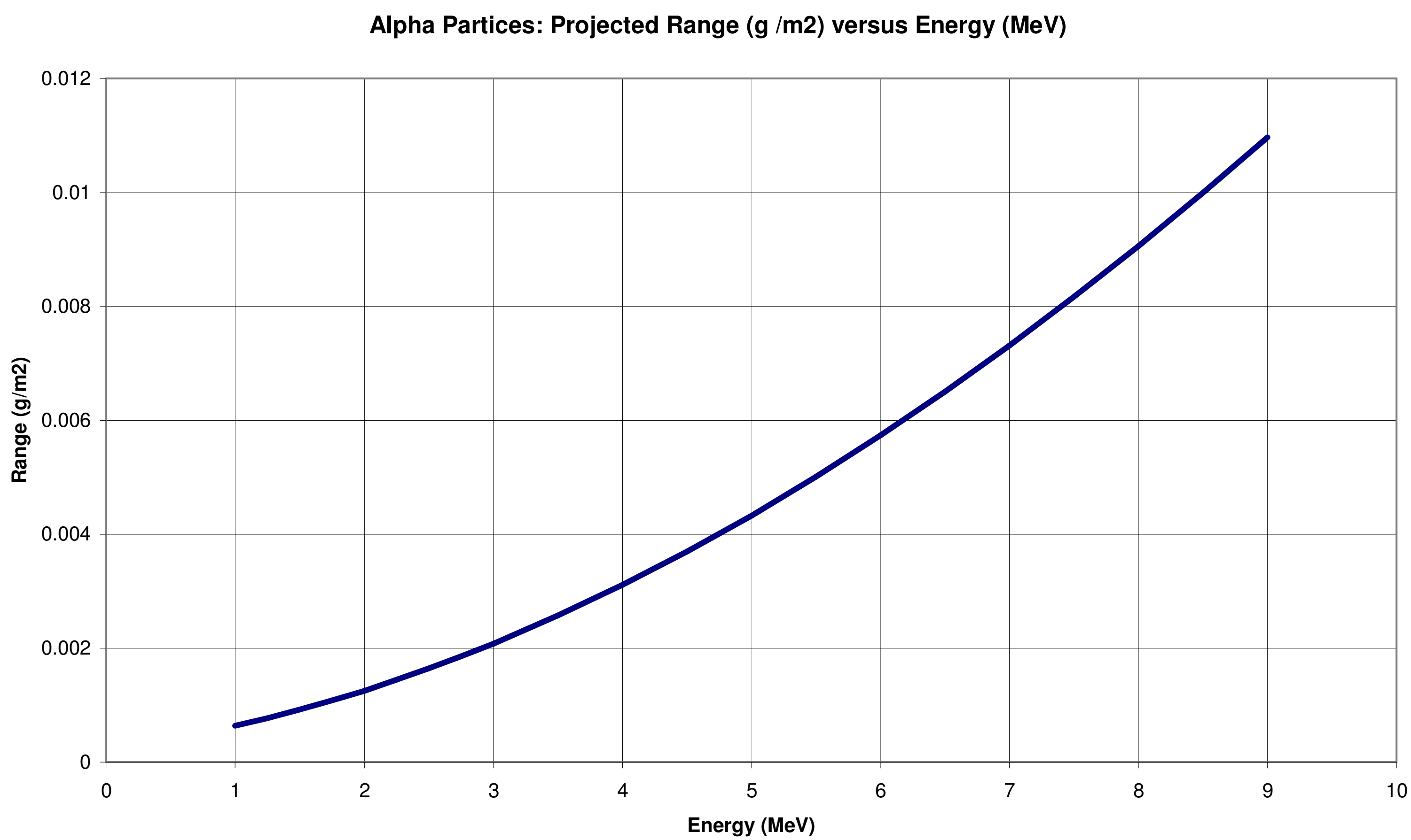
Only the last two are taken into account when calibrating with a reference source

Alpha Particle
Stopping Power and Range

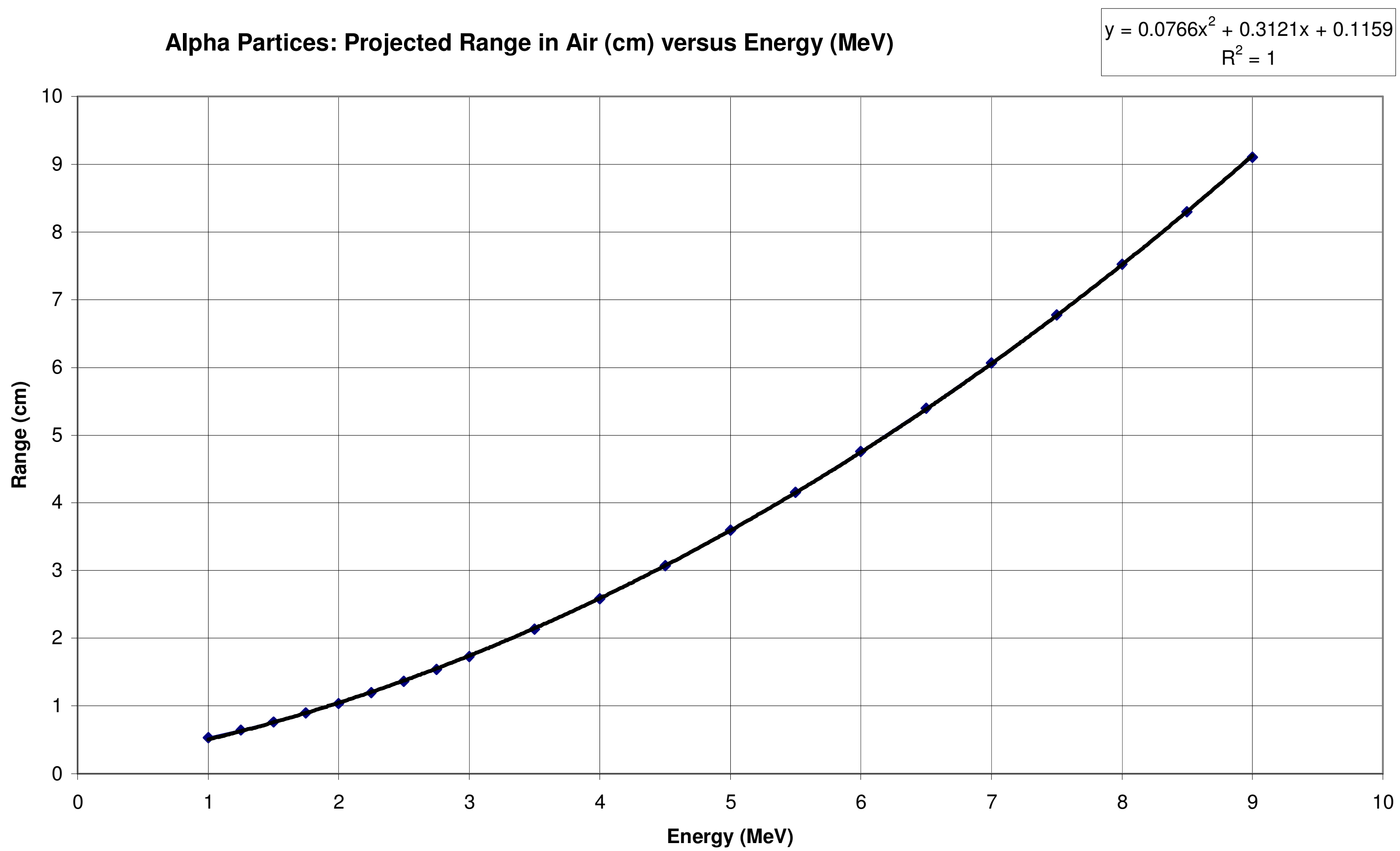
Stopping Power of Alpha Particles (MeV/cm) in Air versus Energy



Range of Alpha Particles (mg/cm^2) versus Energy



Range of Alpha Particles (cm) in Air versus Energy



Approximate ranges of alpha particles

Alpha Energy (MeV)	Material (mg/cm^2)	Air (mm)	Water (μm)
4.0	3.1	26	31
4.5	3.7	30	37
5.0	4.3	36	43
5.5	5.0	42	50
6.0	5.7	48	57
7.0	7.3	61	73
7.5	8.2	68	82
8.0	9.1	75	91
8.5	10	83	100

Mass concentrations of aerosols (PM10) found in air

Typical Urban sites:

Median concentration = 0.05 mg/m^3
(typical range 0.01 to 1mg/m^3)

Waste Isolation Pilot Plant (WIPP):

Median concentration = 5mg/m^3
(range 0.3 to 30mg/m^3)

Time to accumulate significant dust loadings

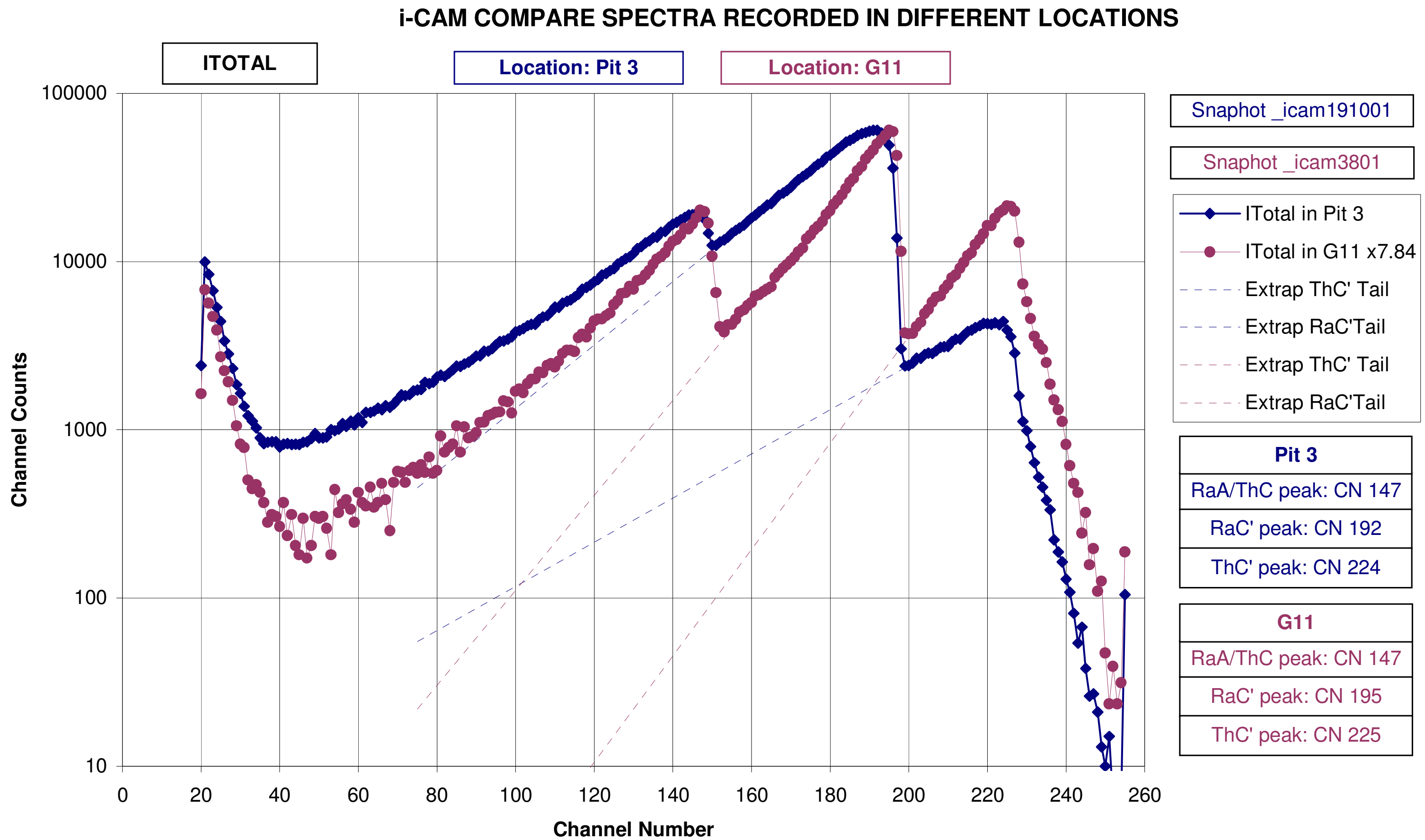
- If air containing 0.05 mg/m^3 of particulates is drawn at a rate of 40 LPM through a 25 mm filter then in 24 hours the filter will accumulate a layer equal to 12% of the range of a 5.5 MeV alpha

The effects of accumulated particulates

- The impedance to flow increases as particulates accumulate
- The efficiency of collecting aerosols tends to increase as the particulates accumulate.
- For this reason the alphas emitted by freshly collected activity may sometimes suffer less energy degradation with a dirty than with a clean filter.
- However the energy of alphas emitted by activity already collected on the filter will obviously tend to be degraded by the further collection of additional particulates

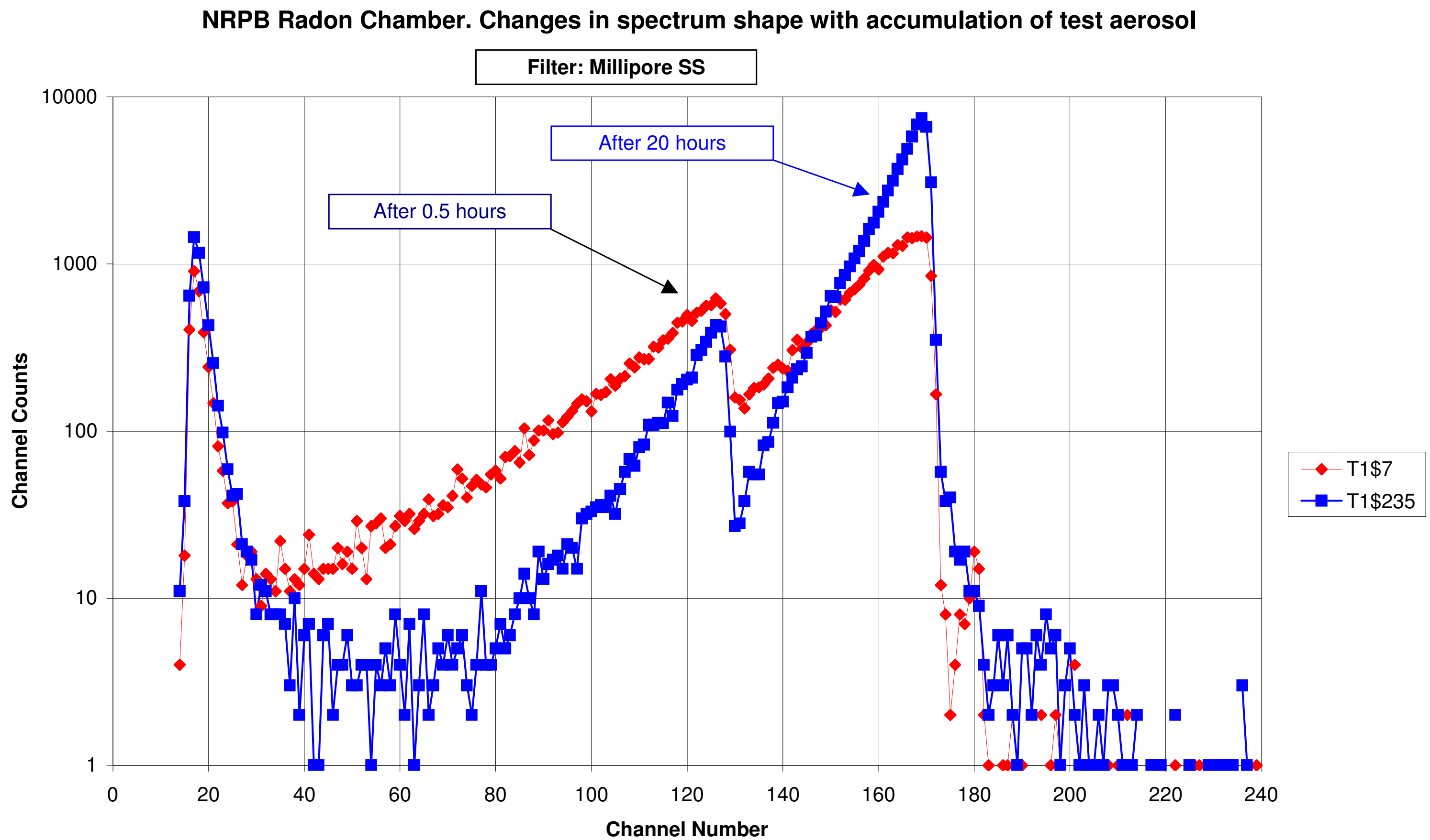
Some differences in Alpha Spectrum Shapes

Difference in spectrum shape with location



The next slide shows an example of improving alpha spectrum resolution with increasing accumulation of aerosol

An example of improved energy resolution with accumulated aerosol

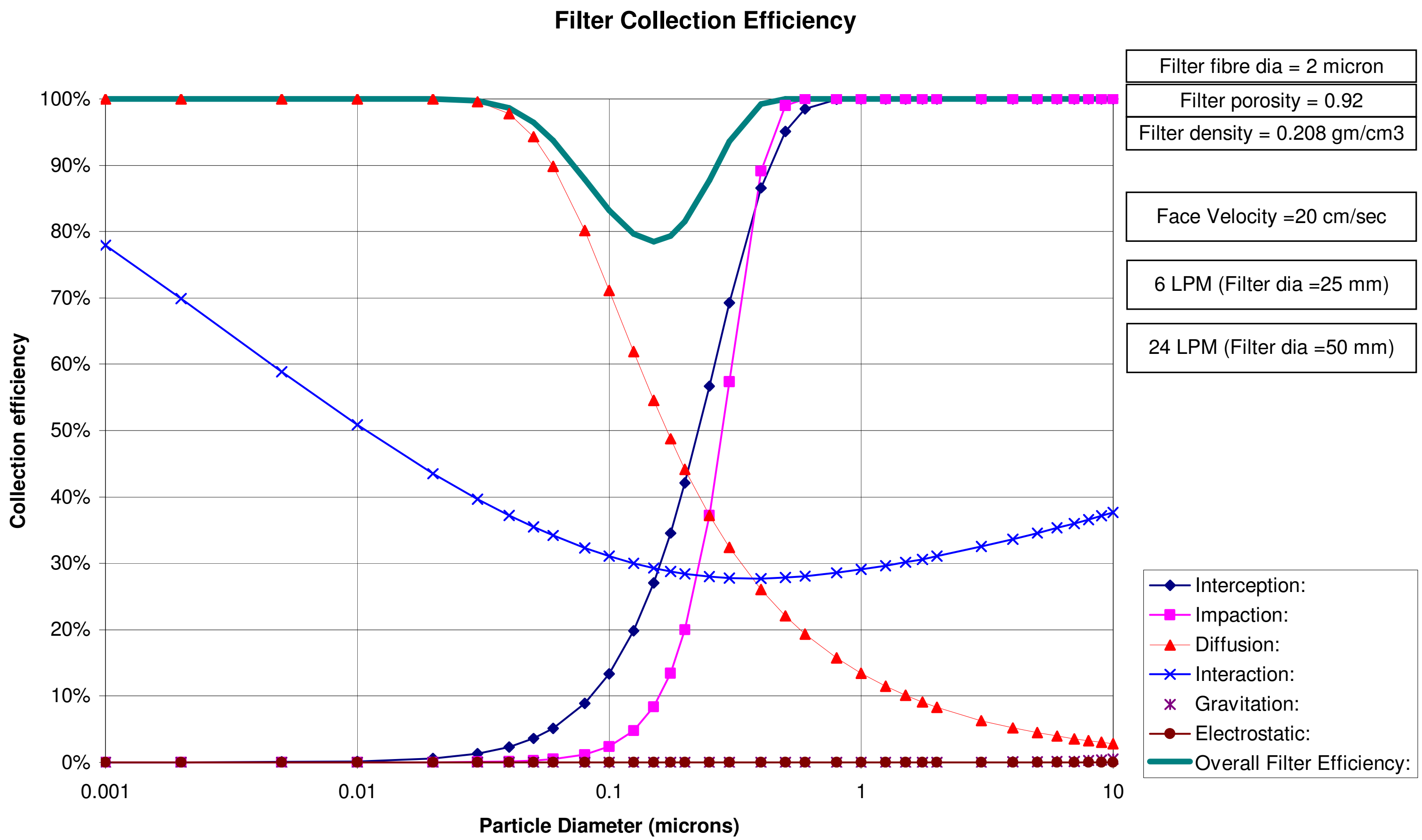


However note that this is radon progeny spectrum.

