Università degli studi di Ferrara
Master’s degree in physics

Study of the rain-induced gamma activity due to atmospheric radon daughters

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Outline

Origin of the atmospheric ^{222}Rn daughters

- Study of ^{222}Rn formation in the soil
- Study of the physical processes controlling the transport of ^{222}Rn from the soil into the atmosphere

Scavenging of the atmospheric ^{222}Rn daughters by precipitation

- Study of the physical properties of ^{222}Rn daughters
- Comparison between different processes controlling the scavenging of the atmospheric ^{222}Rn daughters by precipitation

Analysis of the rain-induced gamma activity on the ground

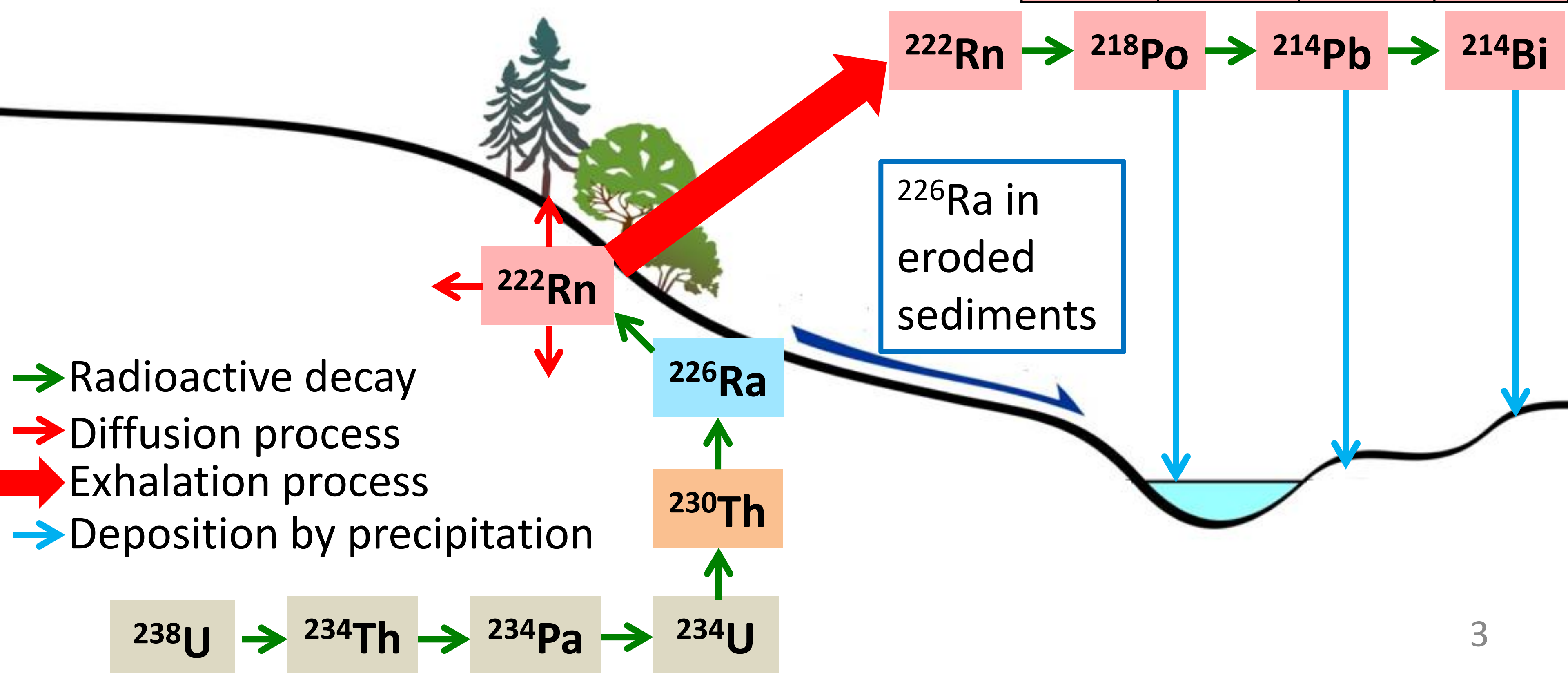
- Experimental site and setup
- Methodology of the data analysis
- Mathematical formalism
- Fit of an analytical model to the rain-induced gamma activity recorded during two different rain events

