NATURAL RADIATION ENVIRONMENT

Science together with industry towards a sustainable and safer environment

Disturbance of radioactive equilibrium in the ashes produced in lignite fired power plants

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Contents

- Lignite combustion for power generation.
- Natural radioactivity and disturbance of radioactive equilibrium in the fly-ash along different stages of the process, within a Power Plant.
- Radon exhalation rate of the fly-ash, within a Power Plant.
- Comparison of radon exhalation rate of ashes produced in different Power Plants.

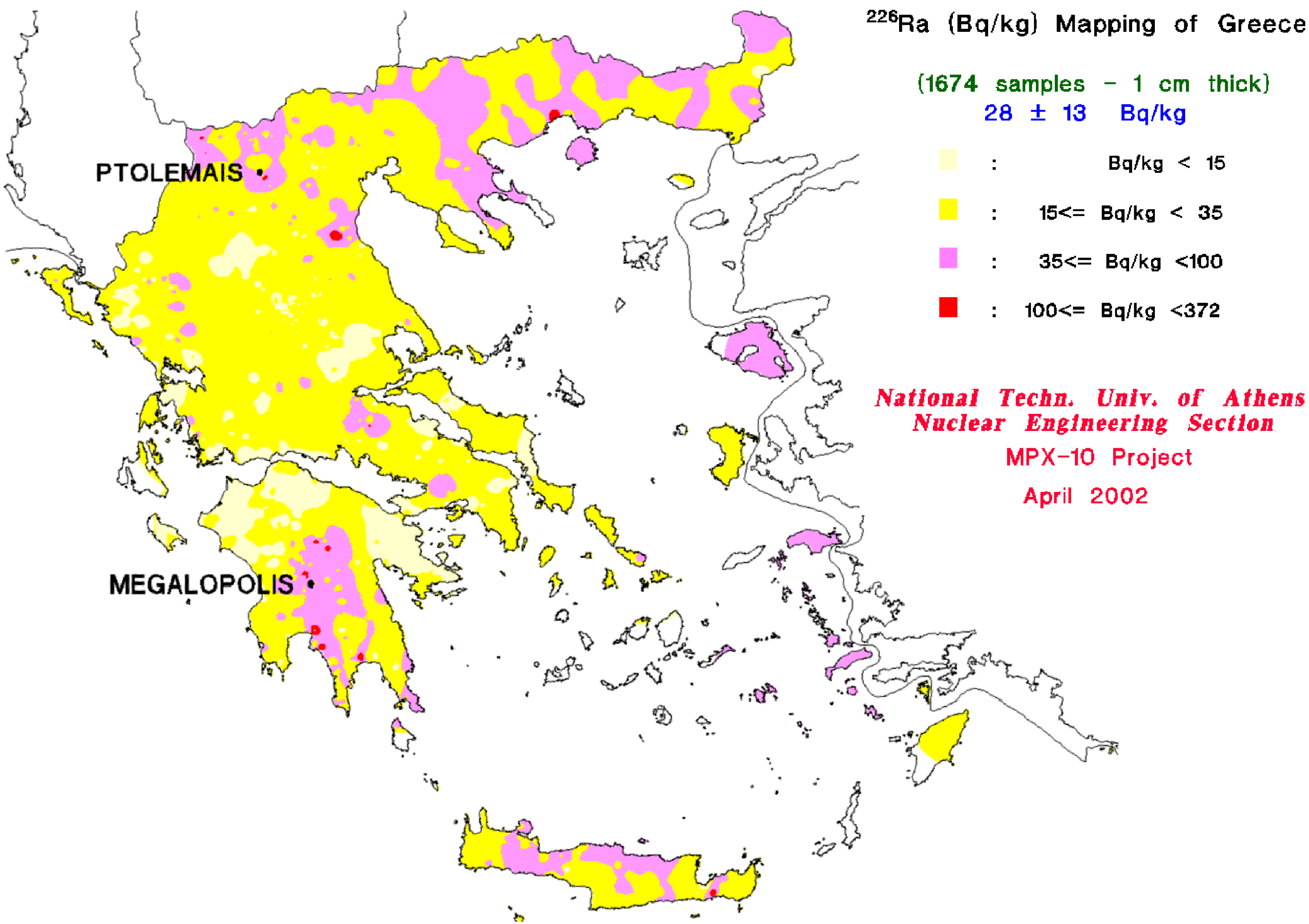
Lignite and Power generation in Greece

- About 65% of electricity in Greece is generated in lignite fired Power Plants (44.5% installed capacity).
- Two major lignite field basins are today under exploitation in Greece:
  - **Ptolemais lignite field** (estimated reserves 2700Mt, 5 Power Plants, 17 Units, installed capacity 4000MW, lignite consumption 51Mt/y, ashes produced 8Mt/y)
  - **Megalopolis lignite field** (estimated reserves 370Mt, 2 Power Plants, 4 Units, installed capacity 850MW, lignite consumption 12.5Mt/y, ashes produced 2.5Mt/y).