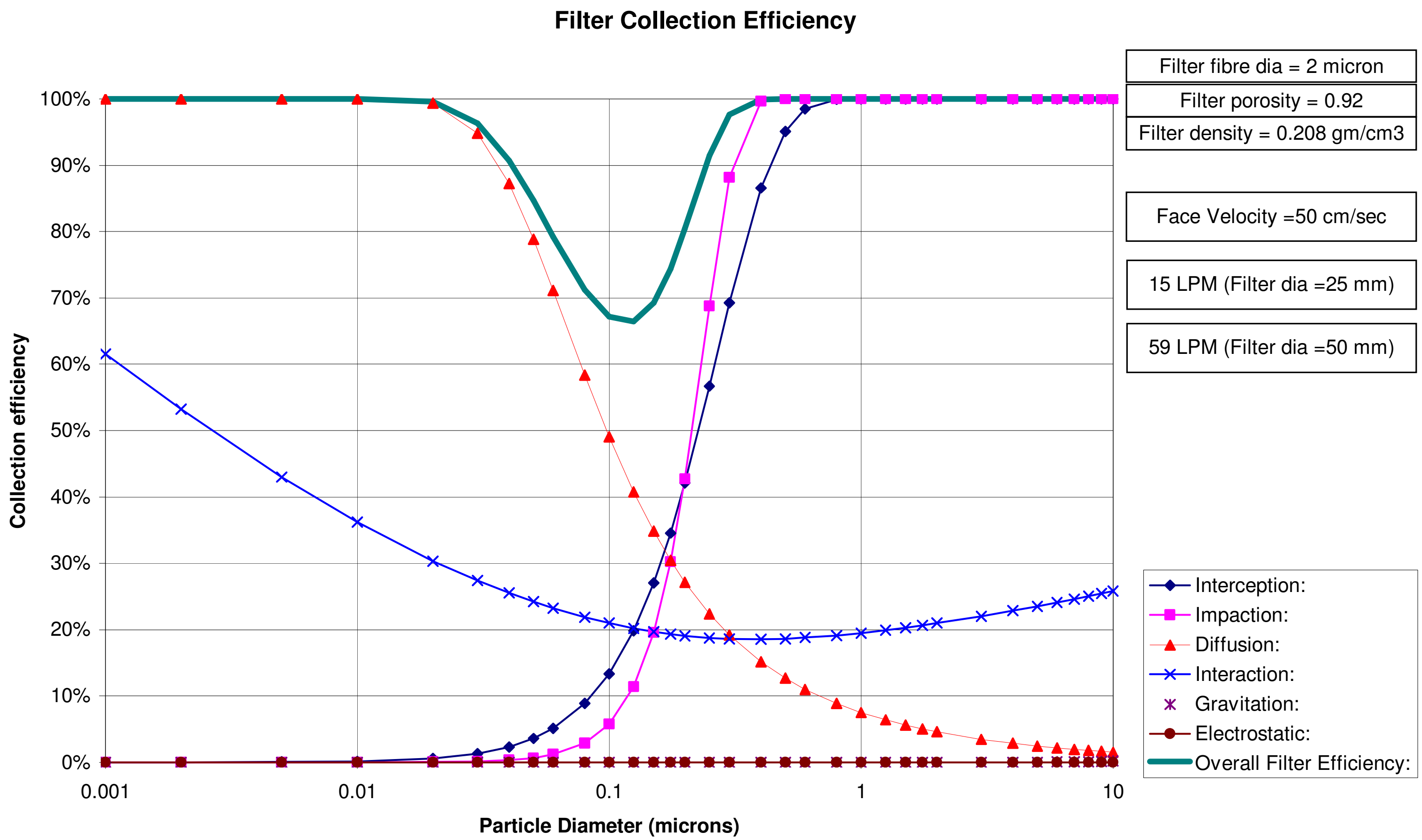
- The short half life of RaA and RaC′ means that most of their alphas are emitted by relatively freshly collected aerosols
- This large change in resolution should not be taken as typical

Filtration Mechanisms

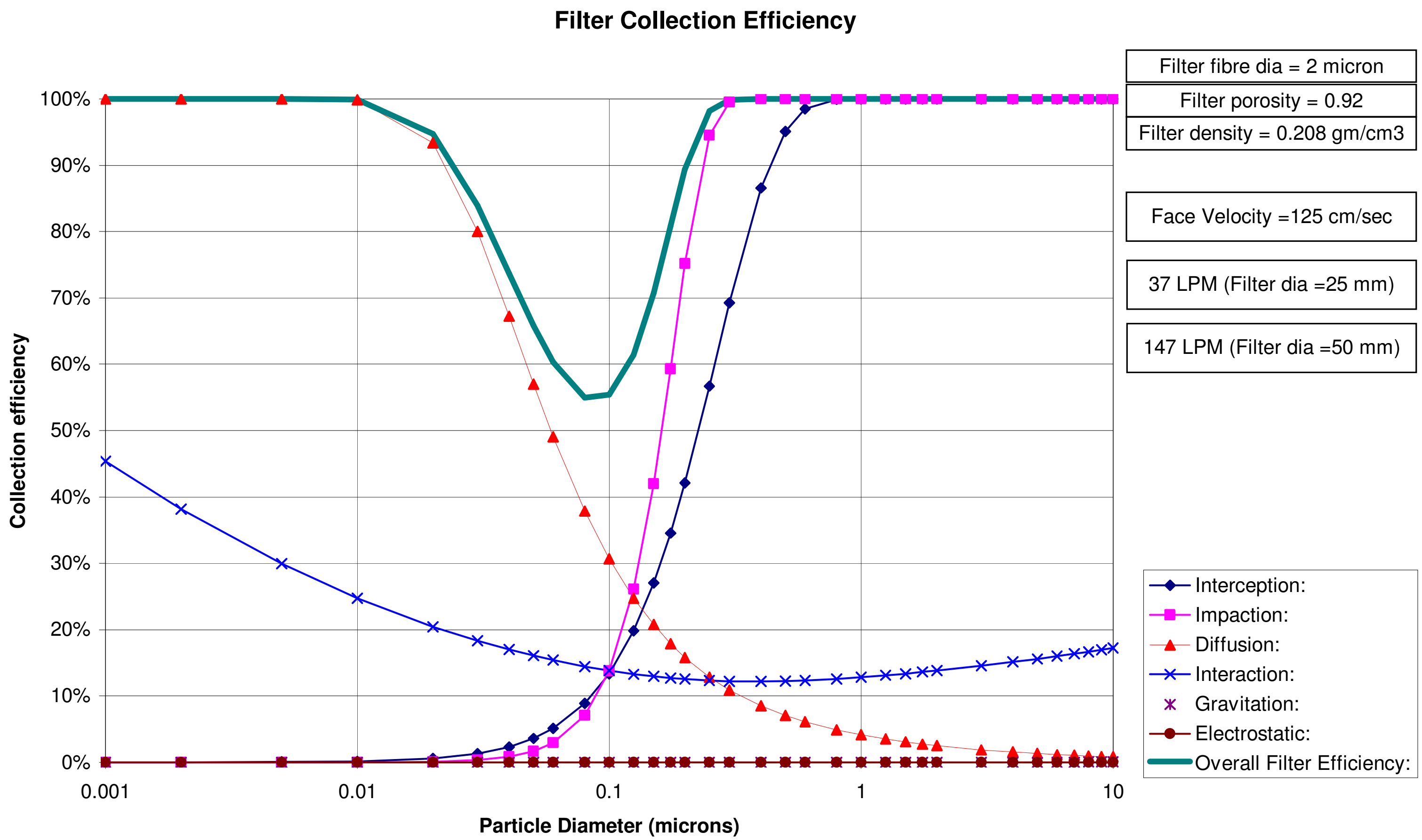
Filtration Efficiency
Contributing collection mechanisms versus aerosol particle size
Face Velocity = 20 cm/s
(6 LPM with 25mm diameter filter, 24 LPM with 50 mm diameter filter)



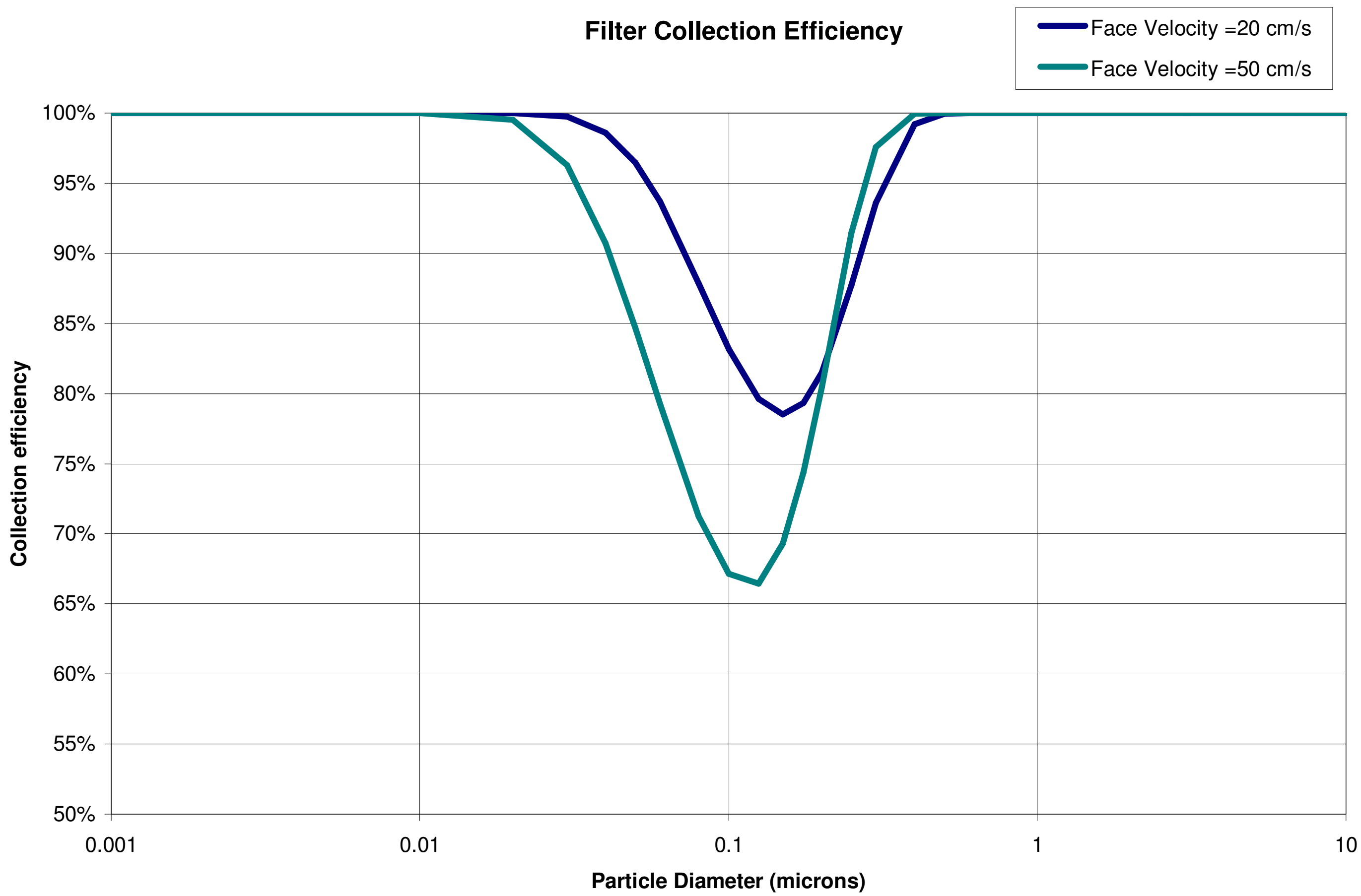
Filtration Efficiency
Contributing collection mechanisms versus aerosol particle size
Face Velocity = 50 cm/s
(15 LPM with 25mm diameter filter, 60 LPM with 50 mm diameter filter)



Filtration Efficiency
Contributing collection mechanisms versus aerosol particle size
Face Velocity = 125 cm/s
(37 LPM with 25mm diameter filter, 147 LPM with 50 mm diameter filter)



Filtration Efficiency at two face velocities



Spectra from calibrated radioactive aerosols collected on two types of filter

^{239}Pu and ^{137}Cs aerosols

Activity Median Aerodynamic Diameters

- AMAD =0.4µm
- AMAD = 4 µm

Radon (^{222}Rn) progeny

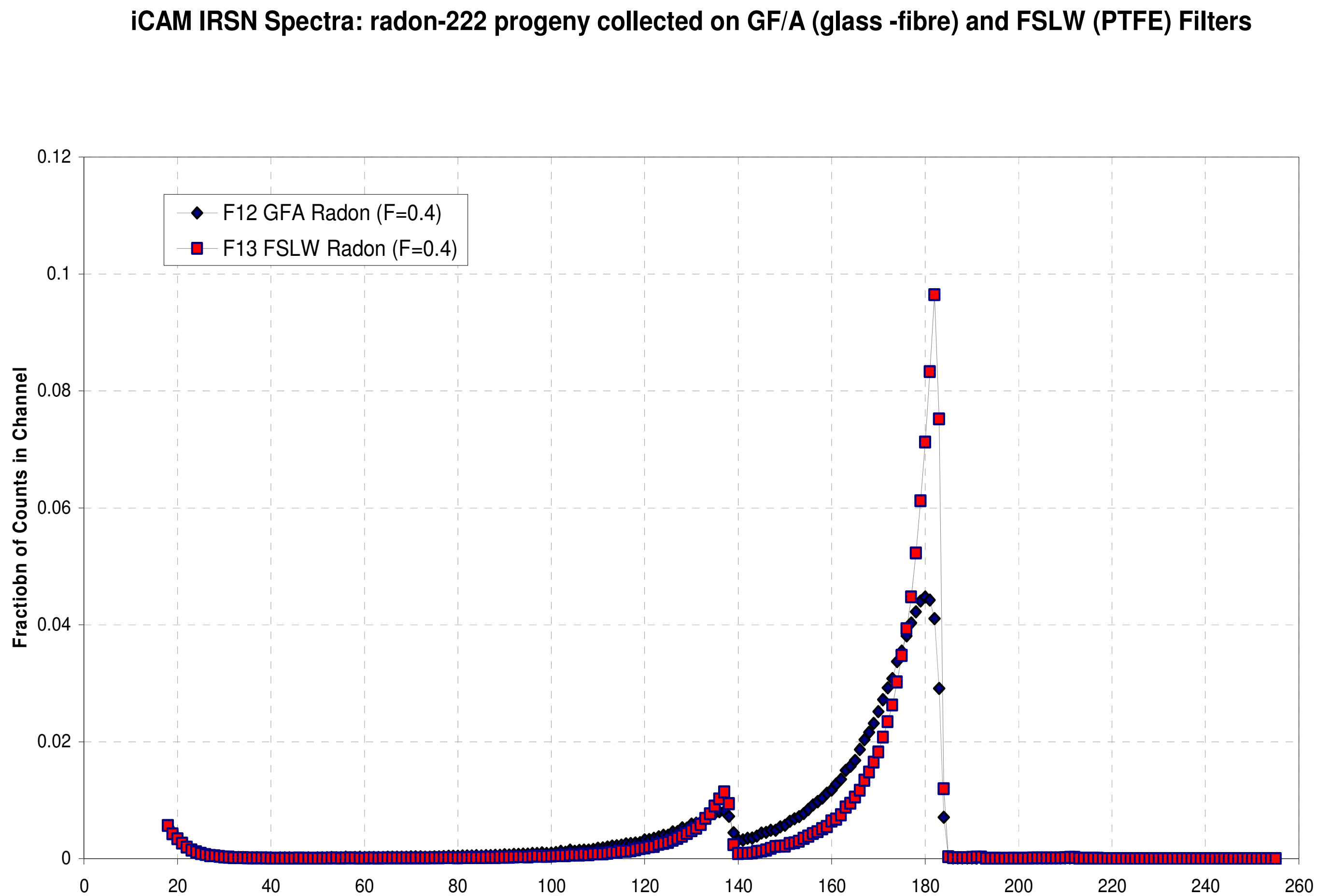
- AMAD =0.2µm
- Equilibrium Factor F=0.4
- Concentration= 40Bq/m3

Filter materials

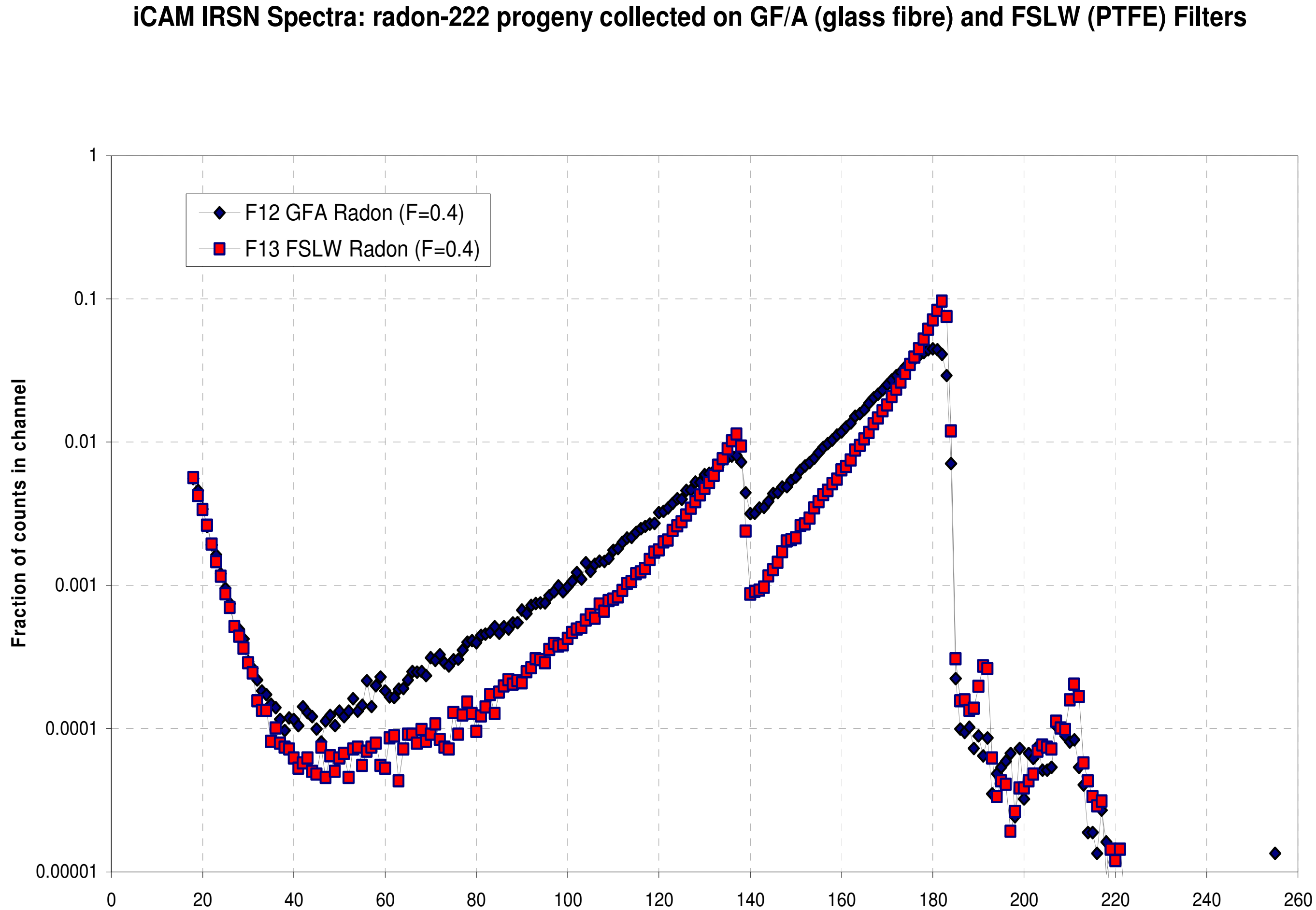
- Whatman GF/A (Glass microfibre)

- Fluoropore FSLW (3µm PTFE filter)

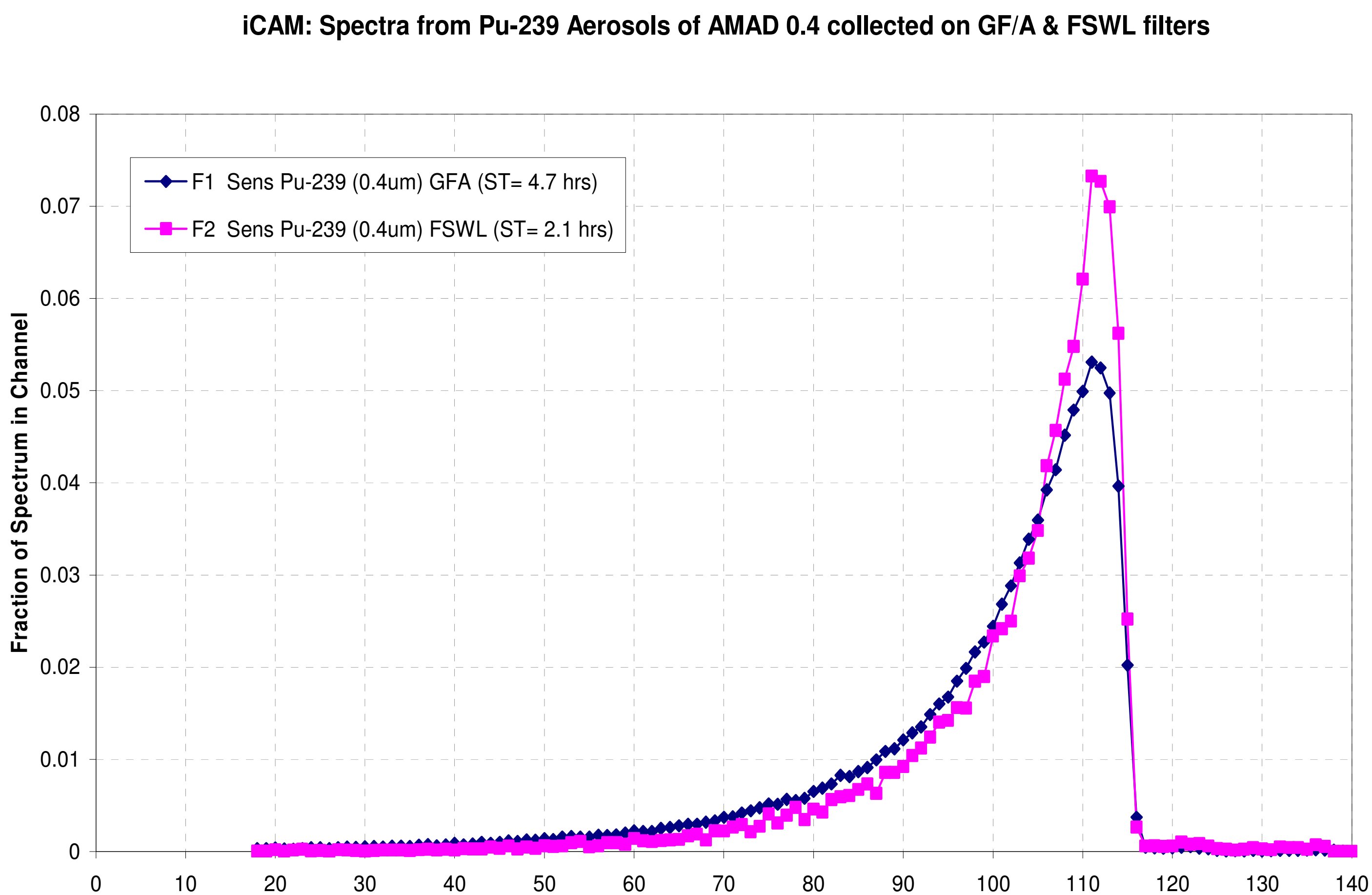
Radon progeny (RaA+RaC′) aerosols (AMAD= 0.4 μm) collected by Whatman GF/A and Fluoropore FSLW filters