Primordial radioactive source of atmospheric ^{222}Rn and its daughters

Radon ^{222}Rn and its daughters are generated by the radioactive decay of uranium ^{238}U which undergoes a chain of successive decays

➡ The ^{238}U decay chain is divided in different sub-series:

Element →	**^{238}U**	**^{234}Th**	**^{234}Pa**	**^{234}U**	**^{230}Th**	**^{226}Ra**	**^{222}Rn**	**^{218}Po**	**^{214}Pb**	**^{214}Bi**
Half-life →	**4.468 × 10^9 y**	**24.10 d**	**6.70 h**	**245.5 × 10^3 y**	**75.38 × 10^3 y**	**1.6 × 10^3 y**	**3.824 d**	**3.10 min**	**26.7 min**	**19.9 min**



Properties of ^{222}Rn

Useful physical and chemical properties of ^{222}Rn for studying its transport from the soil into the atmosphere:

Radon is a colorless and odorless radioactive gas

1. Very limited chemical activity (noble gas)
2. Half-life of 3.824 days
3. Poor solubility

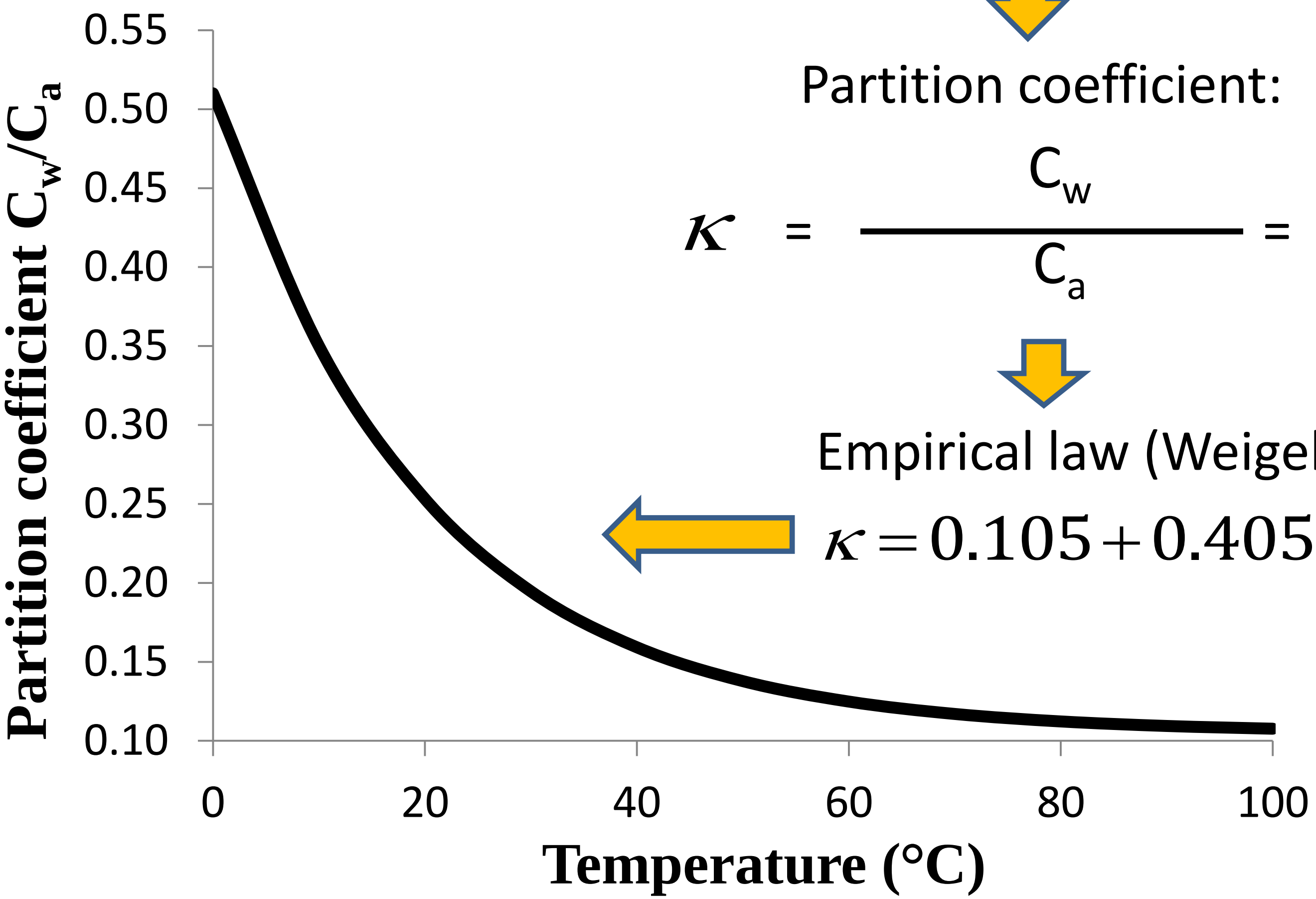
Partition coefficient:

$$\kappa = \frac{C_w}{C_a} = \frac{\text{Concentration of } ^{222}\text{Rn in water (Bq m}^{-3}\text{)}}{\text{Concentration of } ^{222}\text{Rn in gas phase (Bq m}^{-3}\text{)}}$$

Empirical law (Weigel equation):

$$\kappa = 0.105 + 0.405 \times e^{-0.05027 \times T}$$

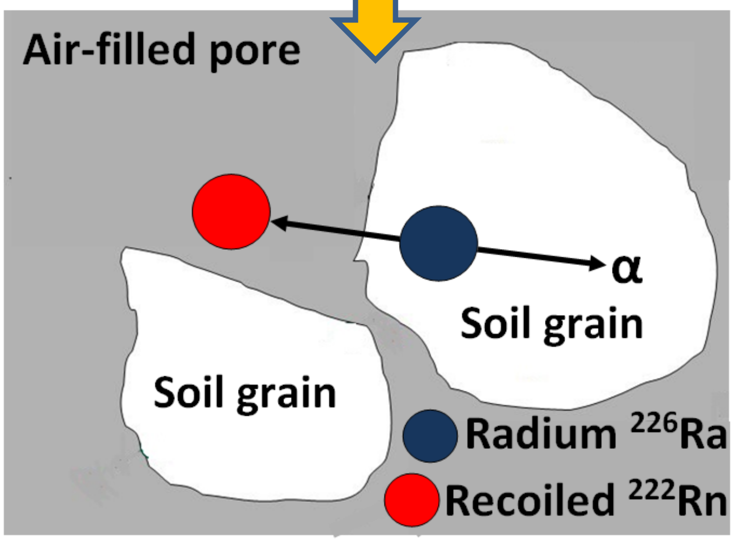
where T : water temperature (°C)



Transport of ^{222}Rn from the soil into the atmosphere

Radon generation (Radium decay) → Radon in air-filled pore → Migration through soil pores → Escape into the atmosphere

Alpha-recoil emanation process



Diffusive transport in soil

Fick's law:

$$j^d = -D_e \nabla C$$

J^d: diffusion flux density ($Bq\ m^{-2}\ s^{-1}$)

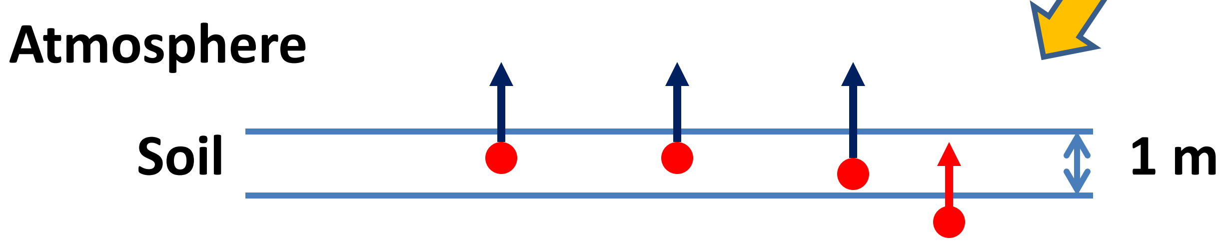
C : radon activity concentration ($Bq\ m^{-3}$)