Characteristics of the Greek lignite as a fossil fuel

- Greek lignite and especially Megalopolis lignite is classified as low-rank coal with:
  - high water content up to 60%,
  - high ash content up to ~20%
  - low calorific value of ~1kcalkg^{-1}
- Greek lignite and especially Megalopolis lignite has relatively high natural radioactivity content.
- Quartz (SiO_2) content in Megalopolis lignite is relatively high (~ 40% in the ashes) .

^{226}Ra mapping of Greek surface soils (1674 samples)



Disturbance of secular equilibrium within a radioactive series

- **Radioactive equilibrium** in a radioactive series is **disturbed** when radionuclides are removed from a sample, as a result of a physical or chemical processes.
- Laboratory measurements indicate in many cases disturbance of radioactive equilibrium in the ashes produced in lignite-fired Power Plants.

Lignite combustion and ash formation mechanism (1)

- All combustible elements are removed with flue-gases.
- Significant enrichment of all incombustible elements such as the natural radionuclides existing in the ash.
- Partitioning of the produced ash.
  - Bottom ash (slag) falling inside the boiler
  - Fly-ash suspended in the flue-gas
  - Volatile compounds and elements (e.g. Pb)
- Cooling of flue-gases.

Lignite combustion and ash formation mechanism (2)

- Volatile compounds and elements condense on fly-ash particulate, preferentially on the finer particles.
- Most fly-ash is collected in the Electrostatic Precipitators (ESP).
- ESP efficiently remove larger fly-ash particles but are less efficient for vapors and finer particles.

Fly-ash sampling

- In most cases fly-ash radioactivity values are reported irrespectively of the fly-ash sampling point inside the Power Plant and its size distribution.
- In the cases that fly-ash is collected for further industrial use, it is collected from the Power Plant back yard, where all fractions of the fly-ash are mixed.

However :

the size distribution of fly-ash may be of great importance, because it may be related to :

- differences in the natural radioactivity content, and
- differences in the radon exhalation rate.

Aim of this work is :

1. To study the disturbance of radioactive equilibrium, within the nuclides of the Uranium series (^{238}U – ^{226}Ra – ^{210}Pb) in the various fractions of the produced ashes, within Megalopolis-B Power Plant.
2. To study the radon exhalation rate in the various fractions of the produced ashes.
3. To compare the radon exhalation rate of the ashes produced in two different Power Plants, fed with lignite of different origin.

Lignite and ashes samplings conducted during this work

- Repeated sampling of fly-ash from various points along the emission control system of Megalopolis-B Power Plant.
- Representative sampling of lignite, fly-ash and bottom ash produced within a five weeks period in Megalopolis-B Power Plant.
- Sampling of lignite, fly-ash and bottom ash from the Ptolemais area Power Plants.

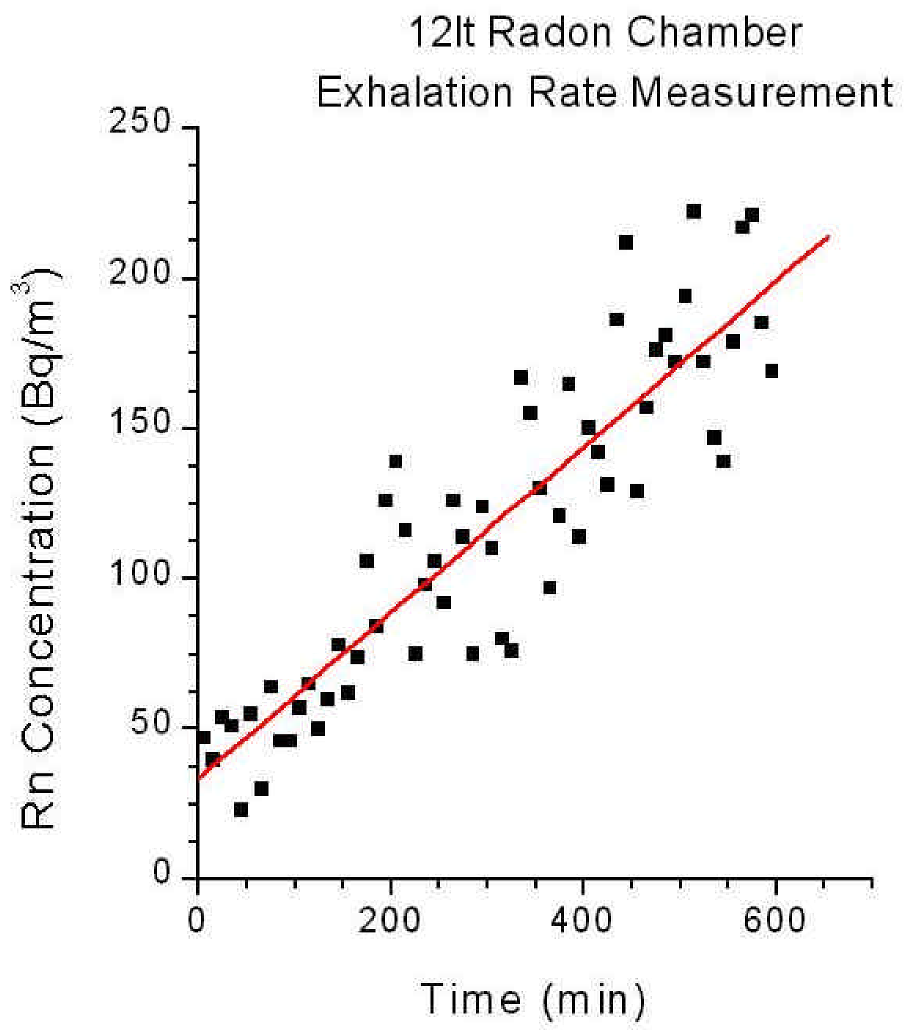
Gamma spectroscopic determination of natural radionuclides