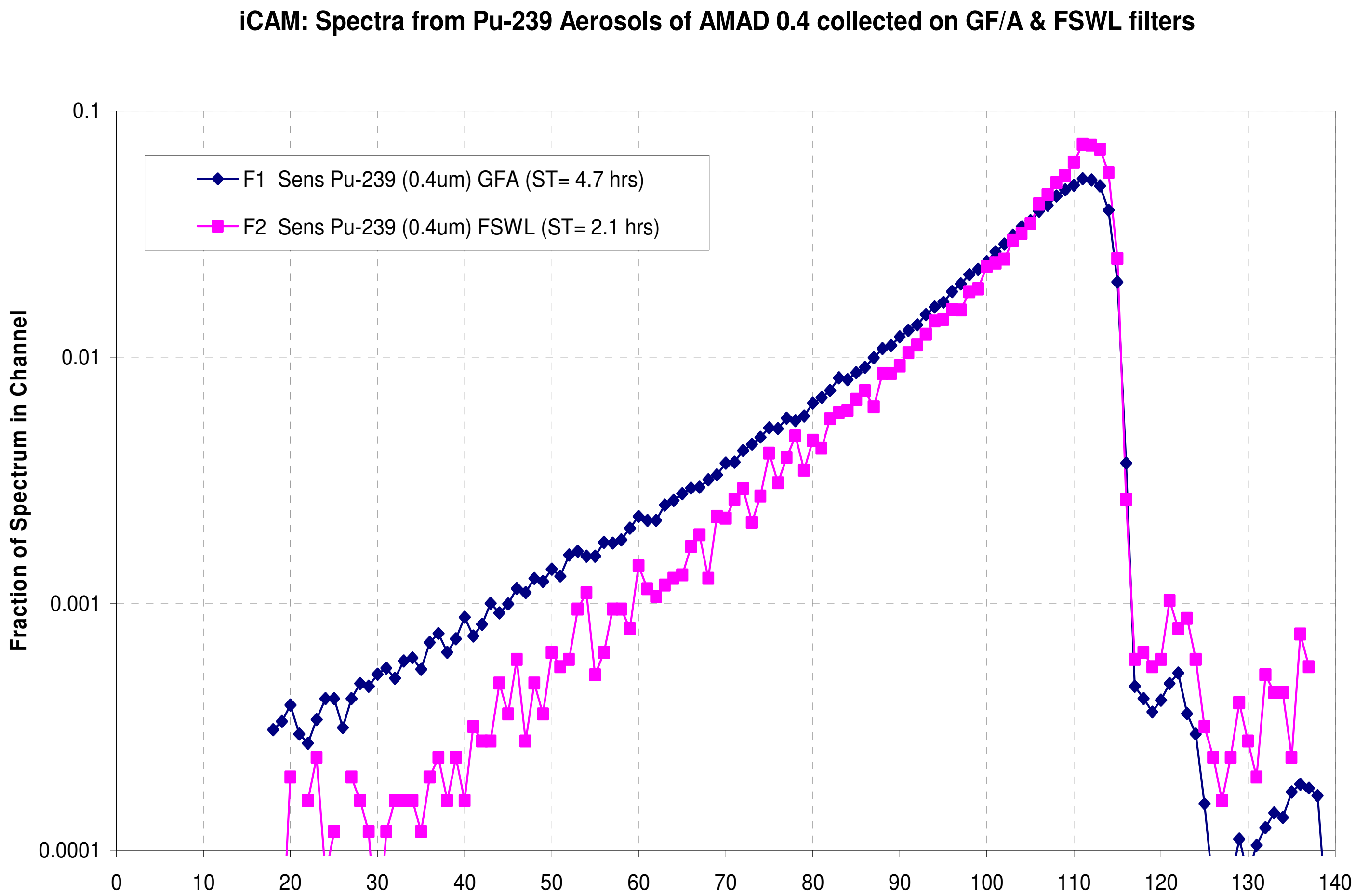
Radon progeny (RaA+RaC′) aerosols (AMAD= 0.4 µm) collected by Whatman GF/A and Fluoropore FSLW filters



Pu-239 aerosols (AMAD= 0.4 µm) collected by Whatman GF/A and Fluoropore FSLW flters



Pu-239 aerosols (AMAD= 0.4 µm) collected by Whatman GF/A and Fluoropore FSLW flters



Three test runs with ^{239}Pu aerosols

- The instrument sampled air containing ^{239}Pu aerosols at calibrated concentration levels for between 5 and 6 hours.
- Air sampling then continued for at least a further 16 hours with activity-free air

Three test runs with ^{239}Pu aerosols

Run 1

Filter:	Whatman GF/A
Aerosol:	^{239}Pu (AMAD=0.4µm)
Mean concentration:	10 Bq/m^3
Duration with ^{239}Pu:	5 hours 20 minutes

Total sampling duration: 22 hours

Run 2

Filter:	Whatman GF/A
Aerosol:	^{239}Pu (AMAD=4µm)
Mean concentration:	13 Bq/m^3
Duration with ^{239}Pu:	6 hours

Total sampling duration: 22 hours

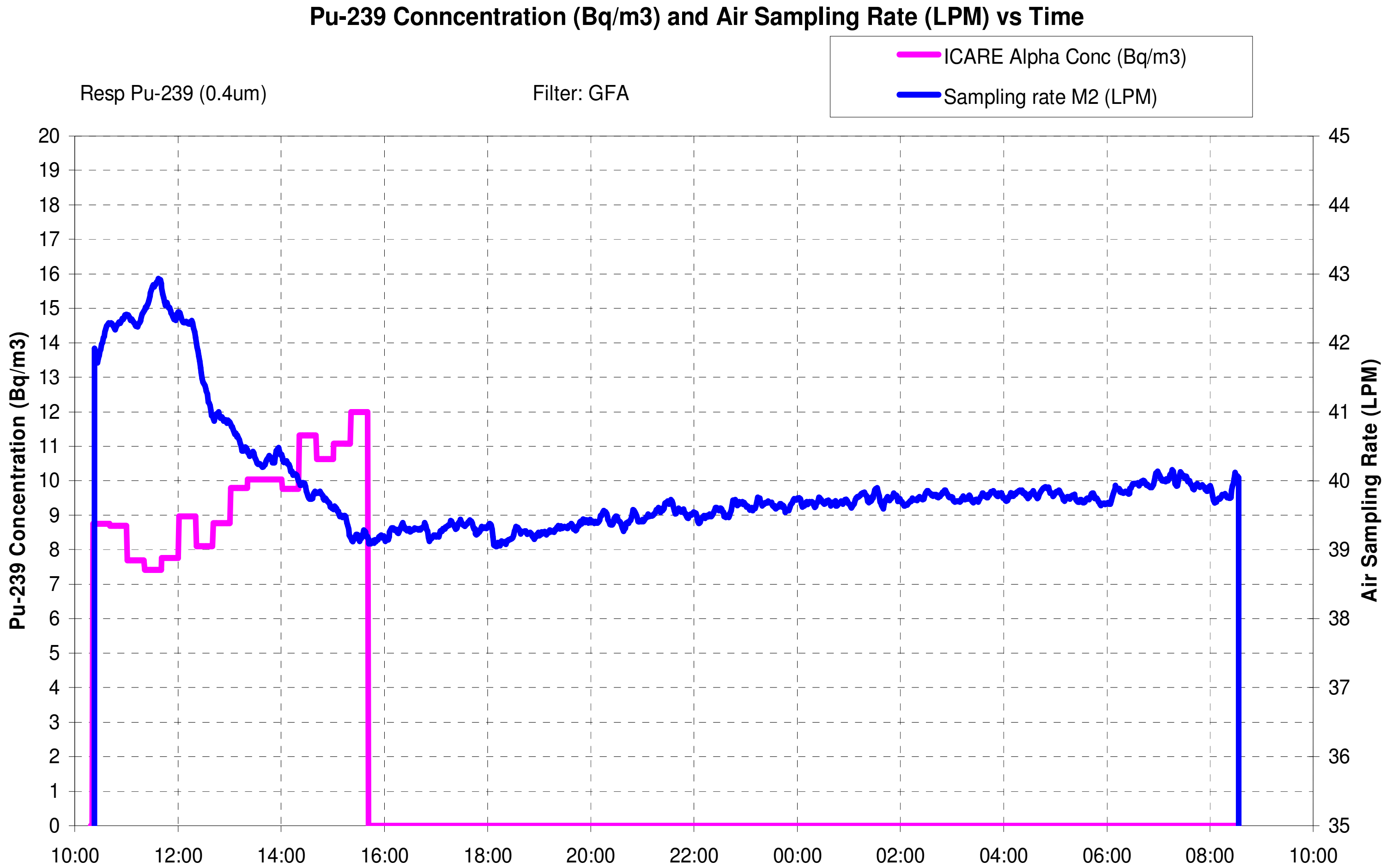
Run 3

Filter:	Fluoropore FSLW
Aerosol:	^{239}Pu (AMAD=0.4µm)
Mean concentration:	9 Bq/m^3
Duration with ^{239}Pu:	5 hours 20 minutes

Total sampling duration: 22 hours

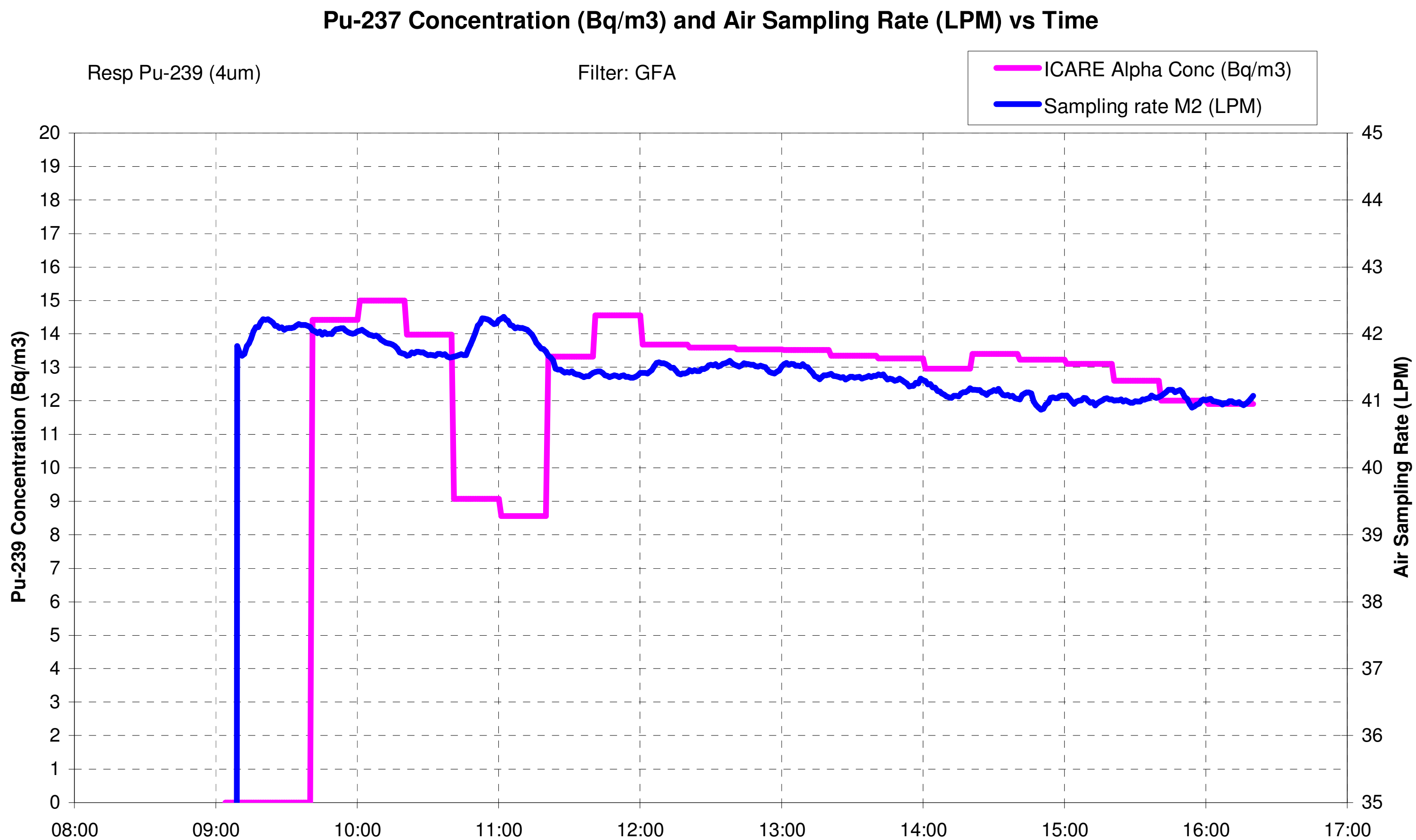
^{239}Pu Concentration and Air Sampling Rate vs Time

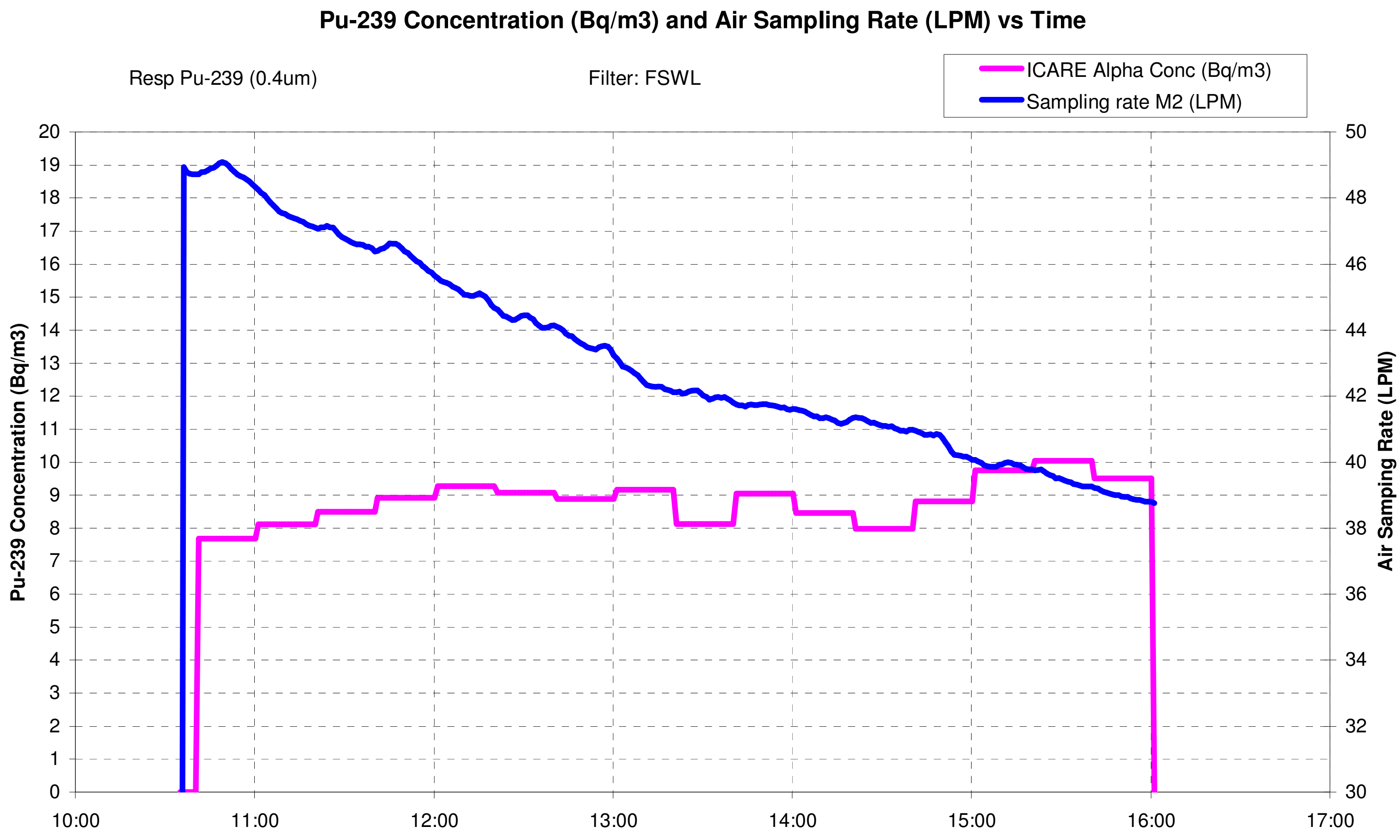
Filter = GF/A   Aerosol AMAD = 0.4µm



^{239}Pu Concentration and Air Sampling Rate vs Time

Filter = GF/A Aerosol AMAD = 4 µm





Pu-239 Concentration (Bq/m3) and Air Sampling Rate (LPM) vs Time
Resp Pu-239 (0.4um)
Filter: FSWL
ICARE Alpha Conc (Bq/m3)
Sampling rate M2 (LPM)
Pu-239 Concentration (Bq/m3)
Air Samping Rate (LPM)
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
30
32
34
36
38
40
42
44
46
48
50
10:00
11:00
12:00
13:00
14:00
15:00
16:00
17:00

Fall in Air Sampling Rate as ^{239}Pu aerosol is collected

- GF/A filter: The impedance to flow increased moderately with time when sampling 0.4 µm AMAD aerosols.
- Fluoropore filter: The impedance to flow increased significantly with time when sampling 0.4 µm AMAD aerosols.
- GF/A filter: The change in impedance to flow was insignificant when sampling 4 µm AMAD aerosols.

Fall in Air Sampling Rate as ^{239}Pu aerosol is collected by filter

Filter	Pu-239 Aerosol AMAD	Initial Air Flow Rate	Final Air Flow Rate	Fall in Air Flow Rate	Fall in Air Flow Rate
GF/A	0.4 µm	43 LPM	39 LPM	4 LPM	9 %
GF/A	4 µm	42 LPM	41 LPM	1 LPM	2 %
FSLW	0.4 µm	49 LPM	39 LPM	10 LPM	20 %

Alpha Spectra. Run 1

Filter: Whatman GF/A

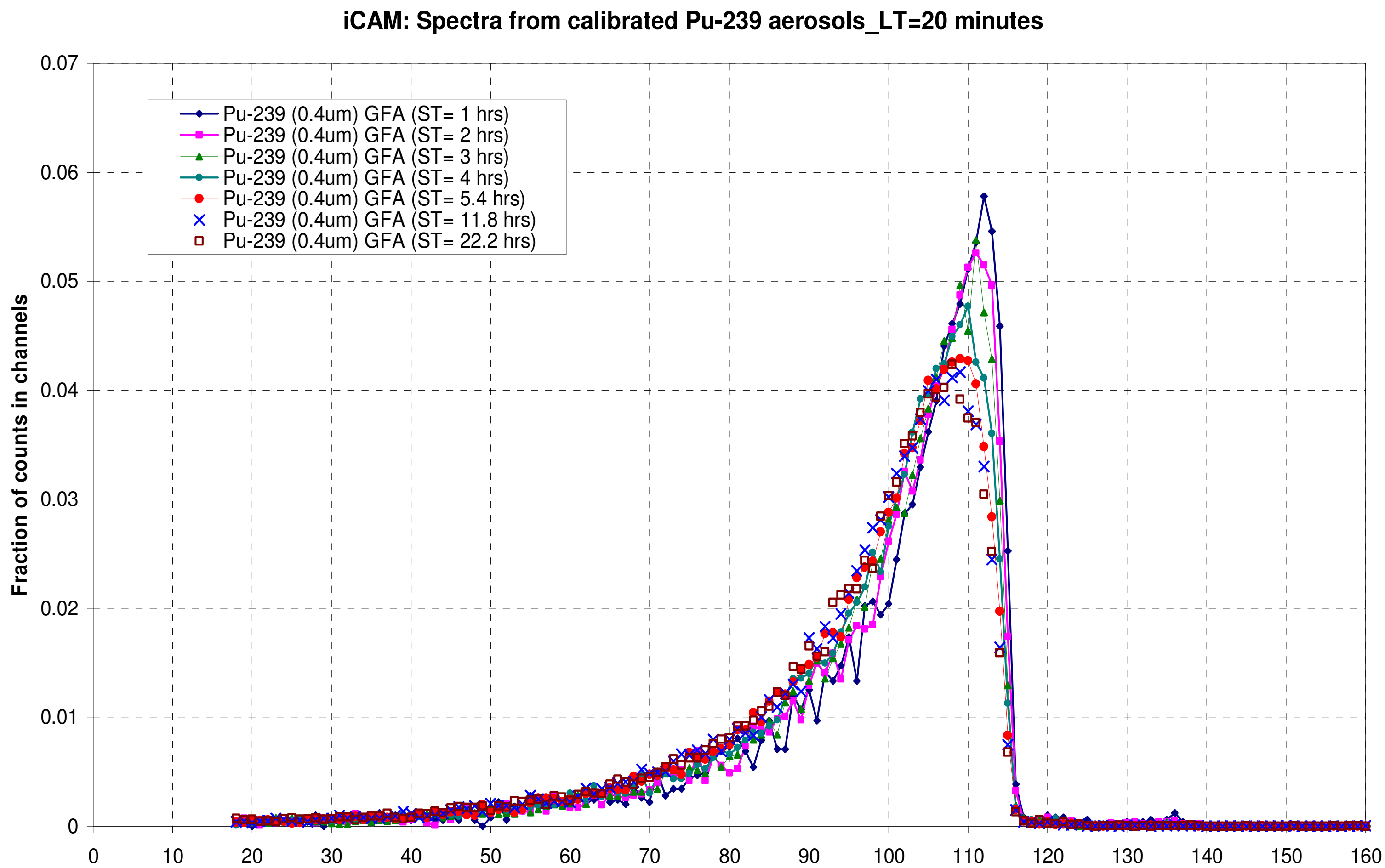
Aerosol: ^{239}Pu (AMAD=0.4µm)