D_e: effective diffusion coefficient ($m^2\ s^{-1}$)

Exhalation process

Diffusive transport into the atmosphere

^{222}Rn diffusing into the atmosphere is mainly originated from the first meter below the soil



Vertical distribution of ^{222}Rn in the troposphere

$$\frac{\partial C}{\partial t} = \nabla \cdot (K \nabla C) - \vec{w} \cdot \nabla C - \lambda C$$

Turbulent diffusion — Convection — Radioactive decay

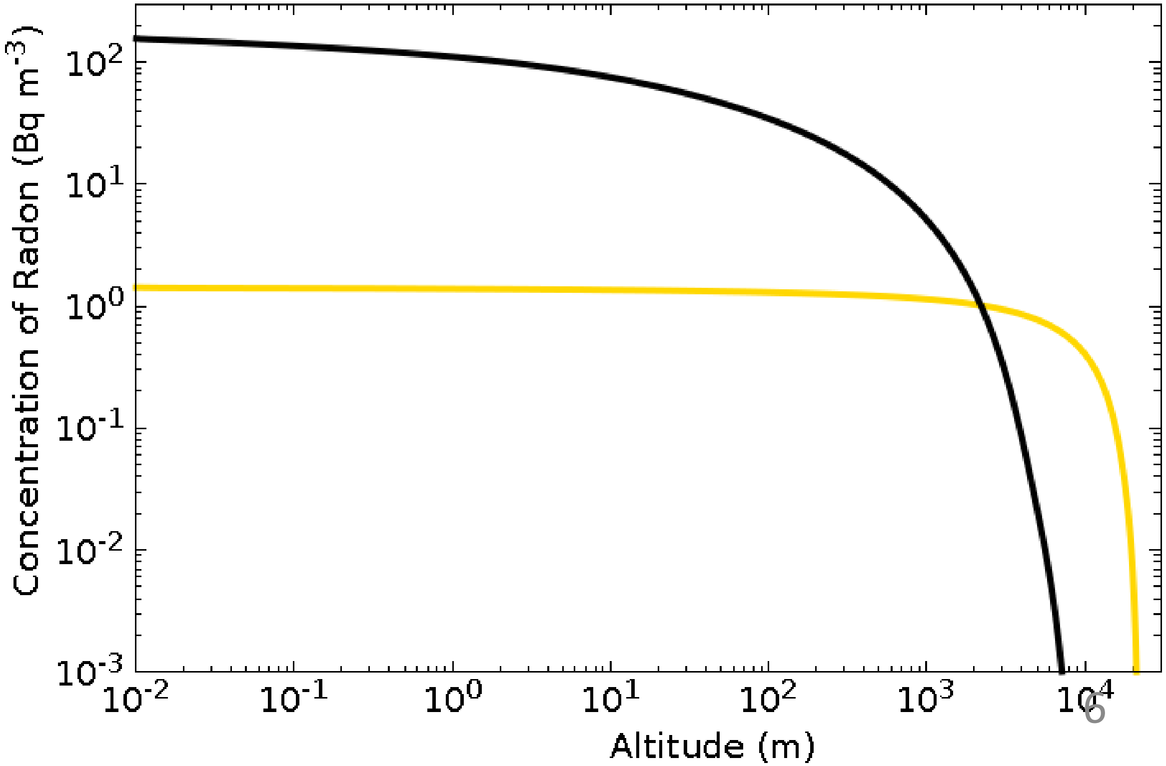
λ : decay constant of ^{222}Rn (s^{-1})
C : radon activity concentration ($Bq\ m^{-3}$)
w : vertical wind speed ($m\ s^{-1}$)
K : turbulent diffusion coefficient ($m^2\ s^{-1}$)

Turbulent diffusion coefficients exceed those in the soil by a factor ranging from 10^6 to 10^{15}

Assuming an horizontal isotropic distribution of ^{222}Rn, the steady-state vertical distribution of ^{222}Rn