- High-resolution high efficiency Ge detector set-ups, such as LEGe and XtRa detectors are used.
- In-house developed software SPUNAL is used.
- ^{226}Ra is determined both through its daughters in equilibrium and through its 186.25keV photons.
- For the analysis of low energy photons (below 200keV) special techniques are used to take into account for the self-absorption of the photons: 63.29keV (^{234}Th) and 46.52keV (^{210}Pb).

Radon exhalation rate measurements technique

The sample (~300g) is enclosed in a container and the initial growth of radon inside the container is monitored for up to 10h.

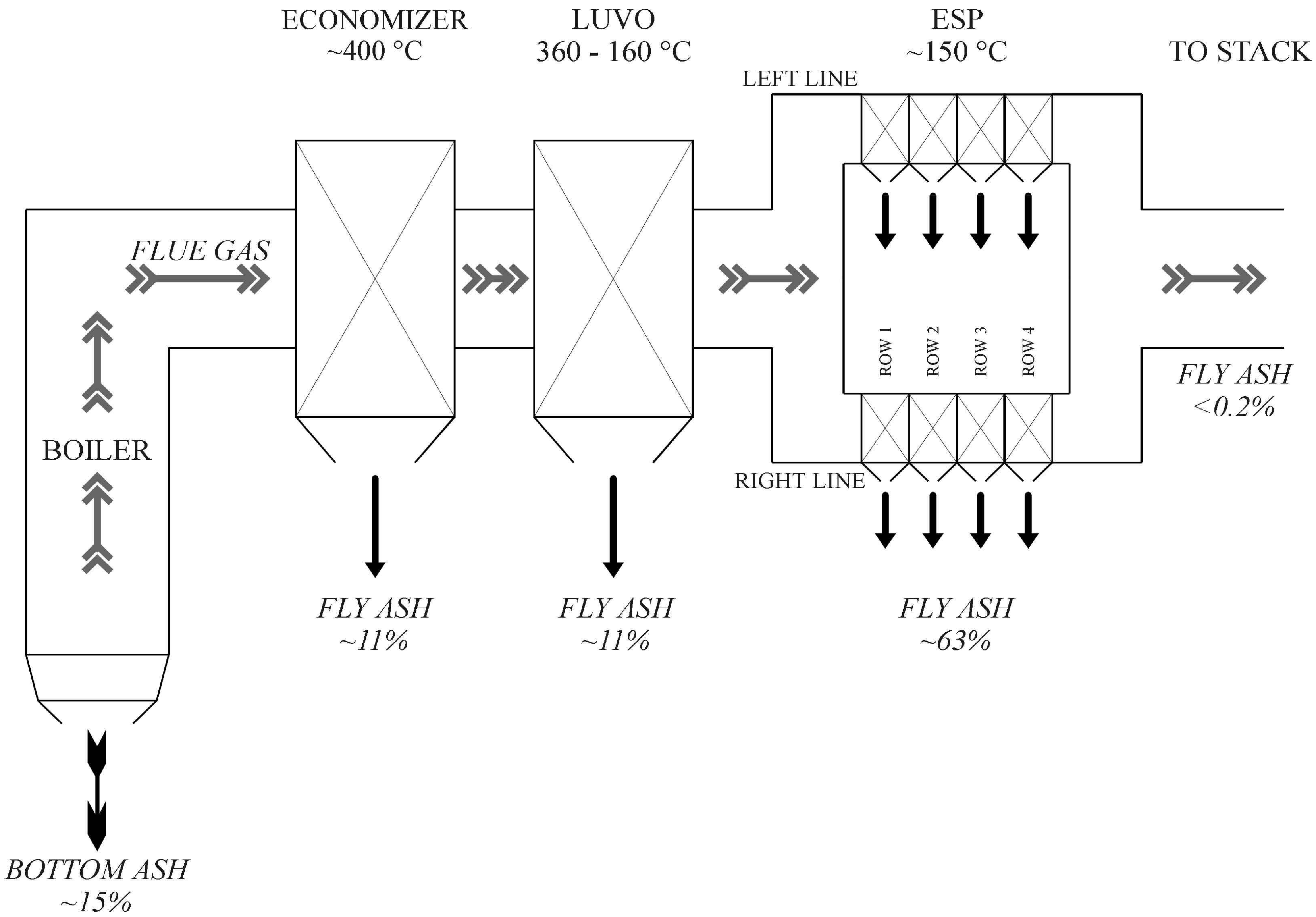
Standard error of the measurement is up to 20%.