Mean concentration: 10 Bq/m^3

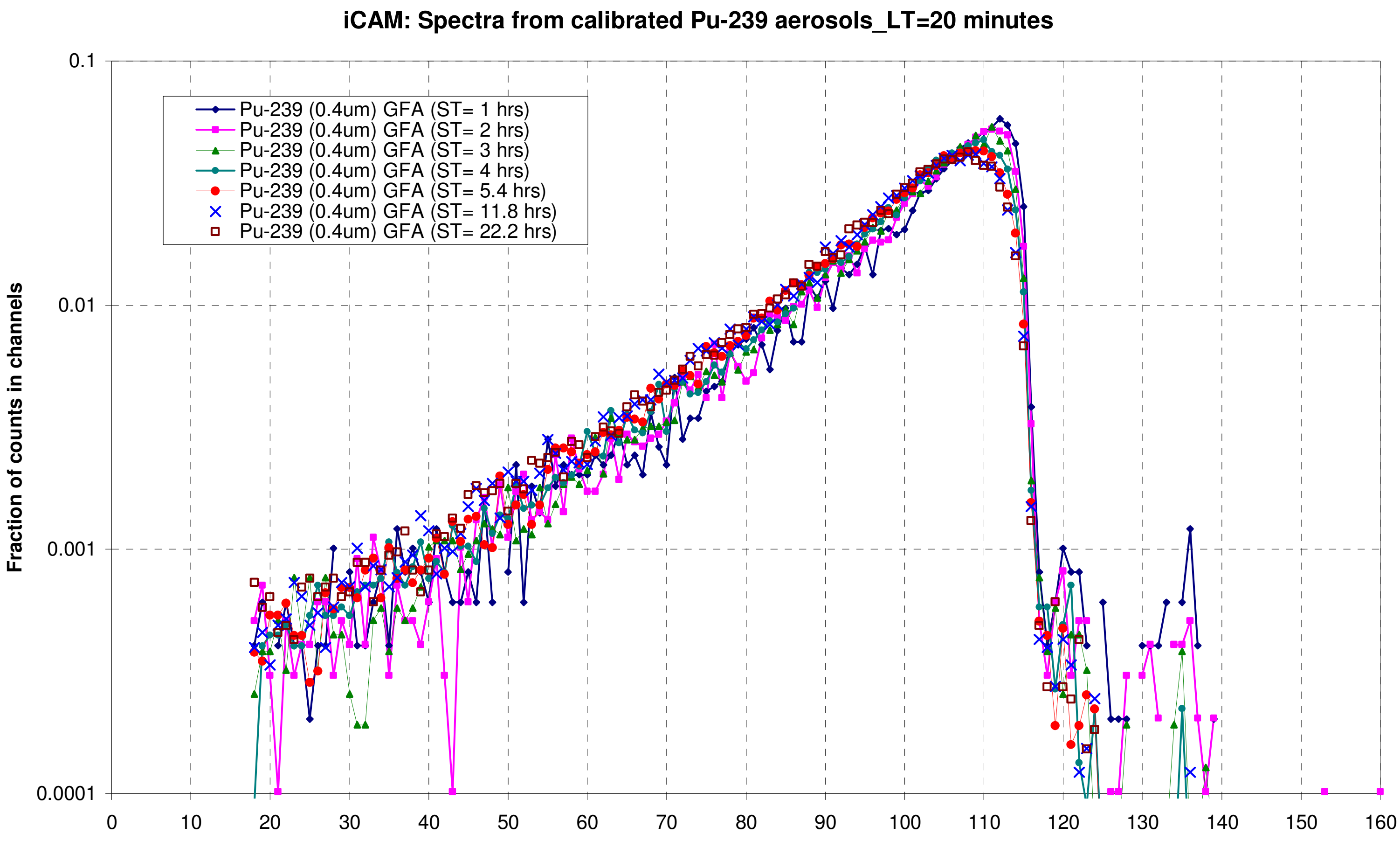
Duration with ^{239}Pu: 5 hour 20 minutes

Total sampling duration: 22 hours

GF/A Filter collecting Pu-239 aerosol (AMAD 0.4)
20 minute counts at hourly intervals
Linear Scale



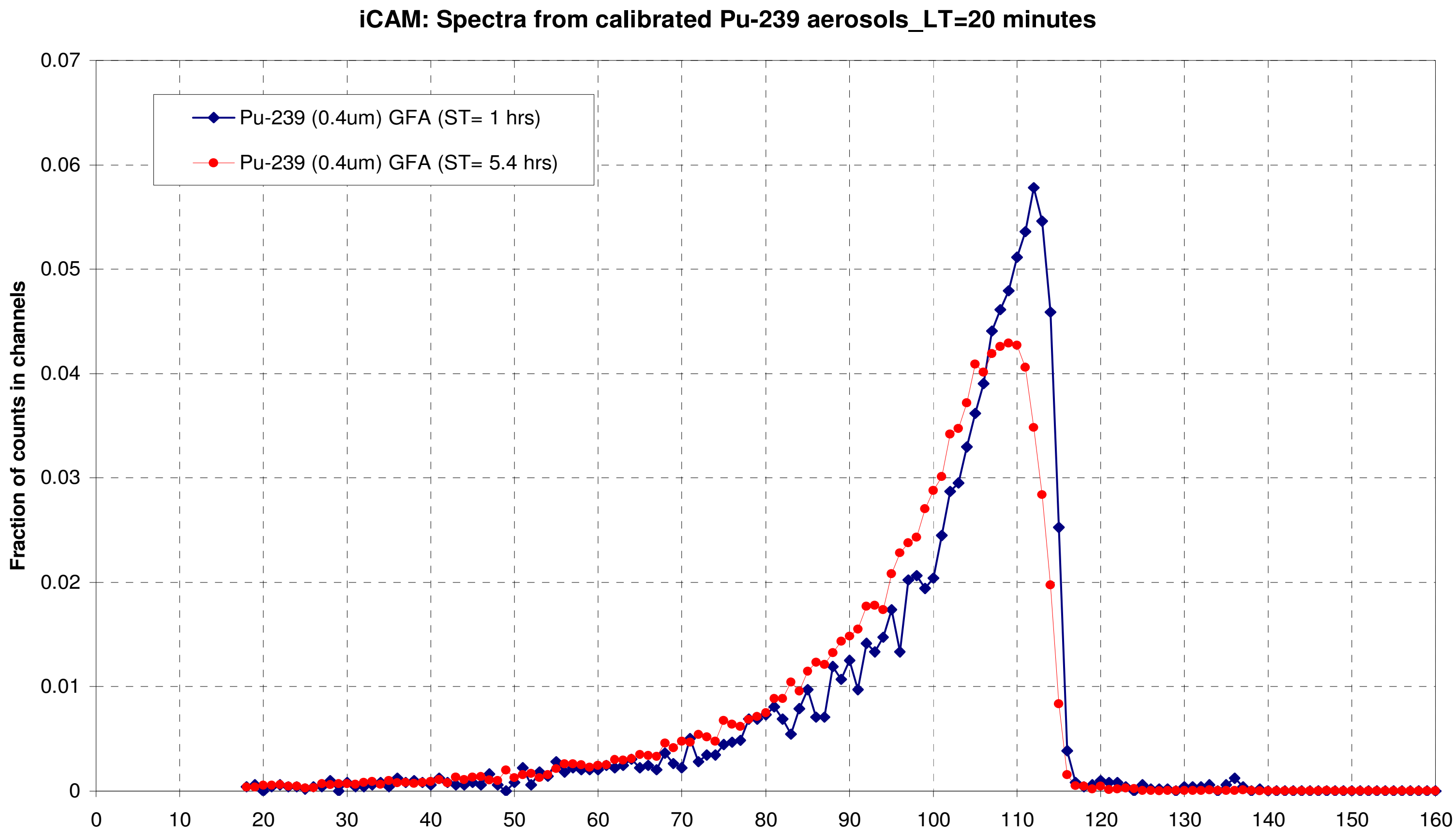
GF/A Filter collecting Pu-239 aerosol (AMAD 0.4)
20 minute counts at hourly intervals
Log Scale



GF/A Filter collecting Pu-239 aerosol (AMAD 0.4)

20 minute counts after 1 and 5 hours sampling activity

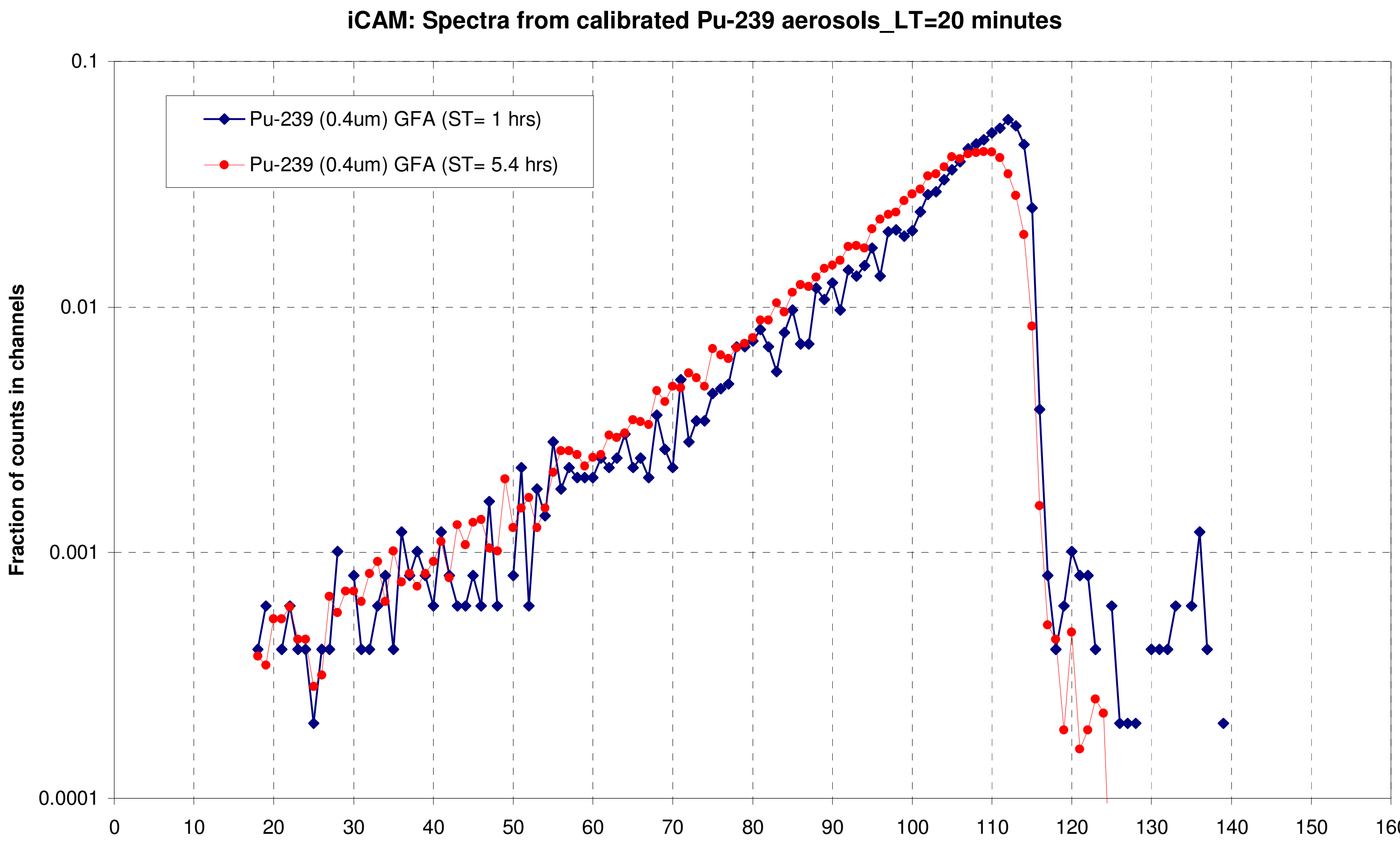
Linear Scale



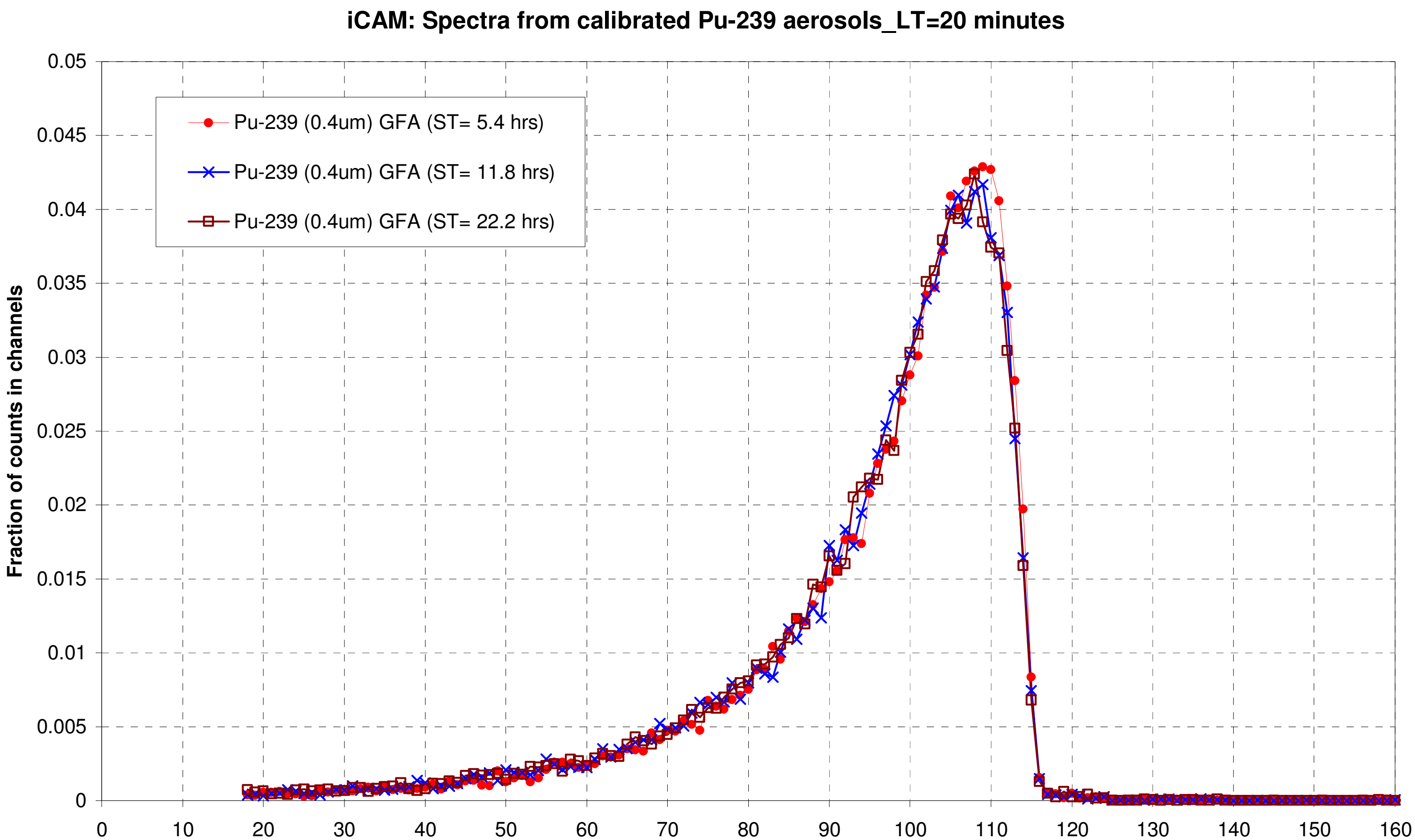
GF/A Filter collecting Pu-239 aerosol (AMAD 0.4)

20 minute counts after 1 and 5 hours sampling activity

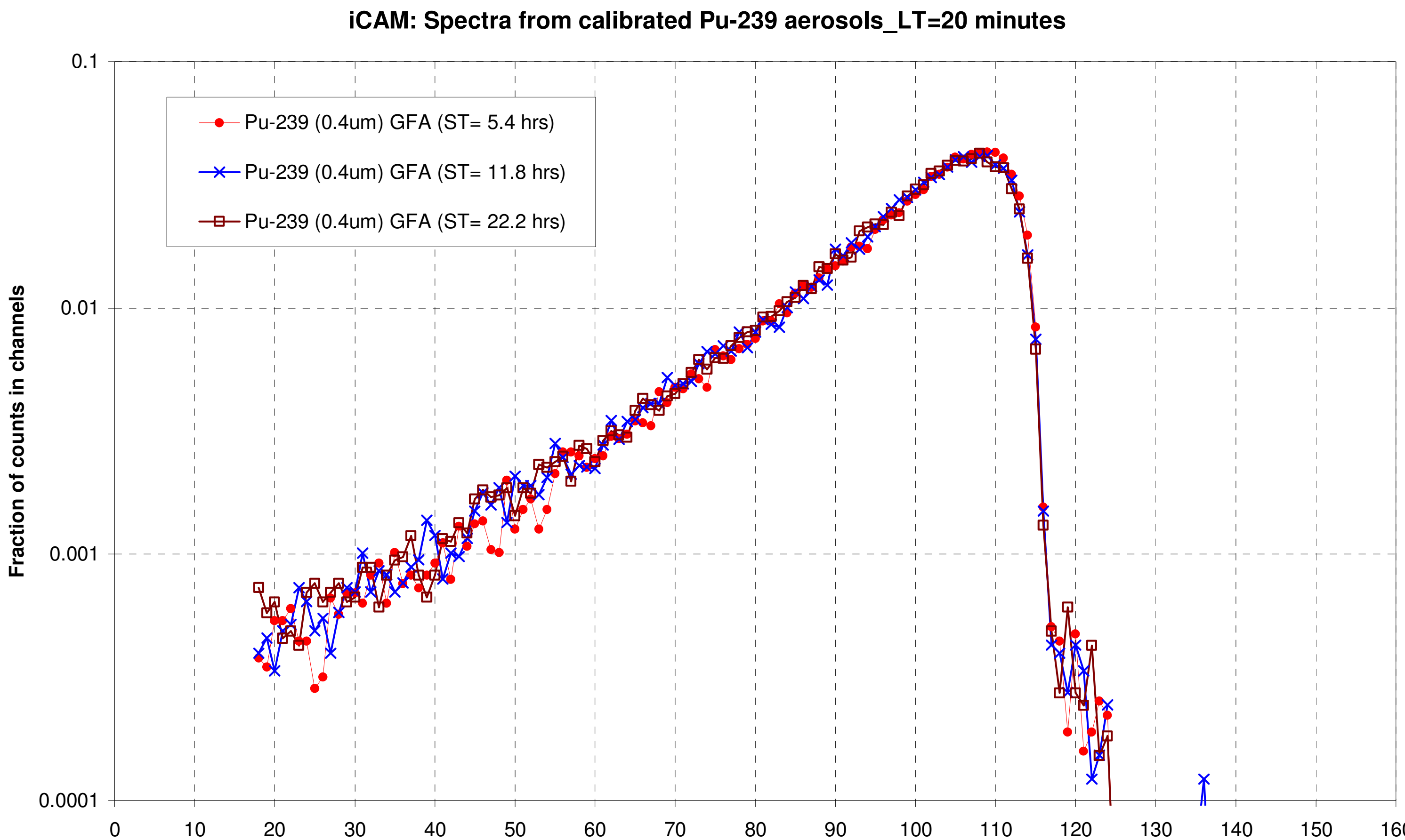
Log Scale



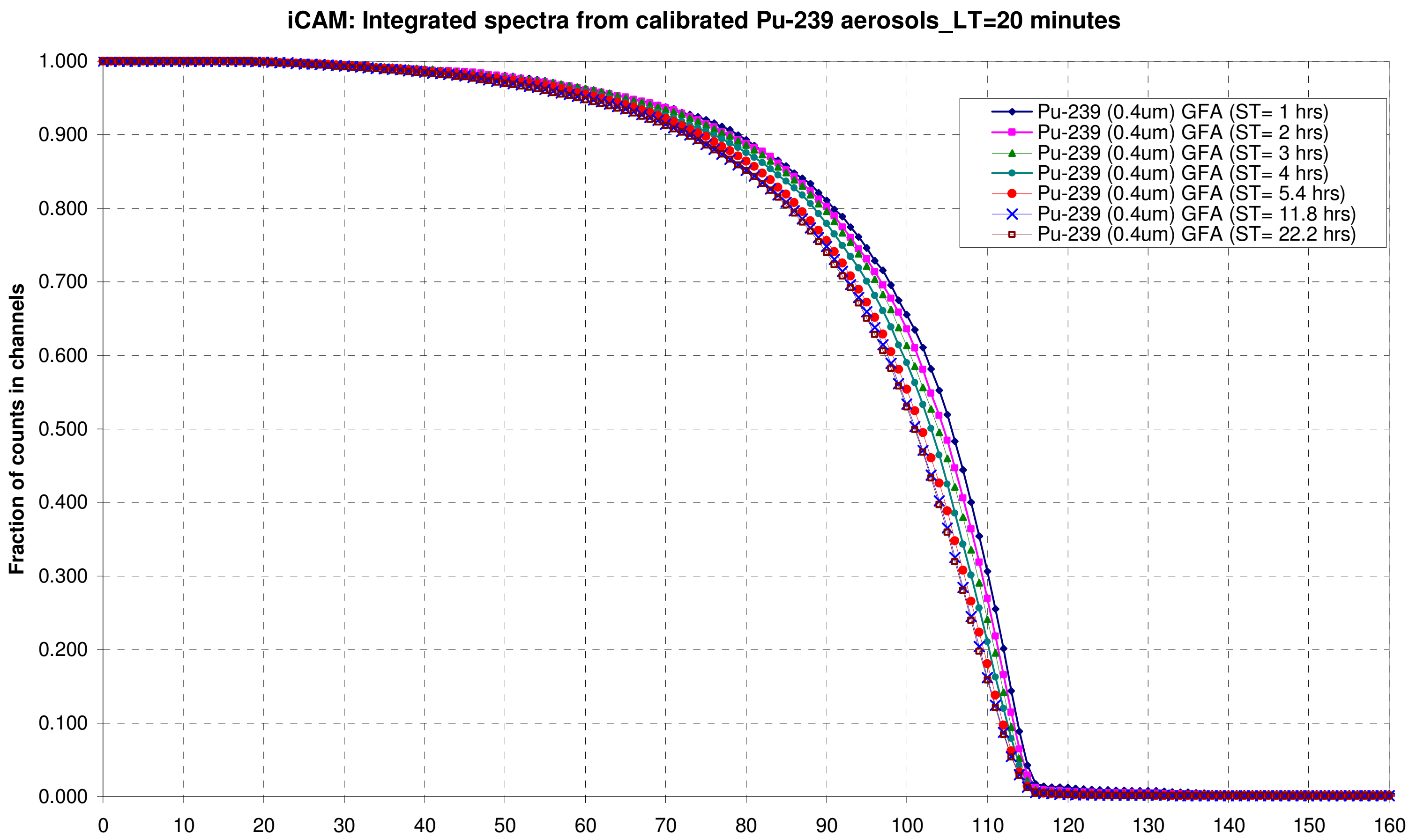
GF/A Filter collecting Pu-239 aerosol (AMAD 0.4)
20 minute counts
At end of activity-sampling period (5h 20m) and at 12 and 22 hours
Linear Scale