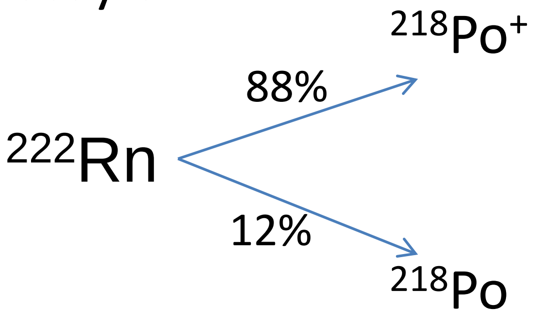
Atmospheric mixing conditions:

- Strong vertical mixing
- Weak vertical mixing



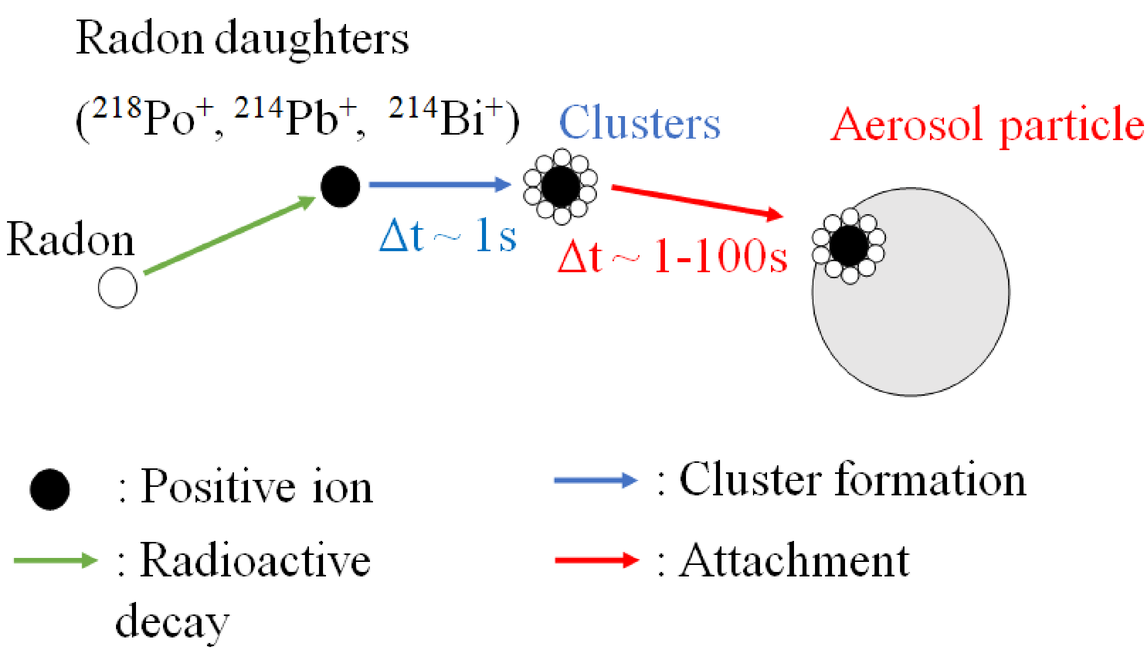
Short-lived ^{222}Rn daughters in the atmosphere

α decay of ^{222}Rn:



Radon daughters are mainly formed and are likely to remain in positive charged states