Megalopolis-4 Power Plant: Mean activity ± standard deviation (%) of lignite and ashes in $Bqkg^{-1}$ (S.Size 5)

Material	^{238}U (^{234}Th)	^{226}Ra	^{210}Pb	^{232}Th (^{228}Th)	^{40}K
Lignite	**306** ± **13**	**346** ±8	**361** ±10	**19** ±9	**173** ±14
Fly-ash	**964** ±7	**904** ±9	**1158** ±11	**52** ±2	**454** ±11
Bottom-ash	**681** ±4	**662** ±9	**275** ±6	**41** ±5	**405** ±11

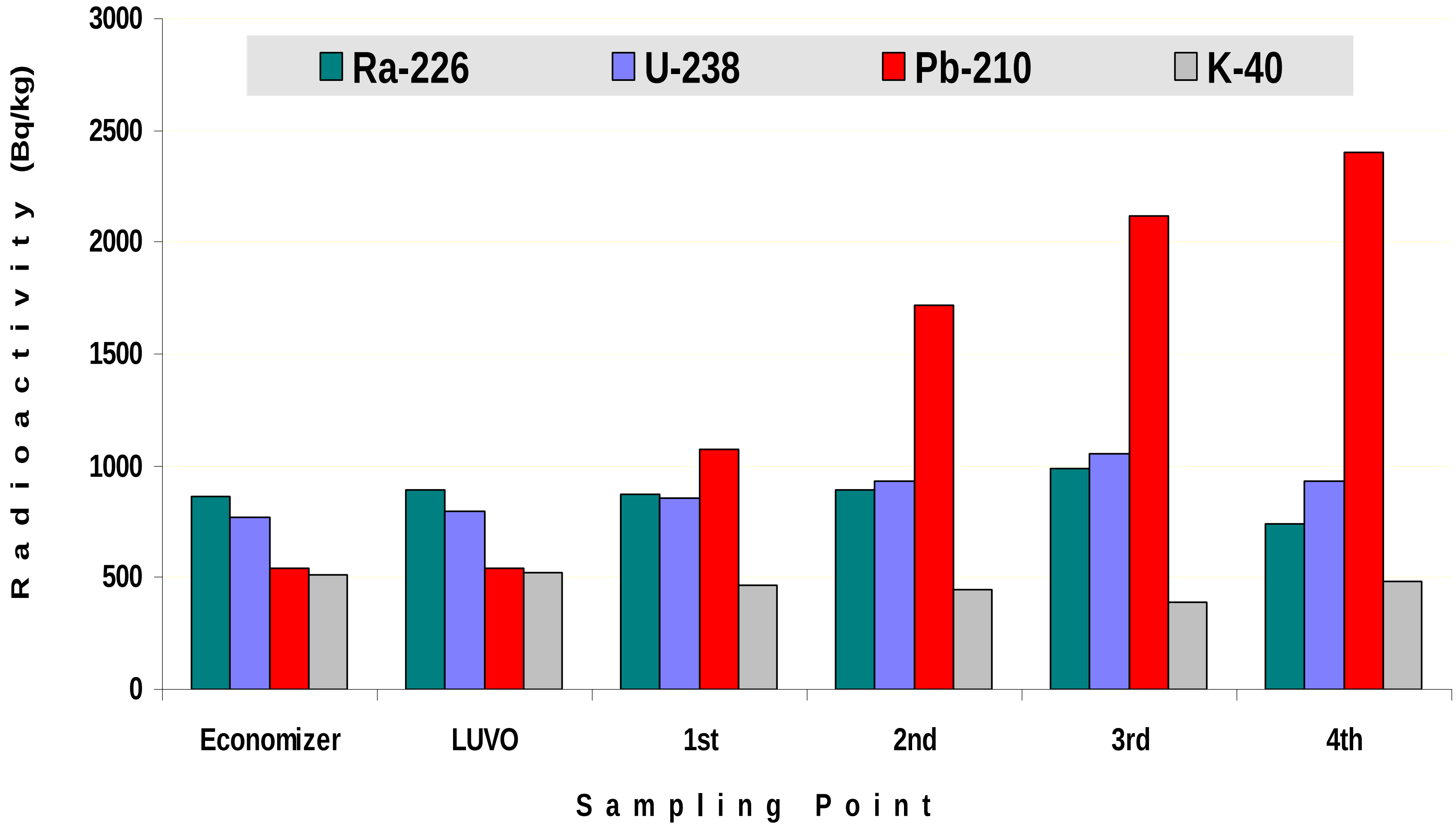
Megalopolis-B Power Plant emission control system (ECS)