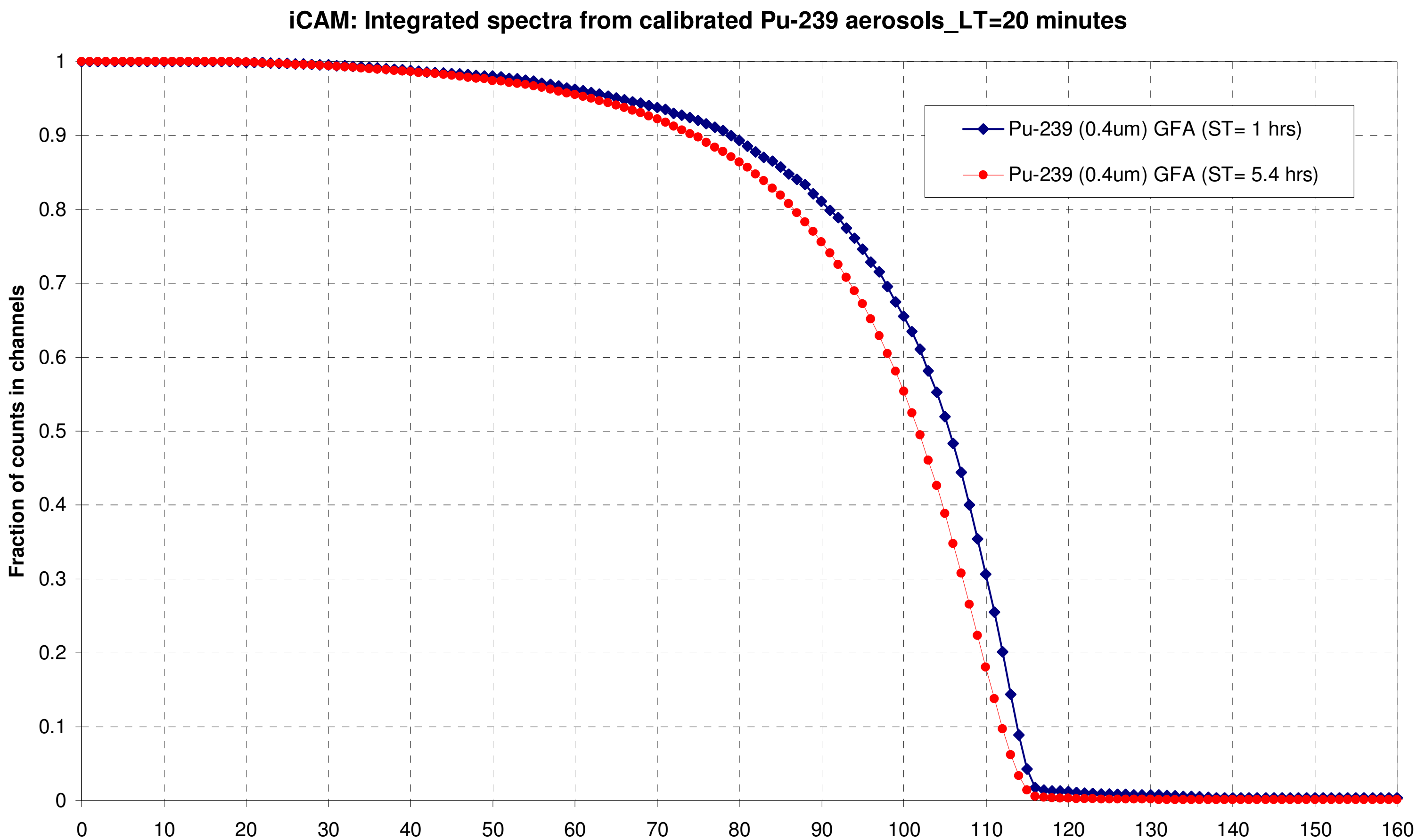
GF/A Filter collecting Pu-239 aerosol (AMAD 0.4)
20 minute counts
At end of activity-sampling period (5h 20m) and at 12 and 22 hours
Log Scale



GF/A Filter collecting Pu-239 aerosol (AMAD 0.4)
20 minute counts at hourly intervals
Integrated Spectrum



GF/A Filter collecting Pu-239 aerosol (AMAD 0.4)
20 minute counts after 1 and 5 hours sampling activity
Integrated Spectrum



Alpha Spectra. Run 2

Filter: Whatman GF/A

Aerosol: ^{239}Pu (AMAD=4μm)

Mean concentration: 13 Bq/m^3

Duration with ^{239}Pu: 5 hour 20 minutes

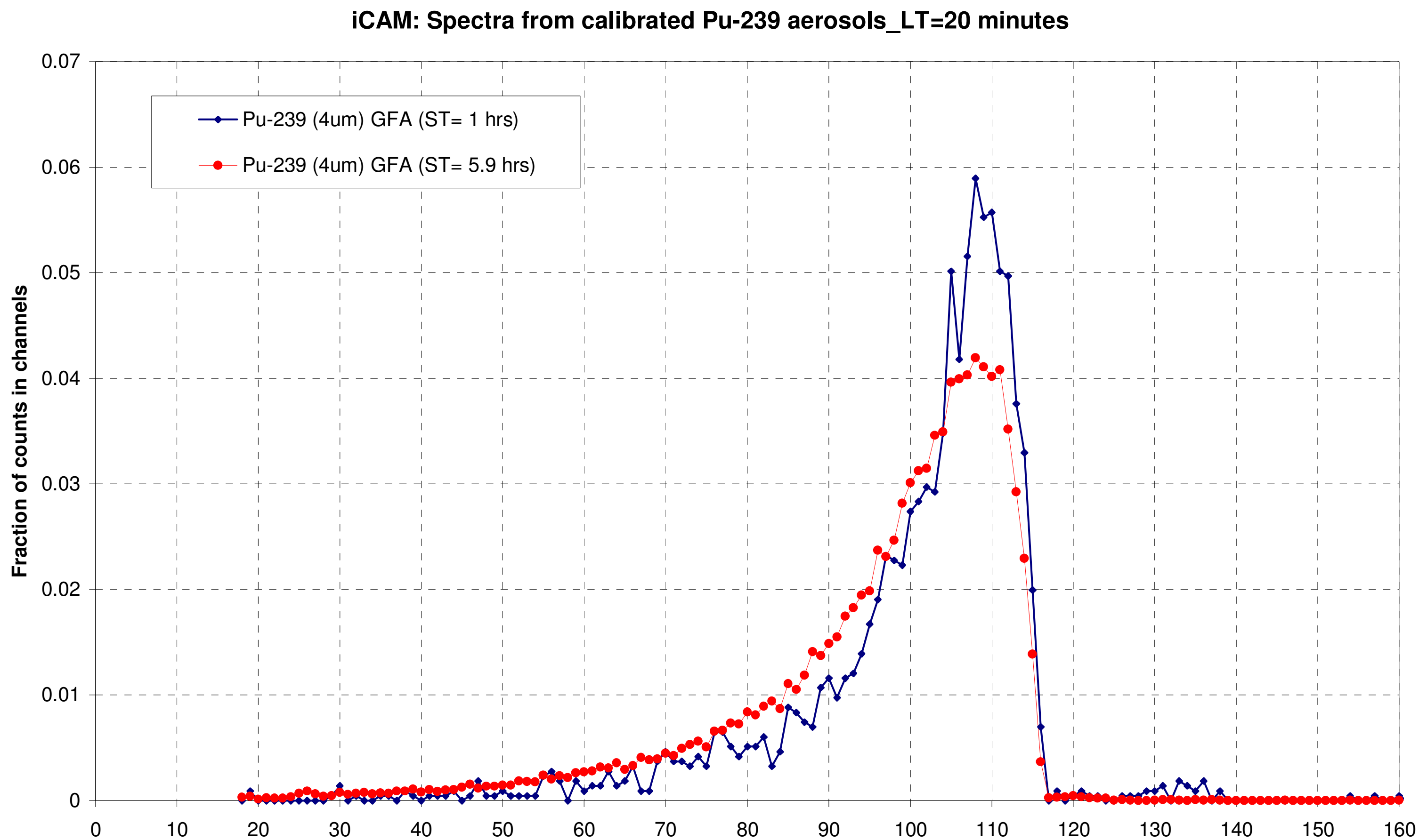
Total sampling duration: 22 hours

GF/A Filter

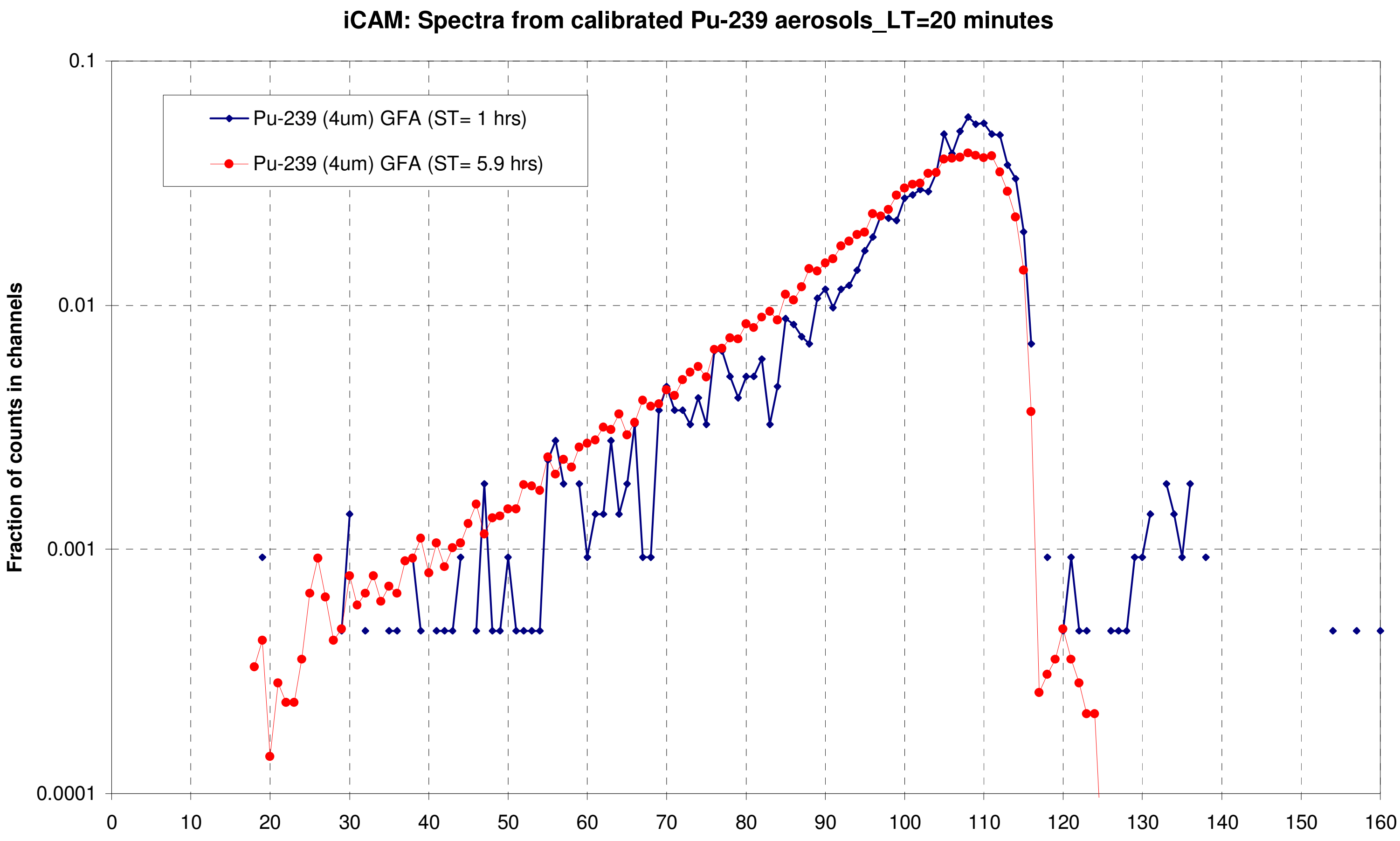
Spectra from 4µm Pu-239 aerosols

20 minute counts after 1 hour and 6 hours sampling activity

LINEAR SCALE



GF/A Filter
Spectra from 4µm Pu-239 aerosols
20 minute counts after 1 hour and 6 hours sampling activity
LOG SCALE



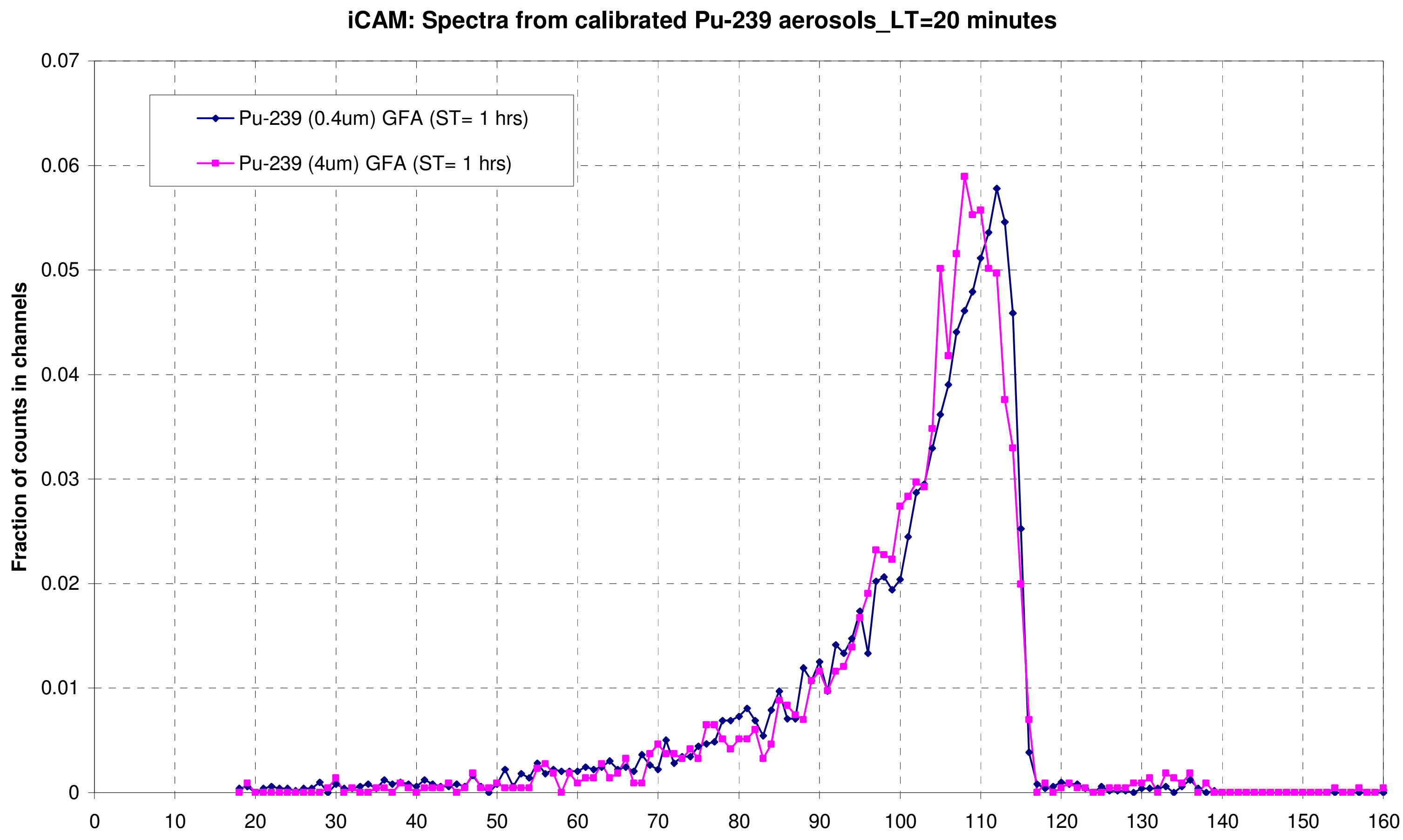
Comparison of Alpha Spectra
from
0.4 µm and 4 µm ^{239}Pu aerosols

GF/A Filters

Spectra from 0.4 μm and 4μm Pu-239 aerosols

20 minute counts after 1 hour sampling activity

LINEAR SCALE

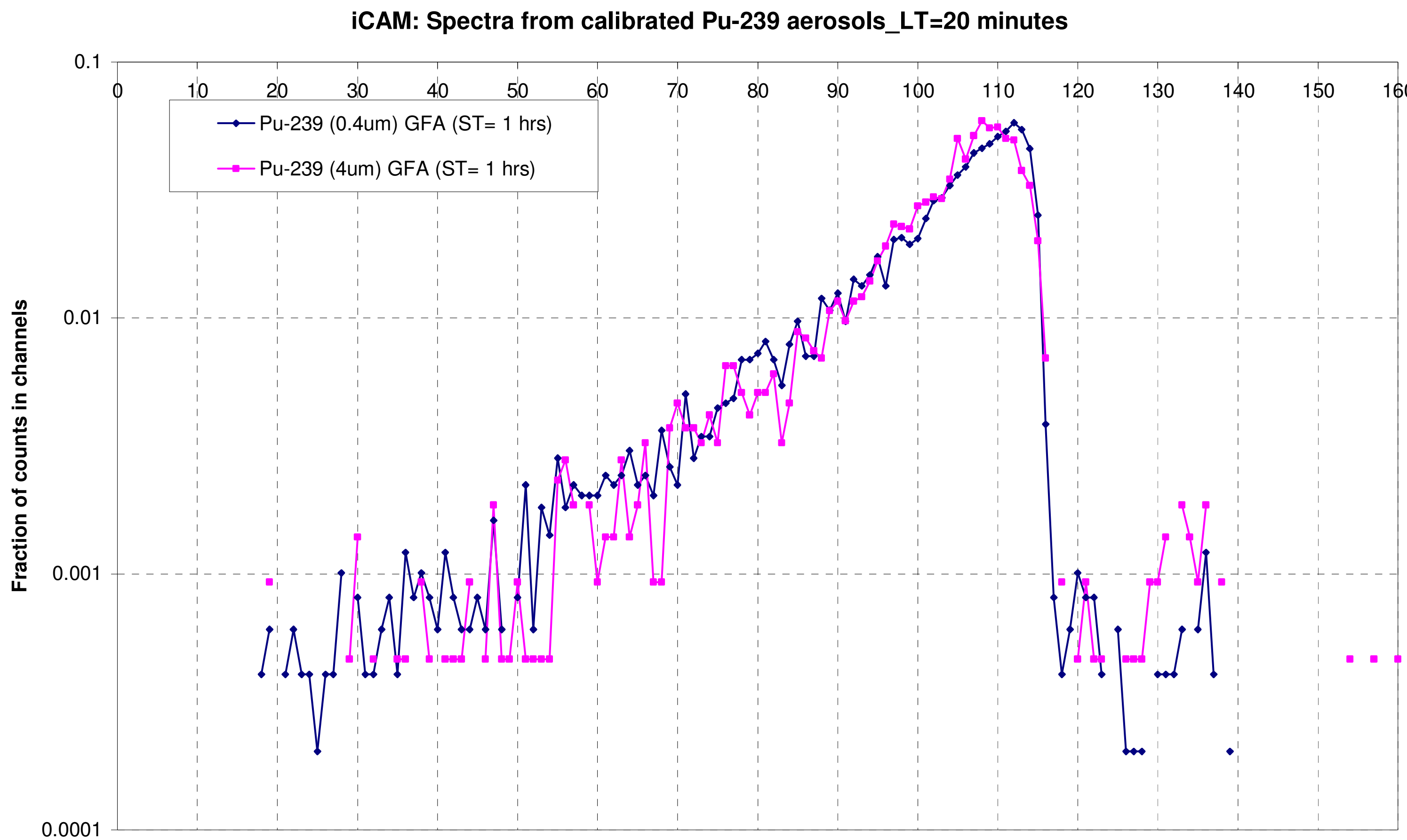


GF/A Filters

Spectra from 0.4 µm and 4µm Pu-239 aerosols

20 minute counts after 1 hour sampling activity

LOG SCALE



Run 3

Filter: Fluoropore FSLW

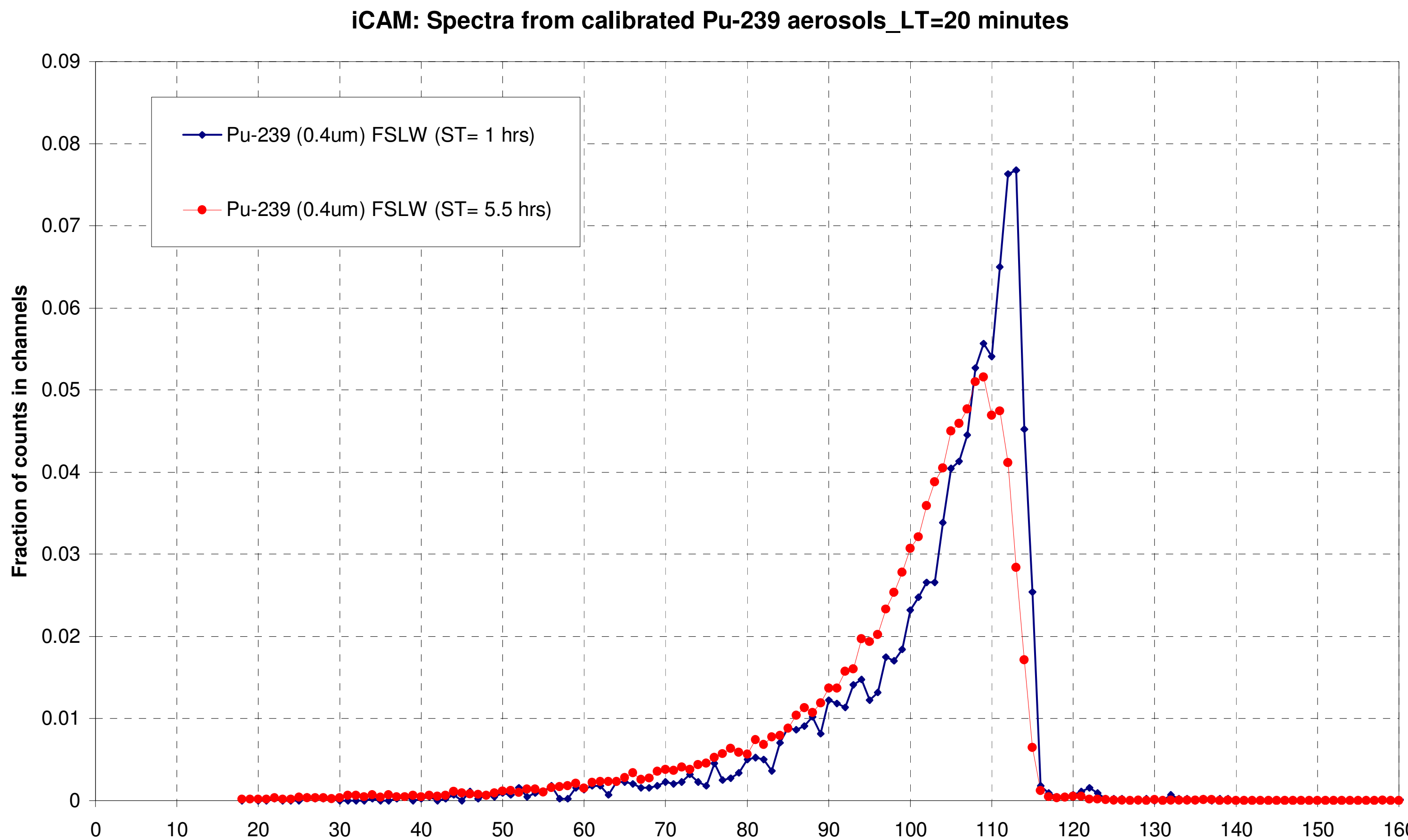
Aerosol: ^{239}Pu (AMAD=0.4µm)

Mean concentration: 9 Bq/m^3

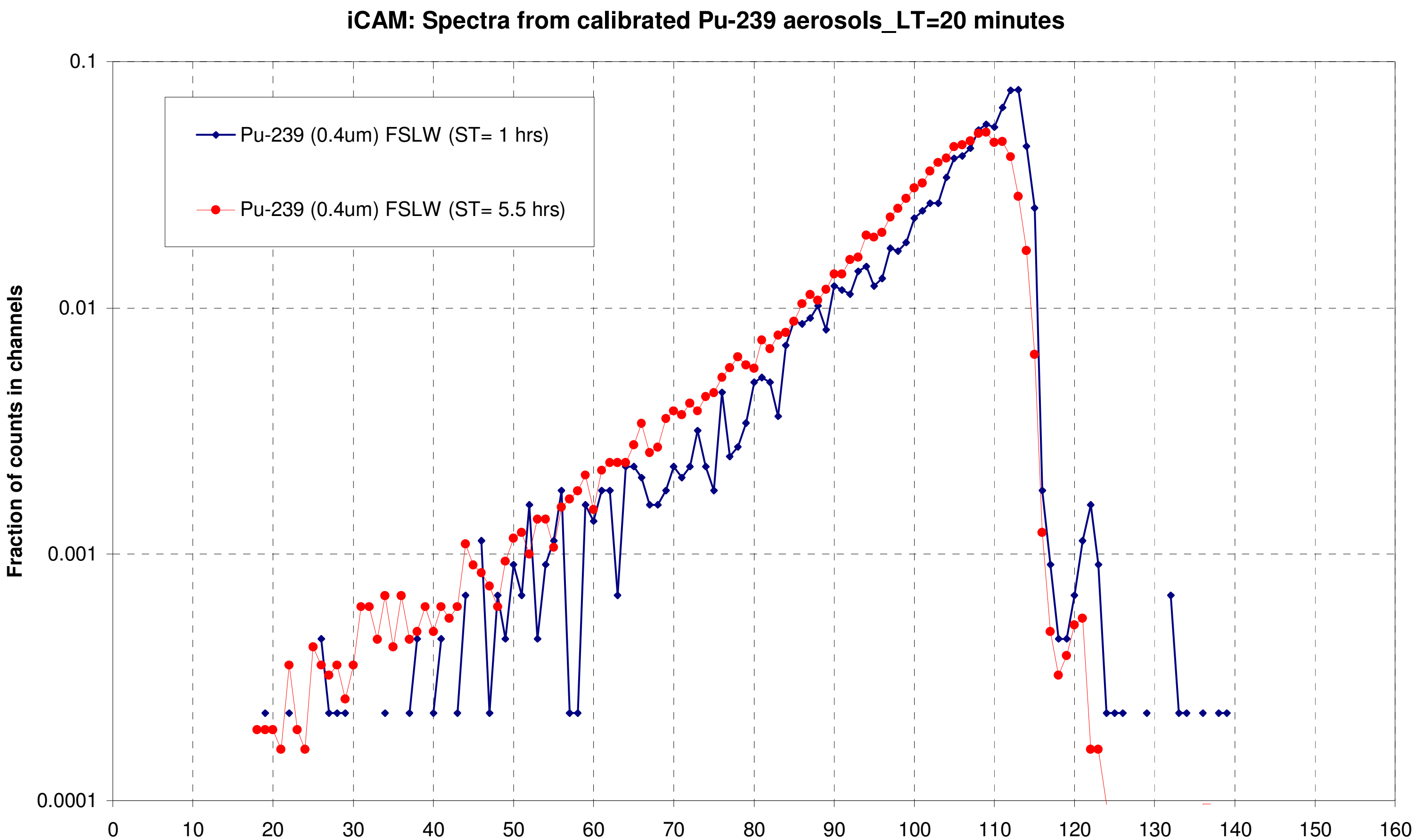
Duration with ^{239}Pu: 5 hour 20 minutes

Total sampling duration: 22 hours

Fluoropore FSLW Filter
Spectra from 0.4µm Pu-239 aerosols
20 minute counts after 1 hour and 5h 20m sampling activity
LINEAR SCALE



Fluoropore FSLW Filter
Spectra from 0.4μm Pu-239 aerosols
20 minute counts after 1 hour and 5h 20m sampling activity
LOG SCALE

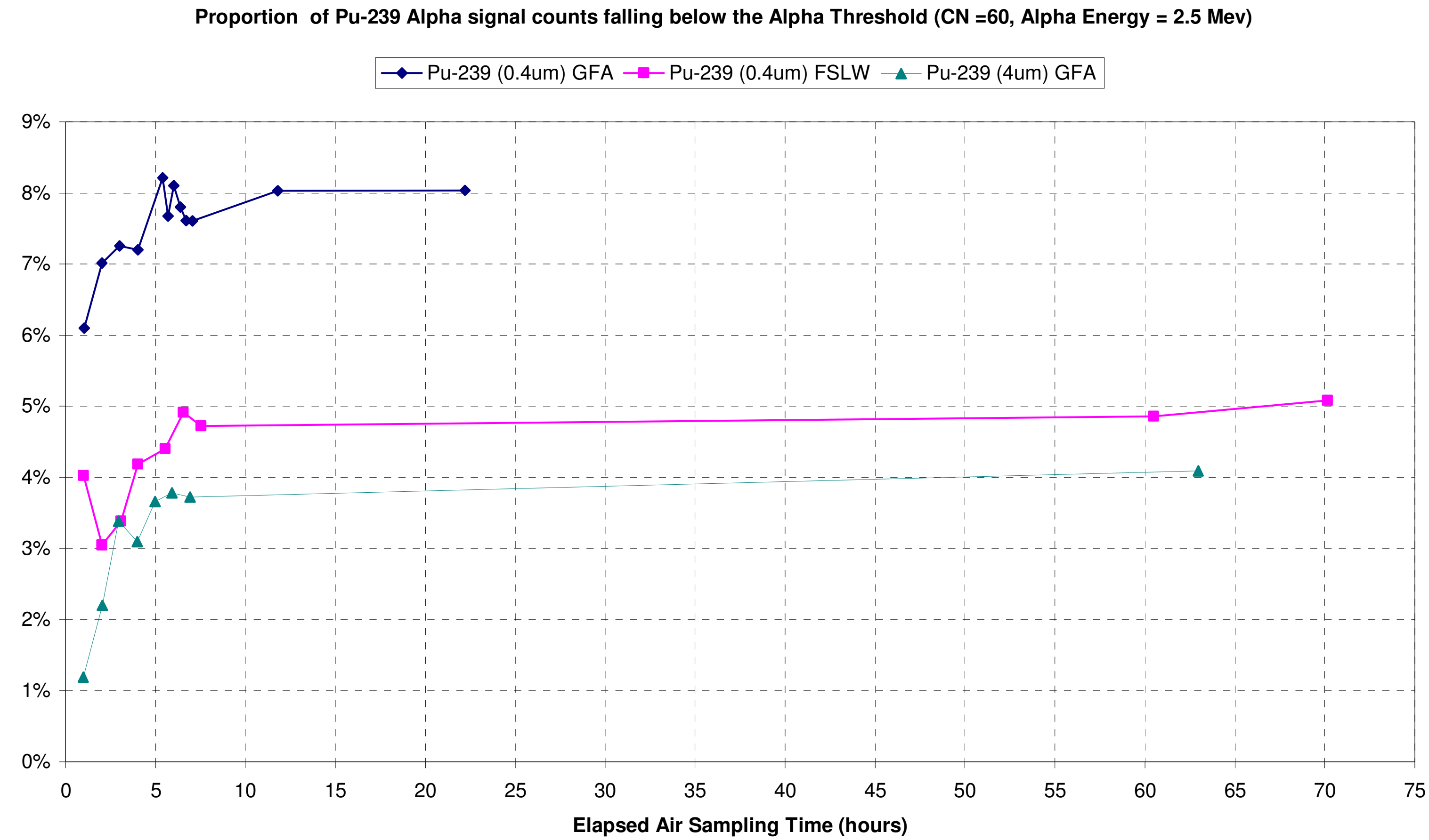


The following slides show the increase in the proportion of the ^{239}Pu alpha counts that fall below the Alpha Region into the Beta Region as aerosol accumulates.

Threshold of Alpha Region :

Channel Number 60 (Alpha Energy = 2.5 MeV)

The increase in the proportion of the ^{239}Pu alpha counts that fall into the Beta Region during aerosol accumulation



In the three test runs the proportion of ^{239}Pu alpha signal counts falling below the Alpha Region threshold was:

- greatest for 0.4µm aerosols collected by a Whatman GF/A filter
- significantly lower for 0.4µm aerosols collected by a Fluoropore FSLW filter
- the least for 4µm aerosols collected by a Whatman GF/A filter
- Presumably the proportion would have been lower with 4µm aerosols collected by a Fluoropore filter
- The collection of 4µm aerosols by GF/A showed no significant increase in impedance during accumulation

The results obtained with calibrated ^{239}Pu aerosols have shown small but significant losses in alpha signal due to alpha energy absorption by the filter material and by aerosols as they accumulate

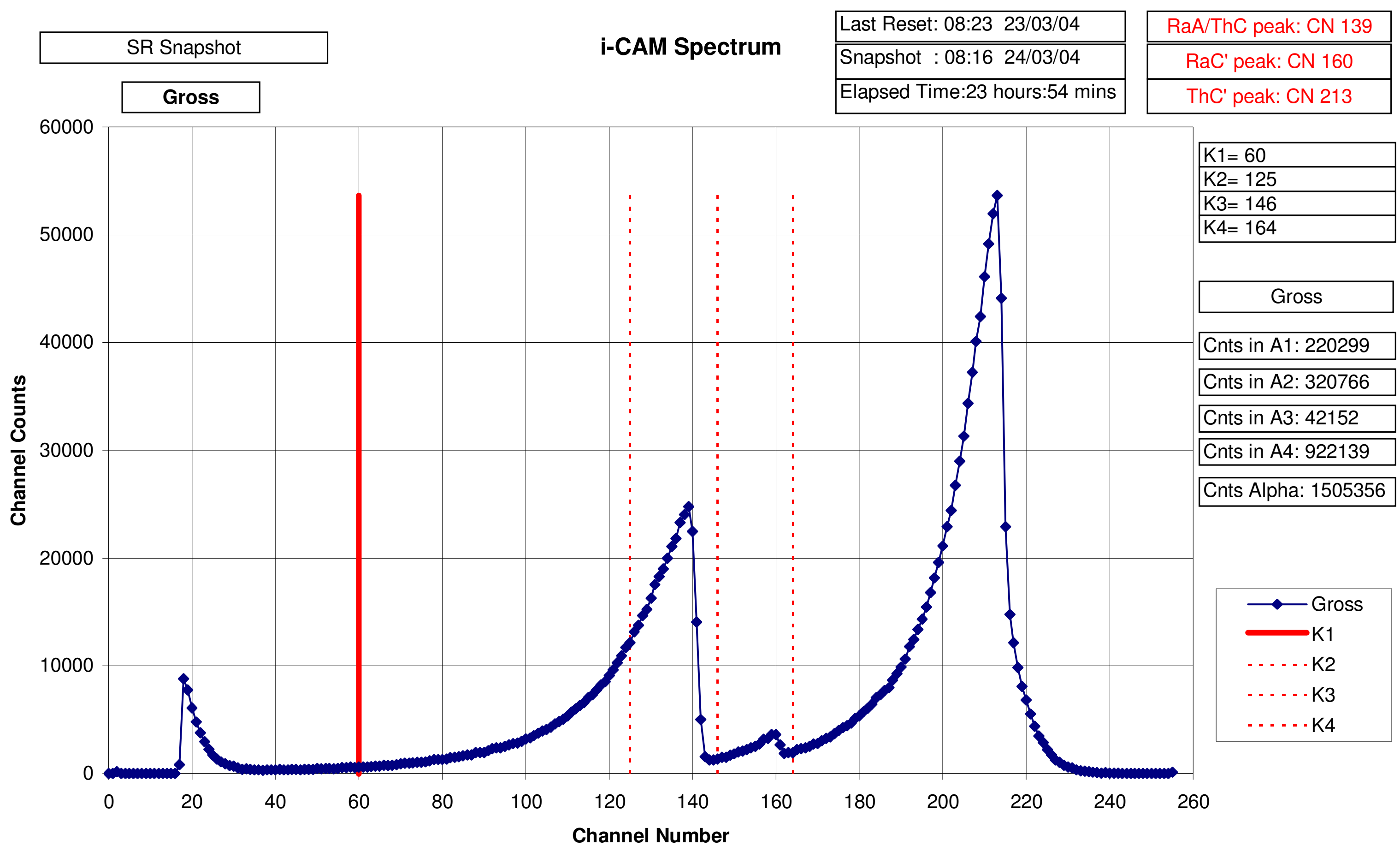
Alpha Spectra heavily degraded by sampling active water droplets

Note: In this example the alpha background is mainly due to thoron progeny ThC and ThC′.

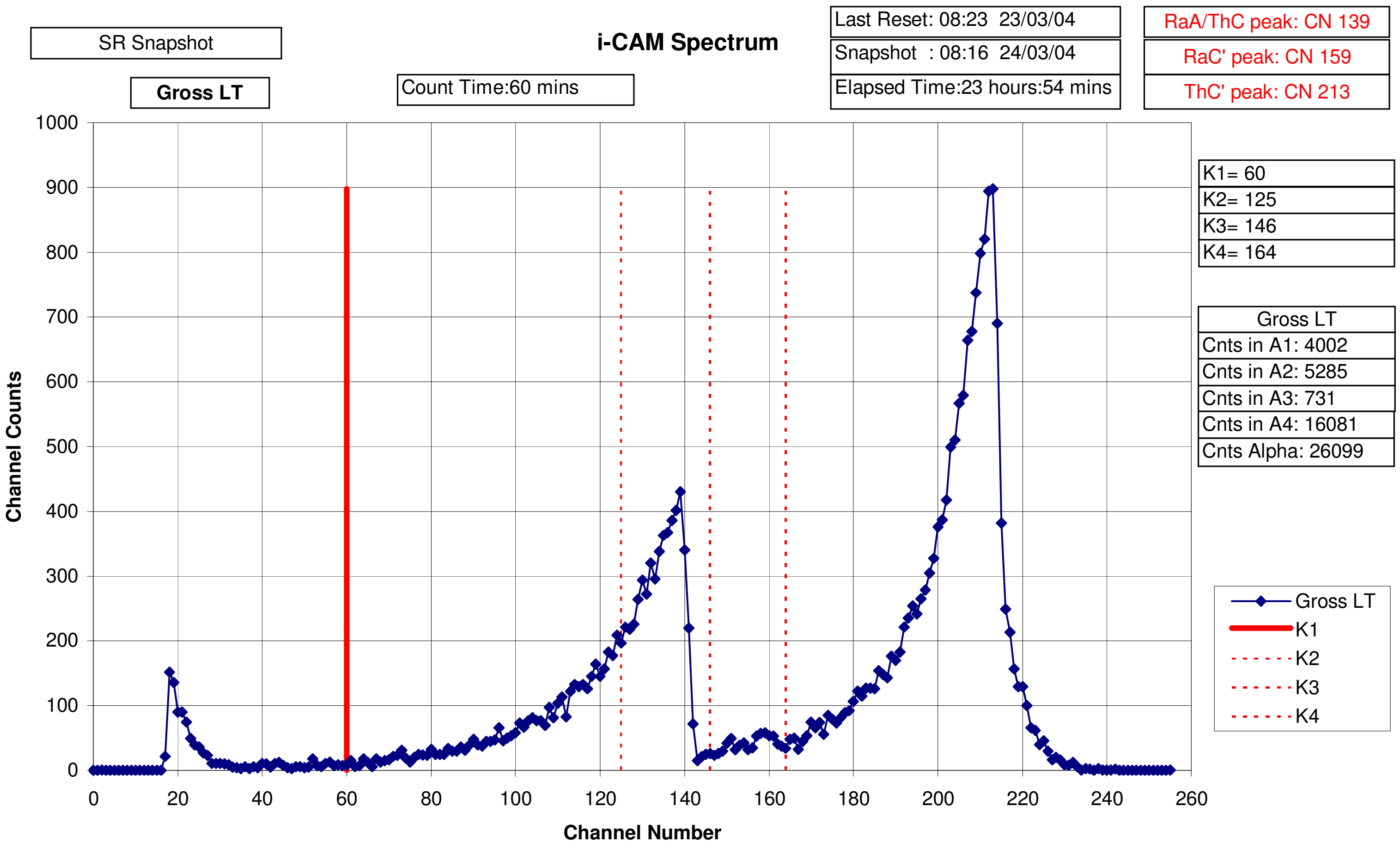
The RaC′ contribution is very small

Alpha Spectrum acquired over 24 hours since last filter change

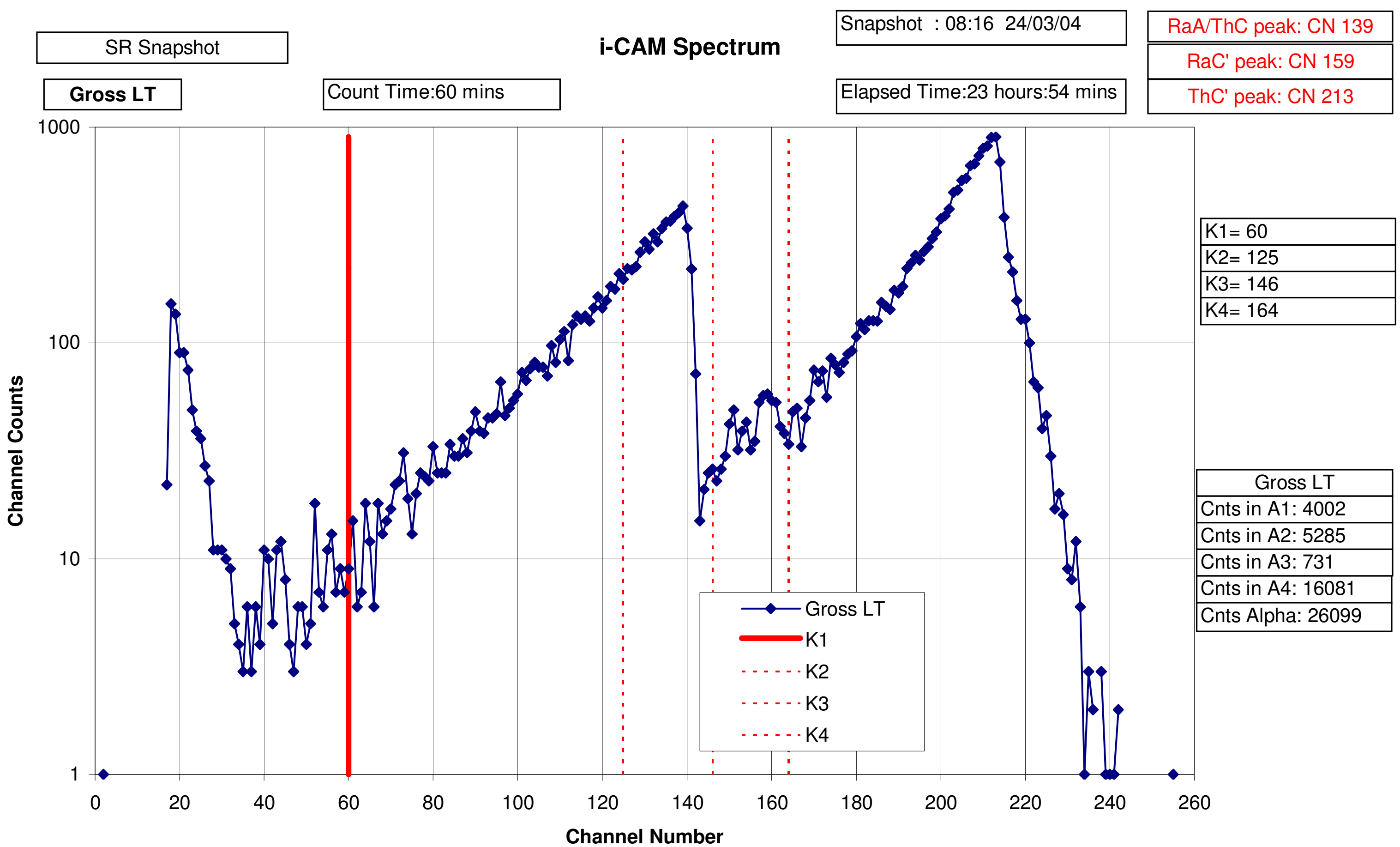
Before arrival of active water drop LINEAR SCALE



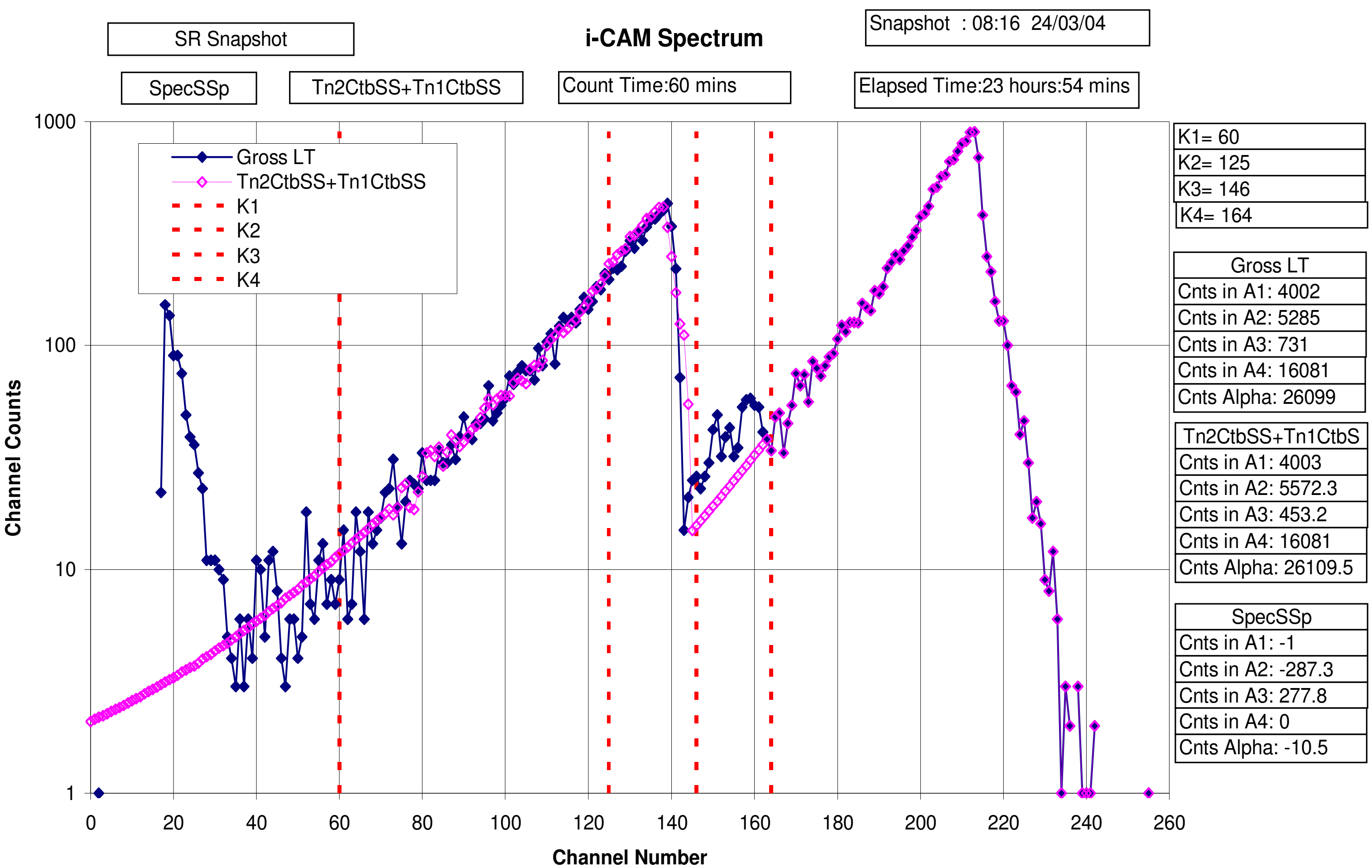
Alpha Spectrum acquired in 60minutes
24 hours since last filter change
Before arrival of active water drop LINEAR SCALE