Radioactivity of ashes collected along the emission control system of Megalopolis-B Power Plant (mean ± std % in $Bqkg^{-1}$)

Sampling point and sample size	^{238}U	^{226}Ra	^{210}Pb	^{232}Th	^{40}K
ECO (12)	**771 ±7**	**863 ±7**	**538 ±17**	**56 ±5**	**508 ±7**
LUVO (15)	**794 ±10**	**896 ±11**	**539 ±11**	**57 ±6**	**520 ±7**
ESP L1 (27)	**859 ±10**	**876 ±9**	**1068 ±24**	**54 ±6**	**466 ±9**
ESP L2 (16)	**934 ±9**	**893 ±9**	**1717 ±10**	**53 ±6**	**444 ±6**
ESP L3 (13)	**1053 ±16**	**987 ±12**	**2119 ±29**	**53 ±10**	**387 ±10**
ESP L4 (4)	**934 ±23**	**739 ±22**	**2404 ±25**	**49 ±14**	**487 ±6**
ESP R1 (29)	**870 ±12**	**885 ±14**	**1167 ±23**	**54 ±5**	**463 ±10**
ESP R2 (22)	**1001 ±10**	**963 ±10**	**1848 ±19**	**54 ±4**	**441 ±6**
ESP R3 (15)	**1155 ±15**	**1067 ±12**	**2252 ±21**	**55 ±3**	**424 ±3**
ESP R4 (3)	**906 ±32**	**654 ±23**	**2280 ±28**	**54 ±3**	**594 ±11**

Natural Radioactivity of fly-ash collected along the ECS of Megalopolis-B Power Plant



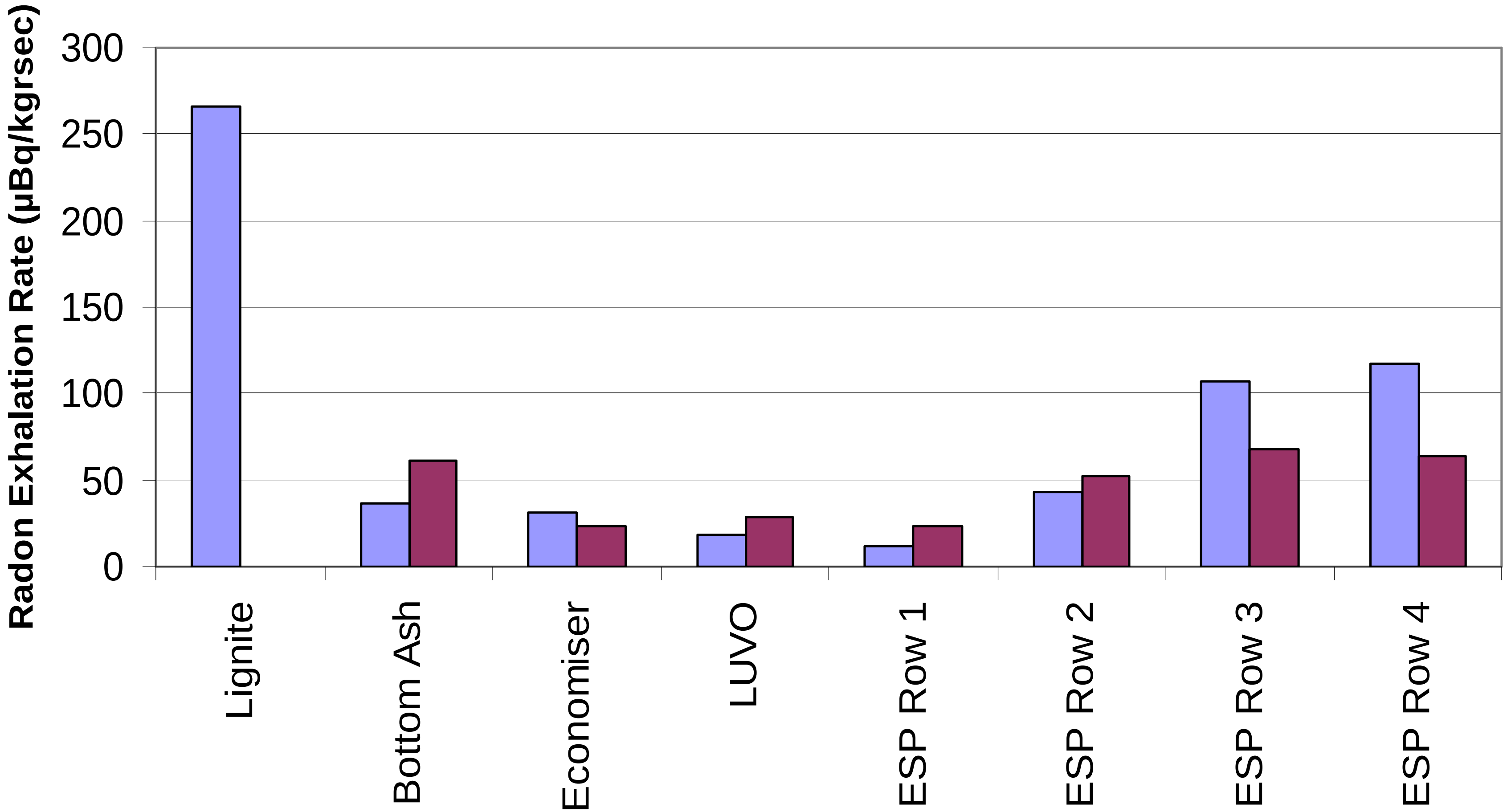
$^{210}Pb/^{226}Ra$ and $^{238}U/^{226}Ra$ activity ratios from Megalopolis-B Power Plant (± standard deviation)