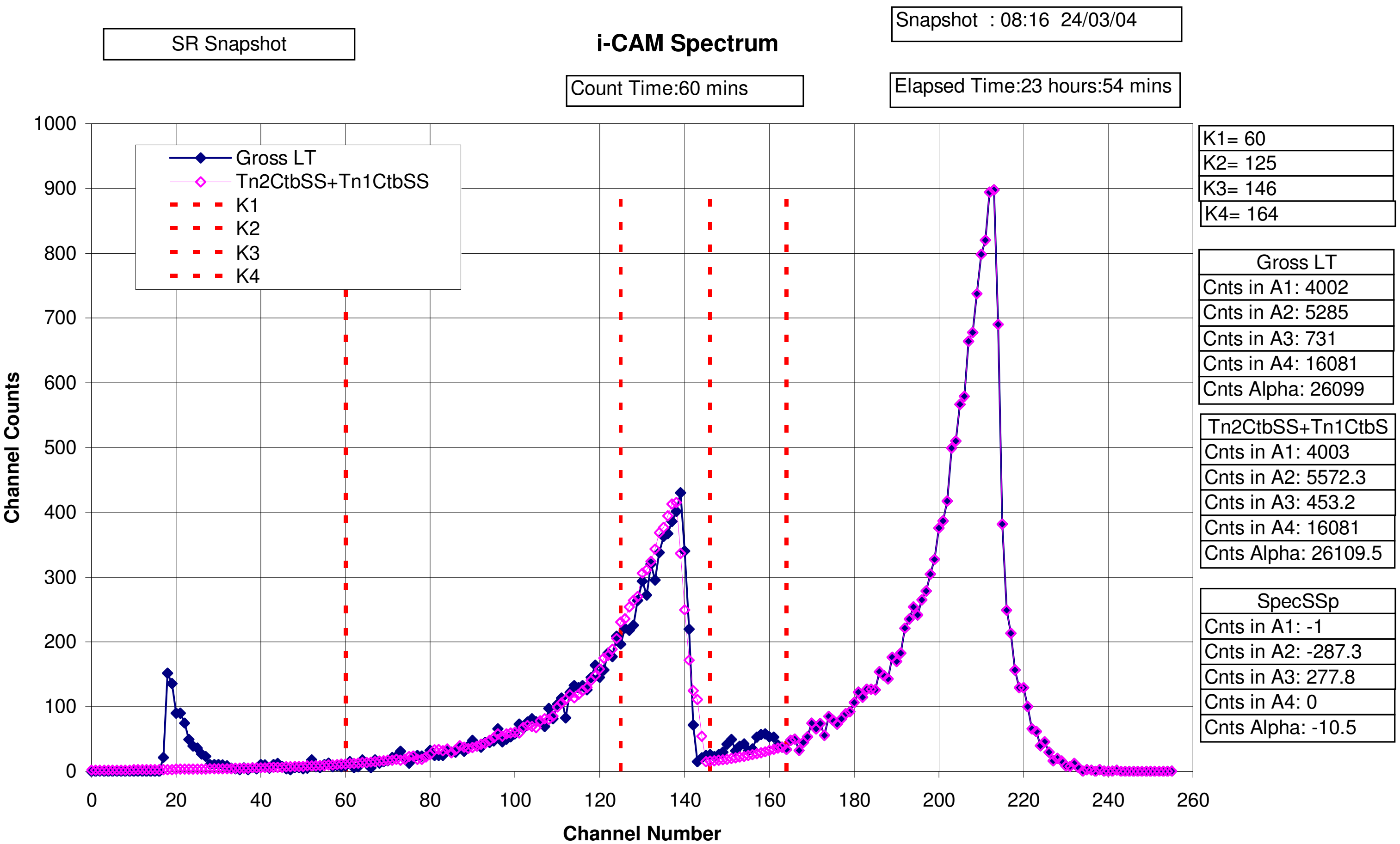
Alpha Spectrum acquired in 60minutes Before arrival of active water drop LOG SCALE



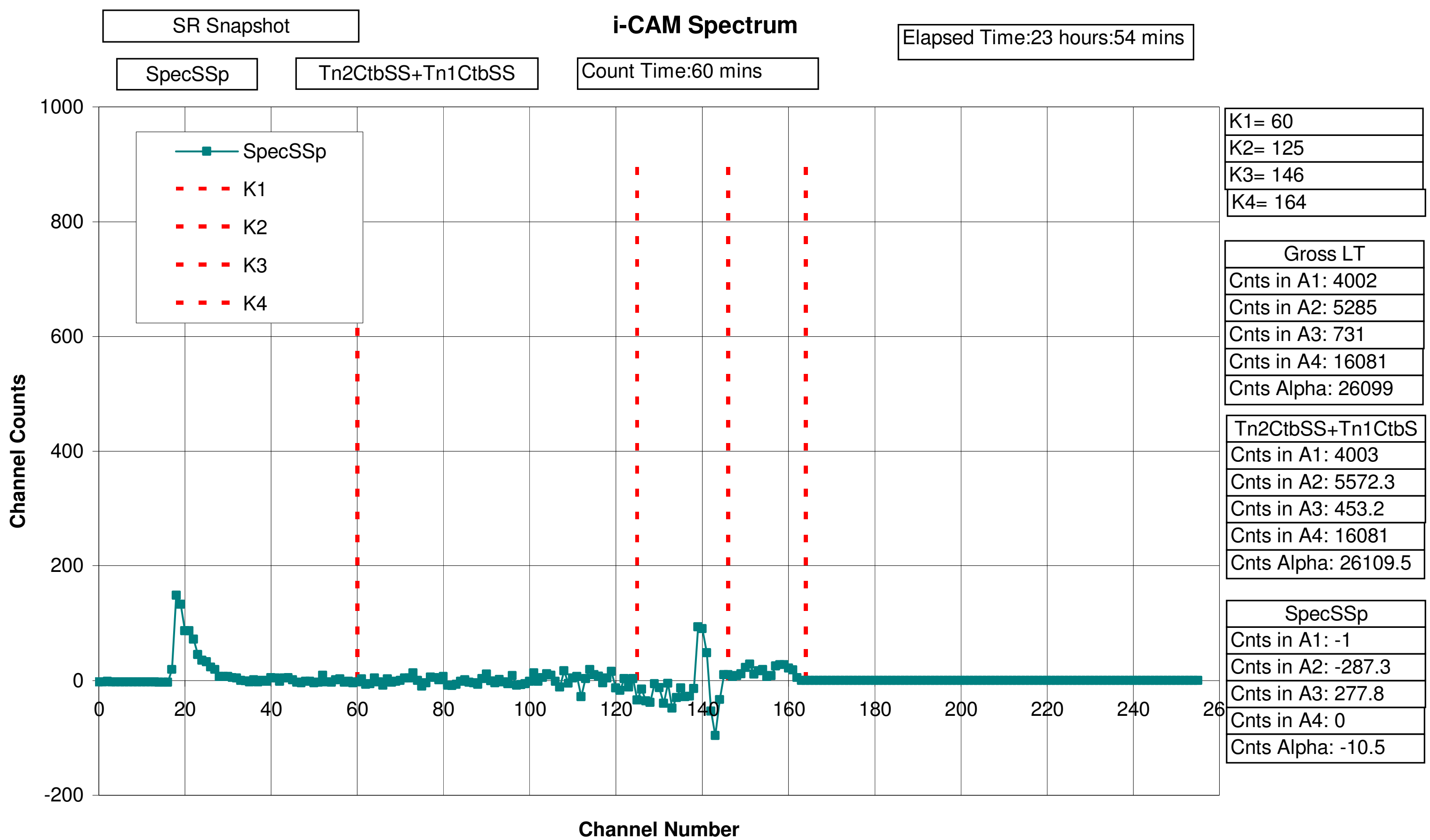
Before arrival of active water drop
Predicted ThC + ThC′ spectrum LOG SCALE



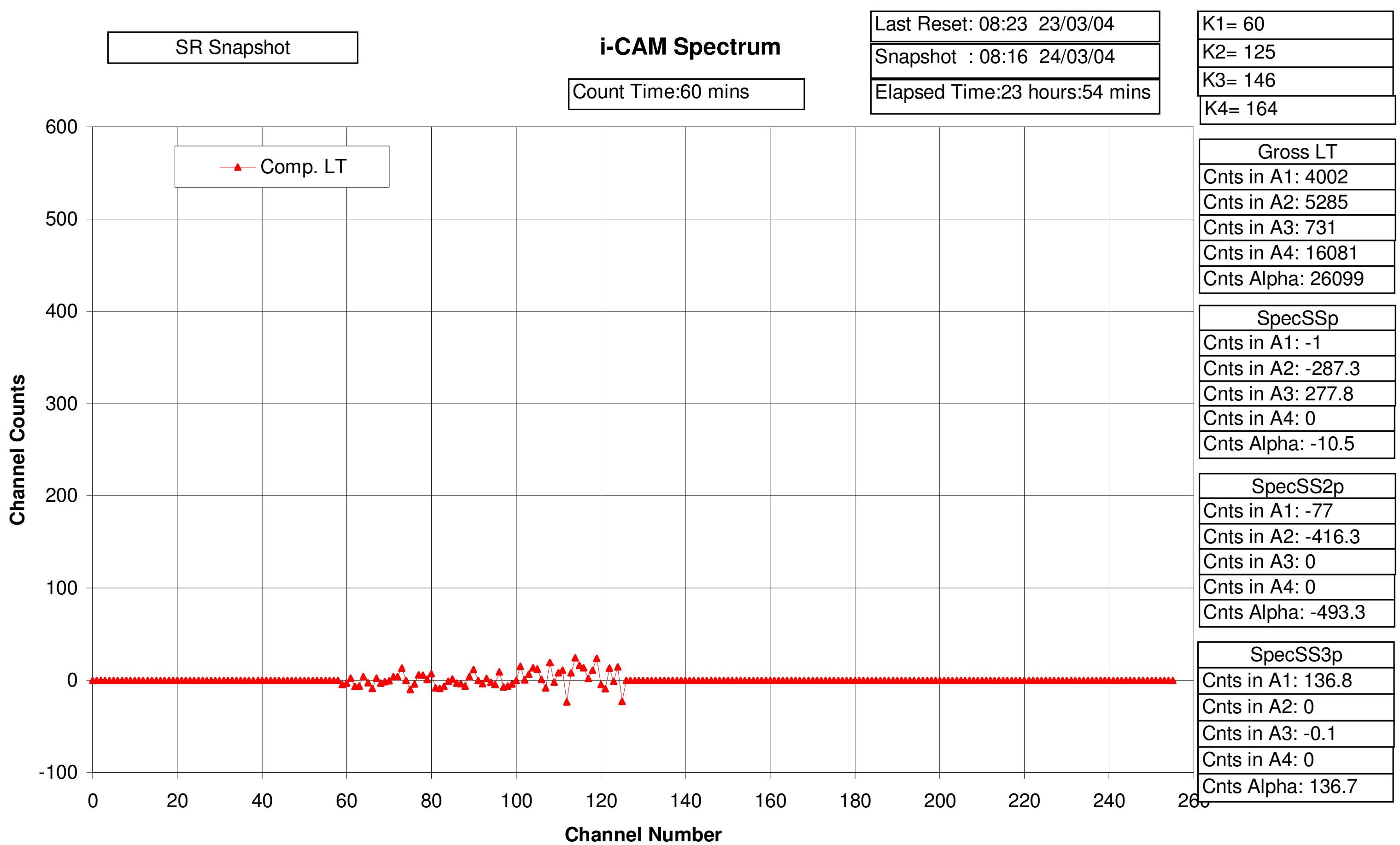
Before arrival of active water drop
Predicted ThC + ThC′ spectrum LINEAR SCALE



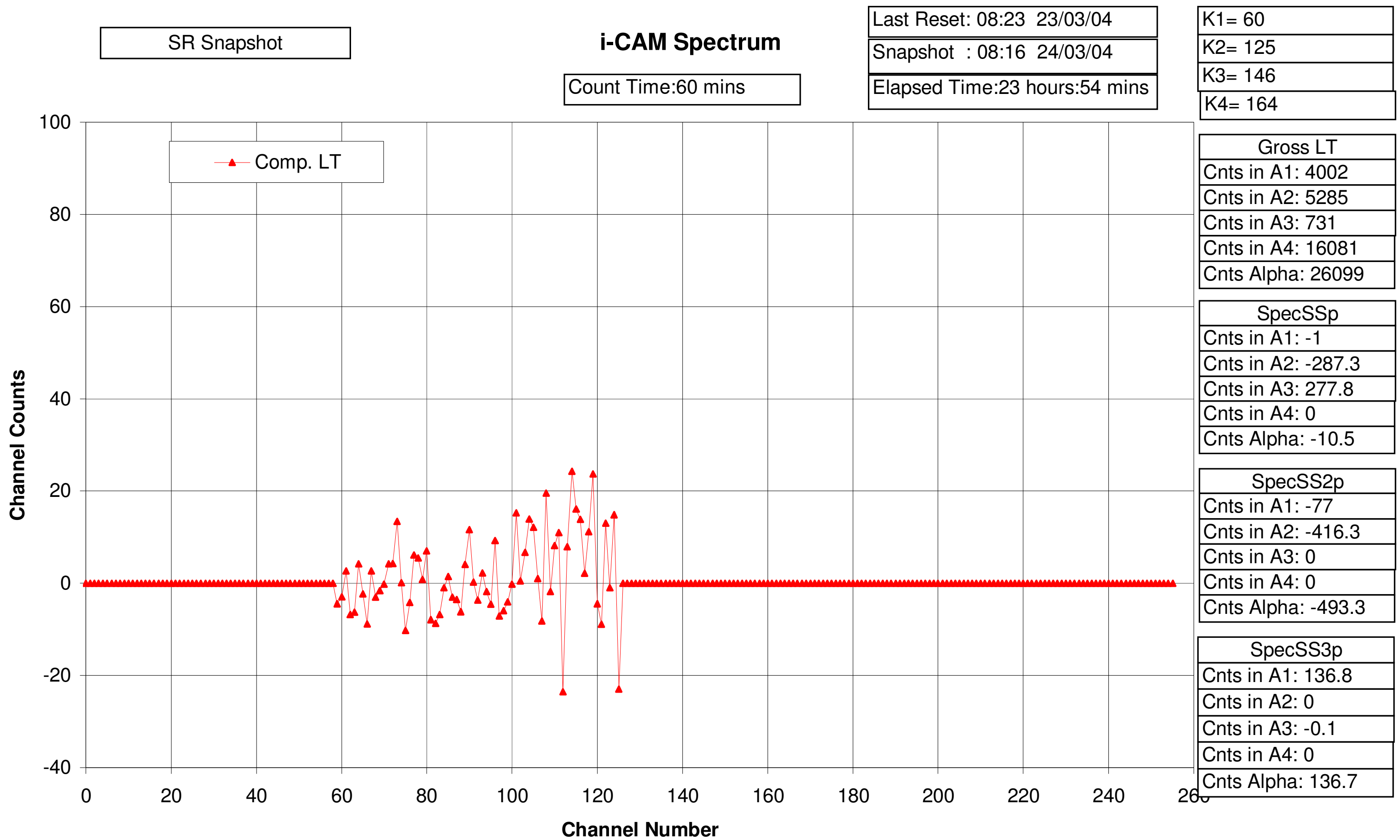
Before arrival of active water drop LINEAR SCALE
Spectrum after thoron progeny spectra stripped



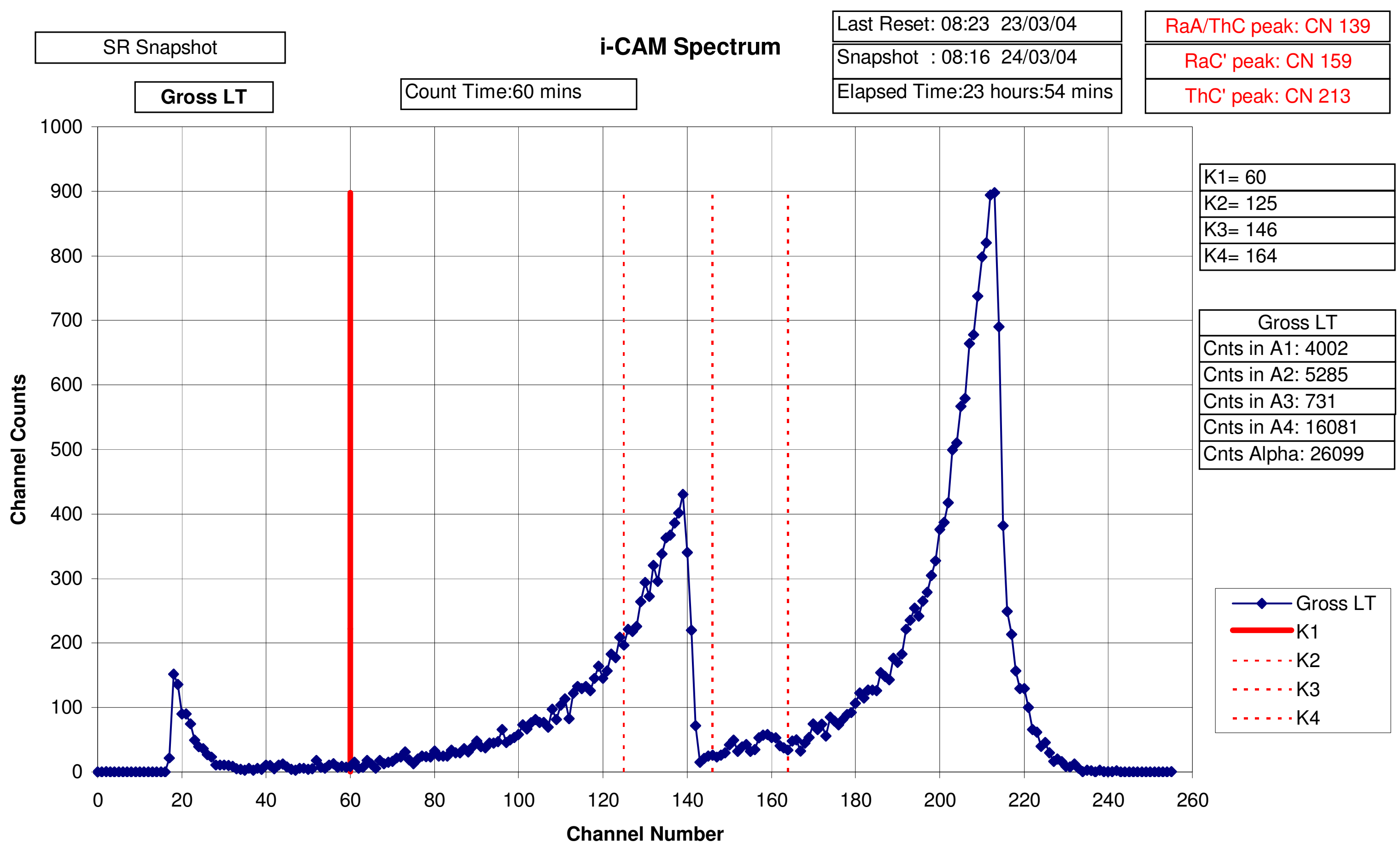
Before arrival of active water drop LINEAR SCALE
Residual spectrum after radon and thoron progeny spectra stripped



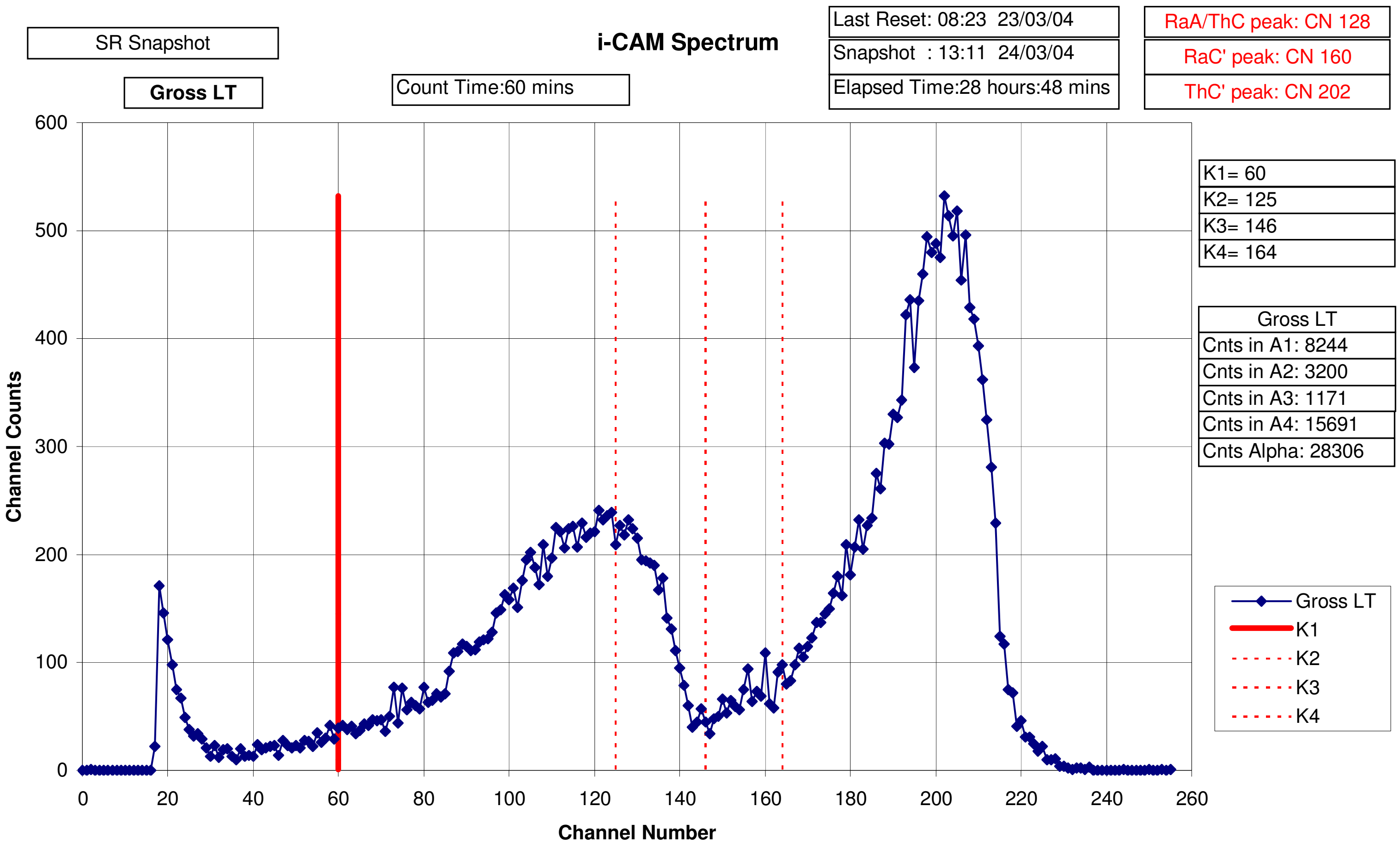
Before arrival of active water drop LINEAR SCALE
Residual spectrum after radon and thoron progeny spectra stripped



Before arrival of active water drop LINEAR SCALE

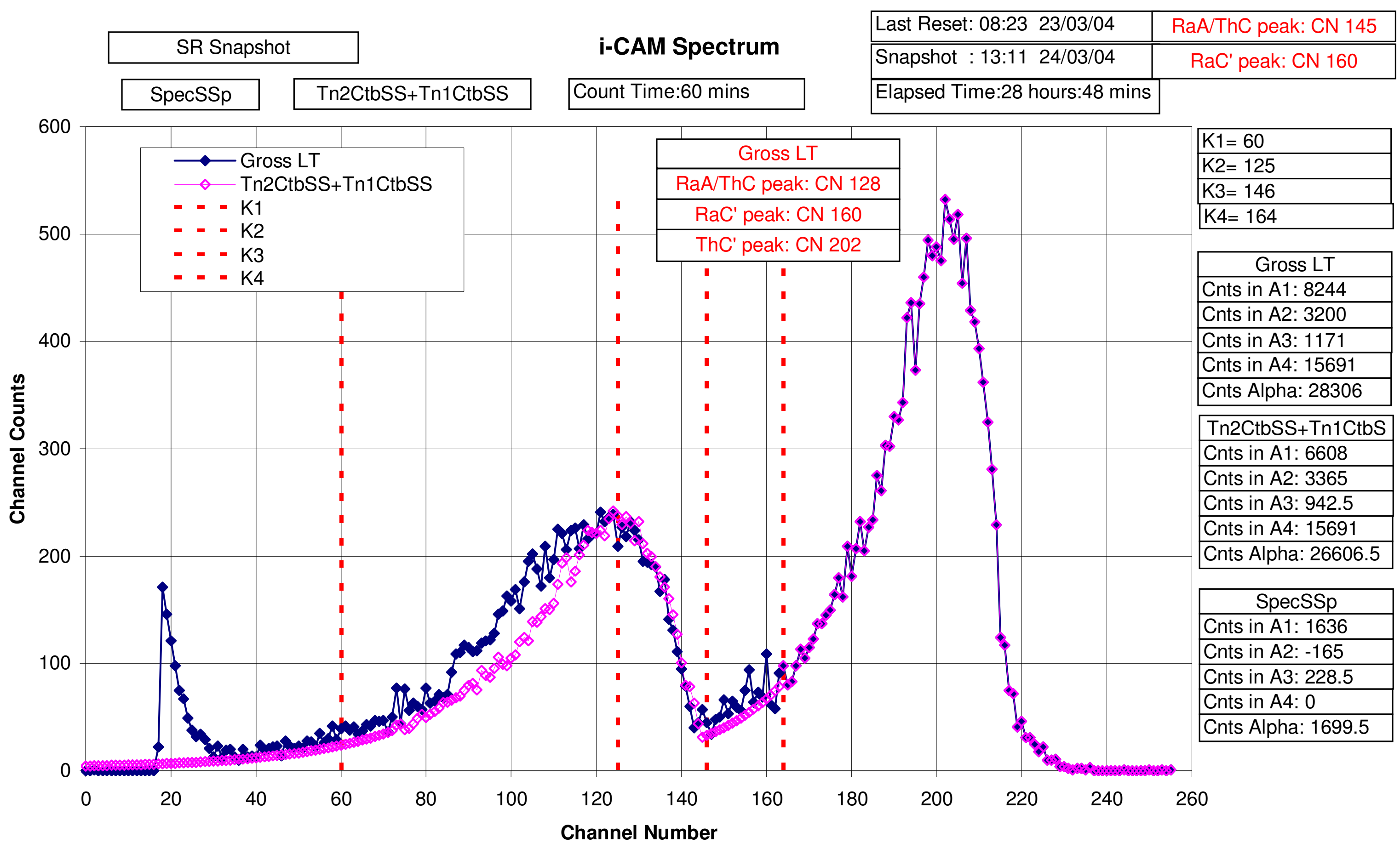


After arrival of active water drop LINEAR SCALE

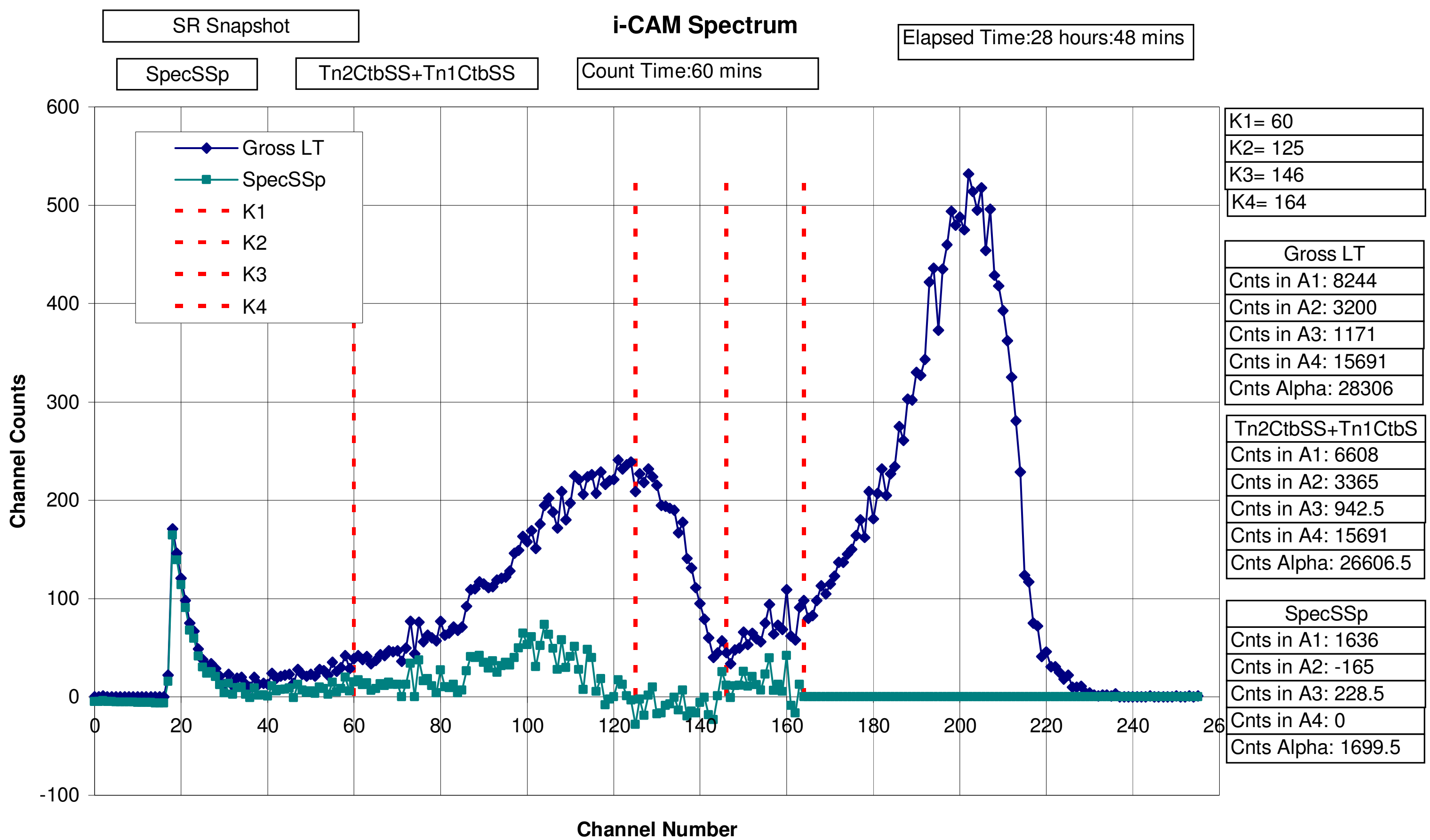


After arrival of active water drop

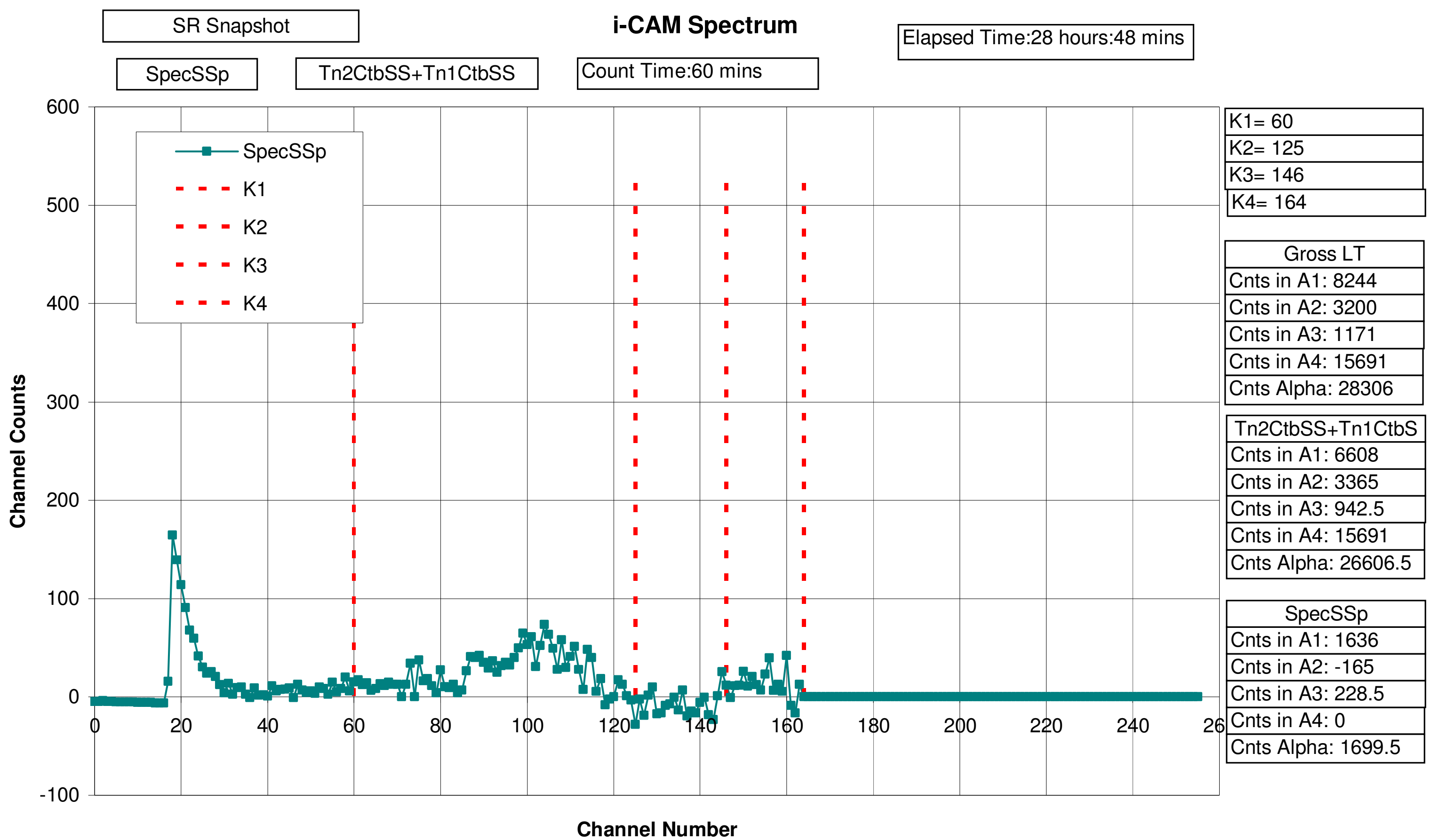
Predicted ThC + ThC′ spectrum LINEAR SCALE



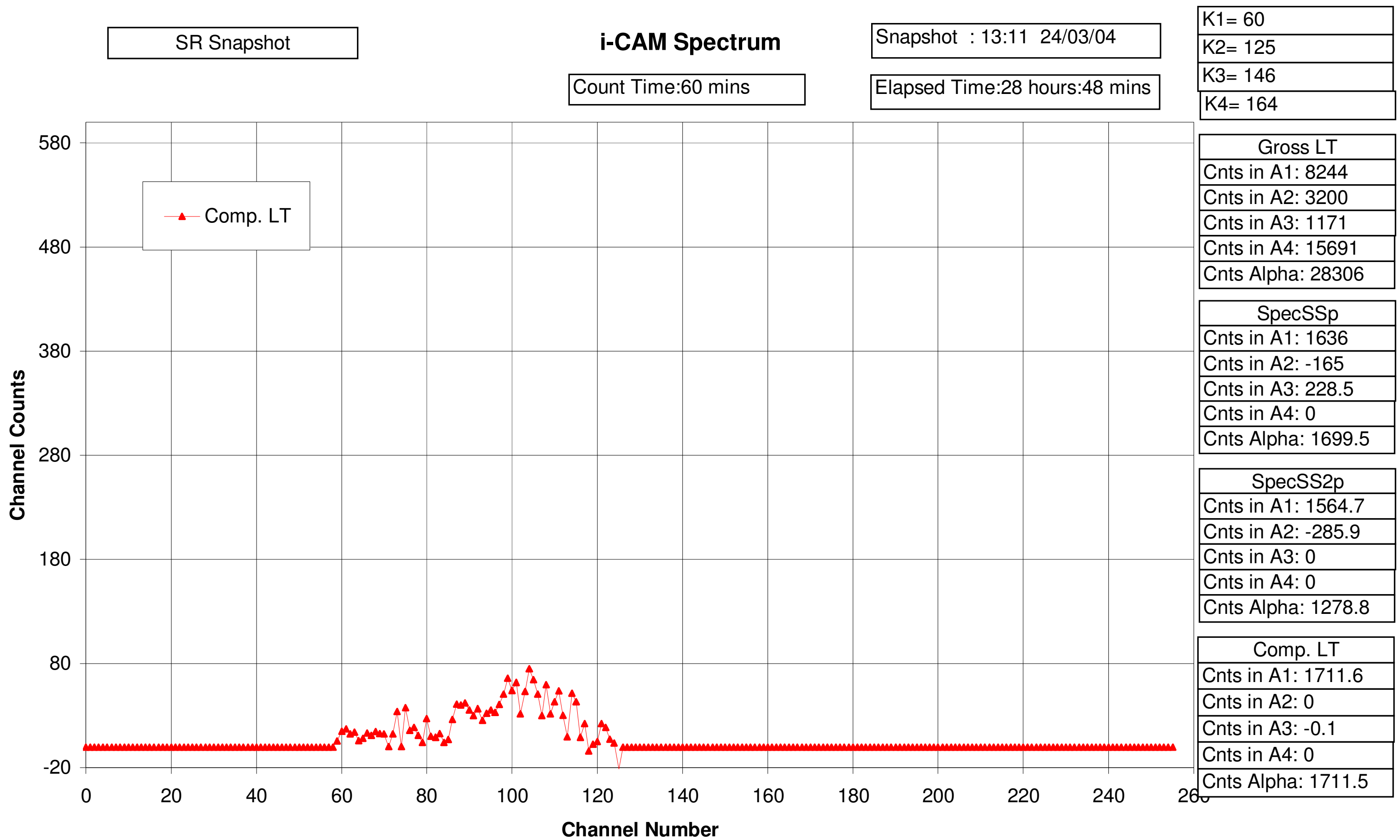
After arrival of active water drop LINEAR SCALE
Spectra before and after thoron progeny spectra stripped



After arrival of active water drop LINEAR SCALE
Spectrum after thoron progeny spectra stripped

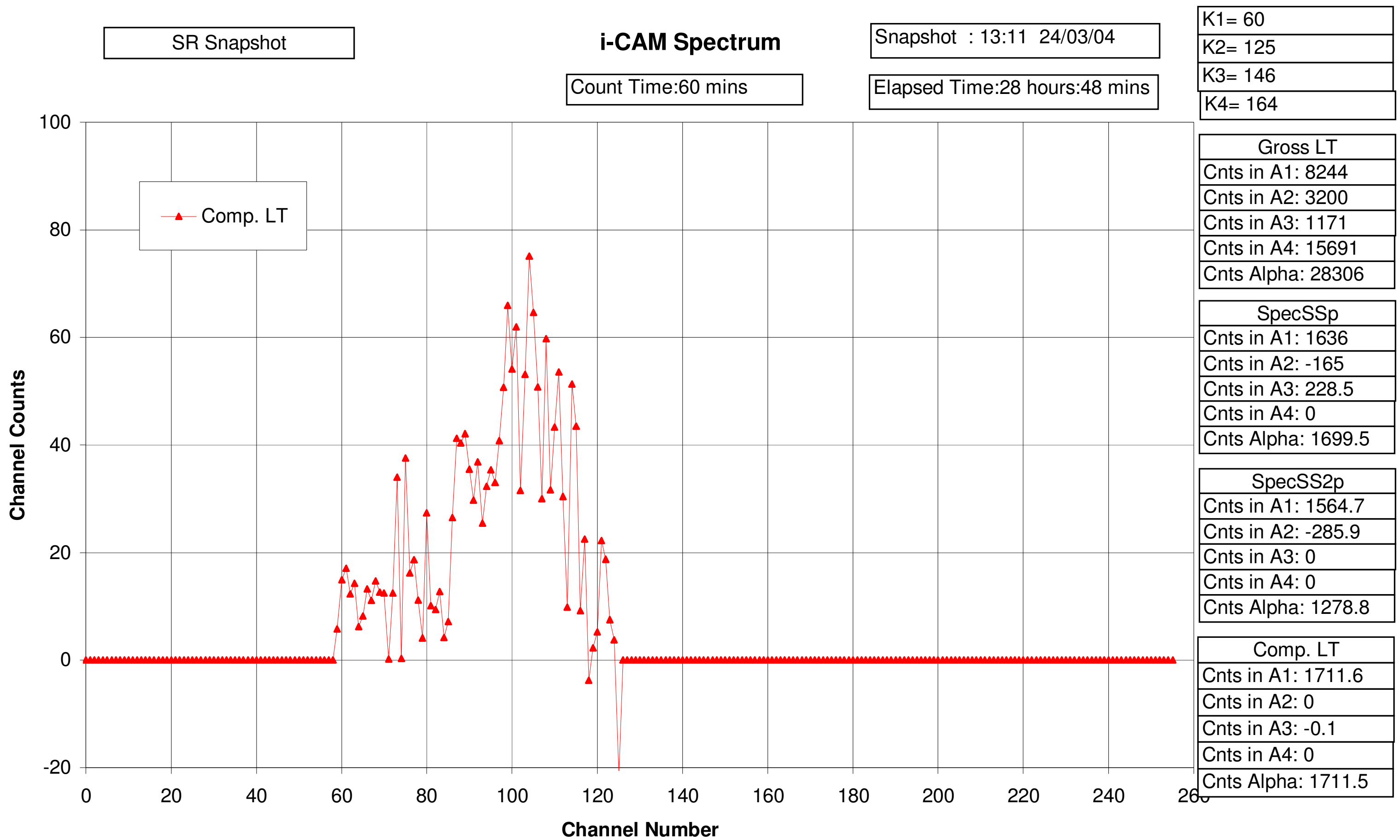


After arrival of active water drop LINEAR SCALE
Residual spectrum after radon and thoron progeny spectra stripped



After arrival of active water drop LINEAR SCALE

Residual spectrum after radon and thoron progeny spectra stripped



Counting the filter 7 days after removing it from the monitor confirmed the that the long lived alpha activity recorded by iCAM in real-time was due to $^{241}Am/^{239}Pu$

Information from papers and reports

The following is a list of some papers and reports that contain useful information

- Pickering (1984) discusses self absorption as alpha energy in escaping from their individual aerosol particles

- Pickering and Fourcaudot (1987) describe how alpha-recoil affects the distribution of aerosols in filters

- Seiler, Newton and Guilmett (1988) give detailed theoretical models for continuous monitoring in a dusty environment

- Moore, McFarland and Rodgers (1993) describe the factors that affect alpha particle detection by continuous air monitors

- Huang, Schery, Alcantara and Rodgers (2002) discuss the influence of dust loading on the alpha-particle energy resolution of continuous air monitors

- Huang, Schery, Alcantara, Dale and Rodgers (2002) provide useful information in a paper on test aerosols

“Investigation of techniques to improve Continuous Air Monitors under conditions of high dust loading in environmental settings” US DoE Report 60163

This is a particularly useful report by Huang et al

Luetzelschwab, Storey, Zraly and Dussinger (200) describes measurements of self absorption of alpha energy in filters but only with sources rather than active aerosols

Grivaud (1992) discusses alpha spectrum resolution obtained with different filters

Grivaud and Pagliardini (2001) give a detailed assessment of Fluoropore FSLW (version 2)

Tony Geryes is two years into a doctoral thesis (Paris University) entitled

“Etude de la degradation de l’energy des particules Alpha dans les filtres de surveillance de la radioactivity atmospheric”

He is supervised by Dr Celine Monsanglant –Louvet at IRSN.

When completed this promises to be a most useful study