Sampling point	sample size	$^{210}Pb/^{226}Ra$	$^{238}U/^{226}Ra$
ECONOMIZER	12	0.63 ± 0.09	0.89 ± 0.04
LUVO	15	0.60 ± 0.05	0.89 ± 0.03
ESP Left line 1st row	27	1.21 ± 0.23	0.98 ± 0.07
ESP Left line 2nd row	16	1.93 ± 0.18	1.05 ± 0.05
ESP Left line 3rd row	13	2.13 ± 0.43	1.16 ± 0.07
ESP Left line 4th row	4	3.12 ± 0.22	1.23 ± 0.05
ESP Right line 1st row	29	1.32 ± 0.24	0.99 ± 0.06
ESP Right line 2nd row	22	1.94 ± 0.43	1.04 ± 0.07
ESP Right line 3rd row	15	2.12 ± 0.42	1.08 ± 0.04
ESP Right line 4th row	3	3.47 ±0 .18	1.37 ± 0.13

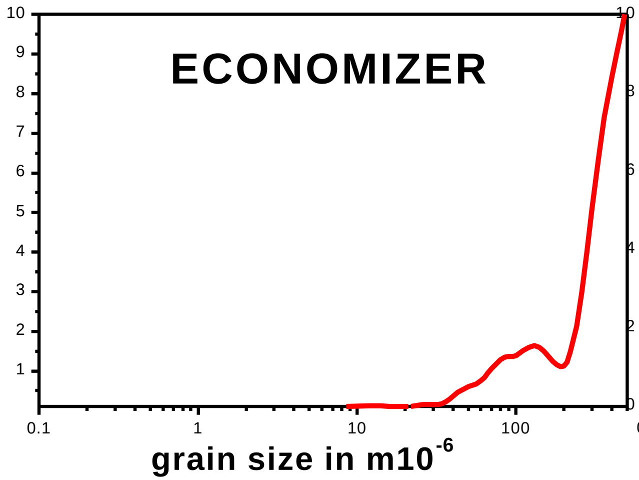
Fly-ash radon exhalation rate

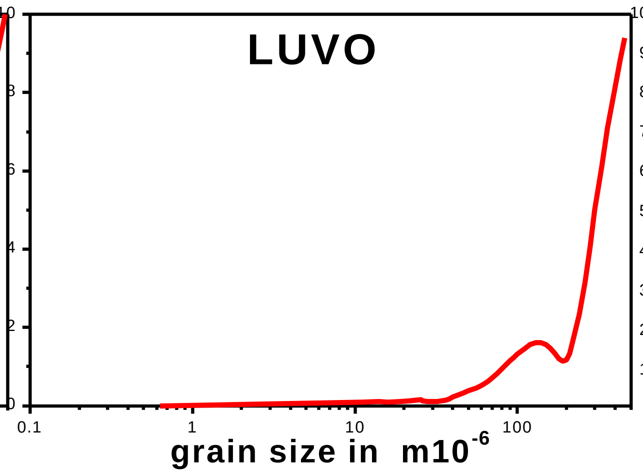
Sampling point	Mean diam. (µm)	^{226}Ra (Bqkg^{-1}) ± **std err (%)**	^{222}Rn exhalation µBqs^{-1}kg^{-1}	^{222}Rn exhalation nBqs$^{-1/}$Bq^{226}Ra
ECO.	**283**	**856 ±4**	**24**	**28**
ECO.	**271**	**763 ±1**	**19**	**25**
LUVO	**287**	**874 ±1**	**14**	**16**
LUVO	**316**	**853 ±1**	**23**	**27**
ESP-row 1	**266**	**932 ±2**	**17**	**18**
ESP-row 1	**190**	**663 ±2**	**23**	**35**
ESP-row 2	**43**	**980 ±2**	**38**	**39**
ESP-row 2	**22**	**1030 ±2**	**45**	**44**
ESP-row 3	**96**	**1155 ±2**	**76**	**66**
ESP-row 3	**21**	**1176 ±2**	**64**	**54**
ESP-row 4	**18**	**938 ±2**	**113**	**121**
ESP-row 4	**21**	**932 ±3**	**64**	**69**

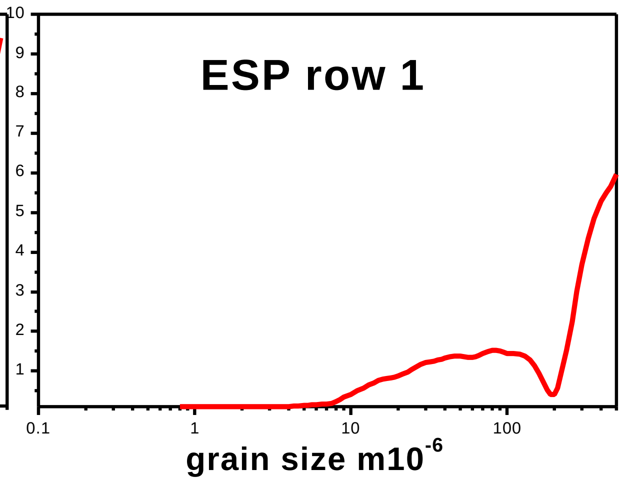
Radon exhalation rate ($\mu Bq s^{-1} kg^{-1}$)

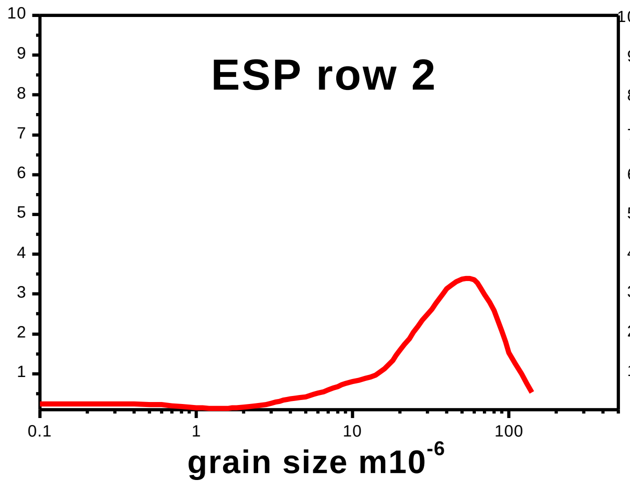


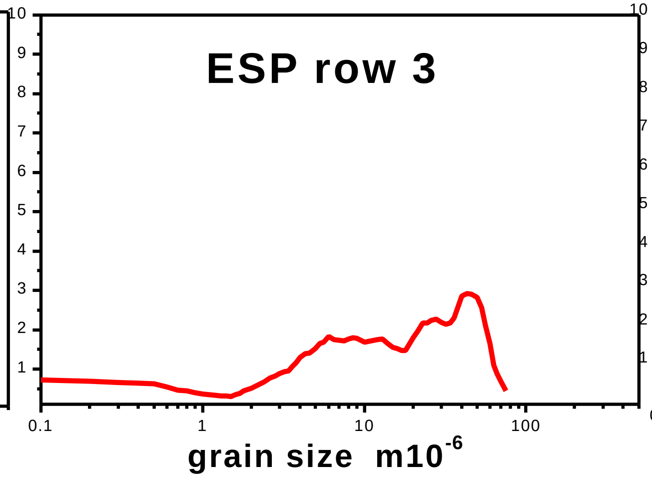
Size distribution of fly-ash

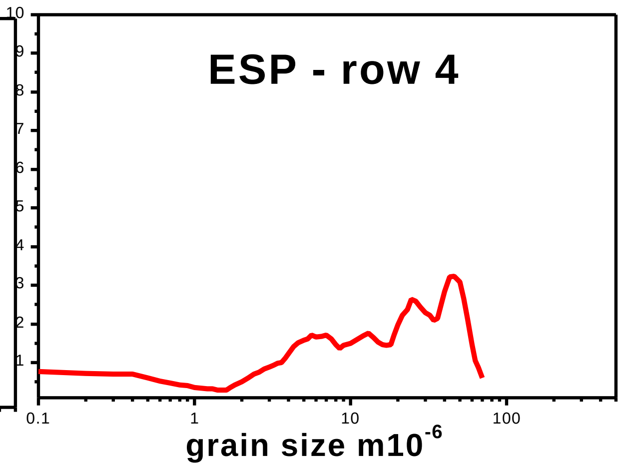




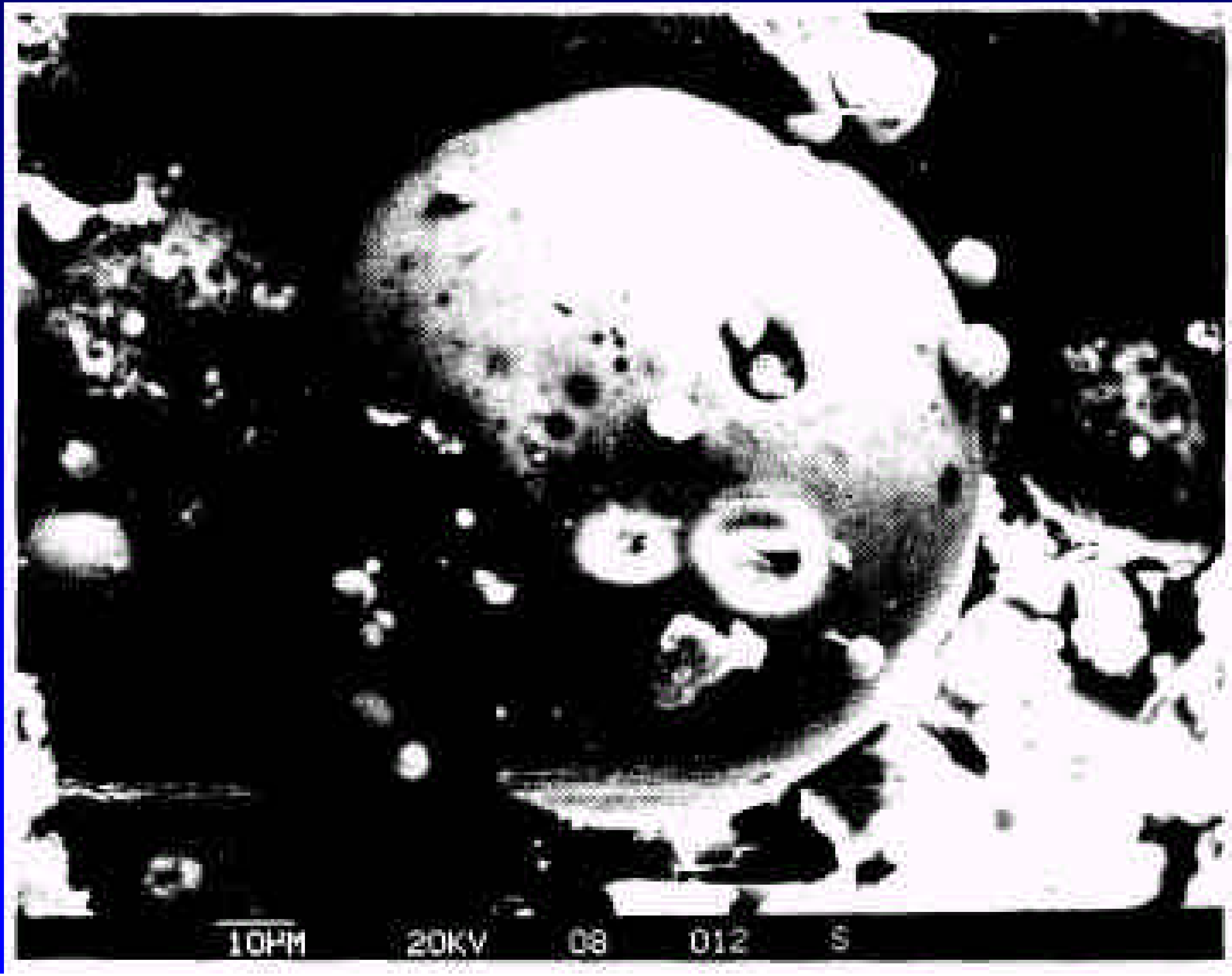








Megalopolis fly-ash particle <5μm



Comparative results of lignite and ashes from two Power Plants (±standard error %)

	Megalopolis Power Plant			Ptolemais Power Plant		
	Lignite	Fly-ash	Bottom ash	Lignite	Fly-ash	Bottom ash
^{226}Ra ($Bqkg^{-1}$)	295 ±1	1003 ±1	583 ±1	88 ±1	309±1	127 ±2
1982-87 samplings mean ± st.dev	314 ±52	807 ± 138	546 ±77	83 ±6	261±19	114 ±16
radon exhalation ($\mu Bqs^{-1}kg^{-1}$)	266±3	35 ± 3	31 ± 3	126 ±6	4 ±25	23 ±25
radon exhalation ($nBqs^{-1}/Bq$ ^{226}Ra)	902 ±4	35 ±3	53 ±4	1432 ±6	13 ±25	181±25

Conclusions (1)

1. Radioactivity of the ashes produced in lignite burning power plants highly depends on the sampling location inside the plant.
2. Radioactive equilibrium among the nuclides of ^{238}U series may be significantly disrupted in the produced ashes.
3. ^{238}U may be slightly enriched compared to ^{226}Ra in the fly-ash collected at the coldest parts of the ESP.

Conclusions (2)

4. ^{210}Pb in fly-ash is in some cases highly enriched compared to ^{226}Ra (up to 3.5 times).
5. ^{210}Pb activity of fly-ash collected in the coldest parts of the emission control system may be up to six times higher than that collected in the hotest parts. This is important for dosimetric calculations.

Conclusions (3)

6. Radon exhalation rate from fly-ash depends on the sampling point along the emission control system of the Power Plant. Radon exhalation is higher towards the coldest parts of ESP, where the finest fly-ash particles, which have the higher surface/volume ratios, are collected.
7. Radon exhalation of fly-ash is much lower than that of lignite, this is more pronounced if exhalation is normalized to ^{226}Ra content. This difference should be attributed to the crystalisation of the produced ashes.