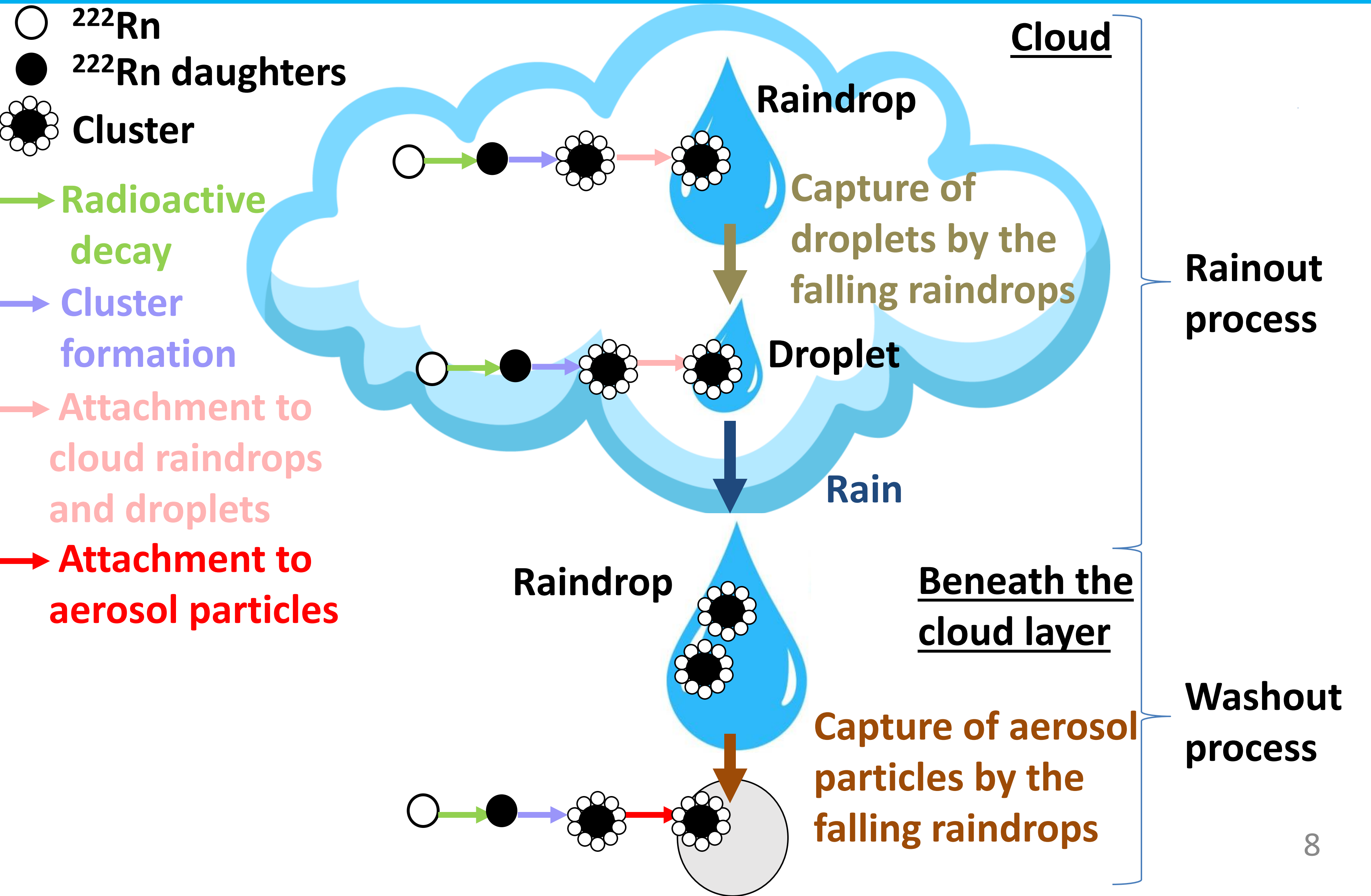
Radon daughters	^{218}Po	^{214}Pb	^{214}Bi
Decay	α	β-γ	β-γ
Half-life (min)	3.10	26.7	19.9
1st ionization potential (eV)	8.34	7.42	7.29



Clusters (with size range 0.5 – 5 nm): agglomeration of polar molecules (H_2O) around ^{222}Rn daughters ions

Attachment to aerosol particles with size range 0.1 – 2 μm

Scavenging of the ^{222}Rn daughters from the atmosphere by precipitation



Dominant mechanism controlling the scavenging of ^{222}Rn daughters from atmosphere by precipitation

The efficiency of the washout process can be described by the washout ratio:

$$\mathbf{WR} = \frac{\text{Concentration of } ^{222}\text{Rn daughters in rainwater on the ground (Bq m}^{-3})}{\text{Concentration of } ^{222}\text{Rn daughters in ground-level air (Bq m}^{-3})}$$

If washout process were the dominant mechanism:

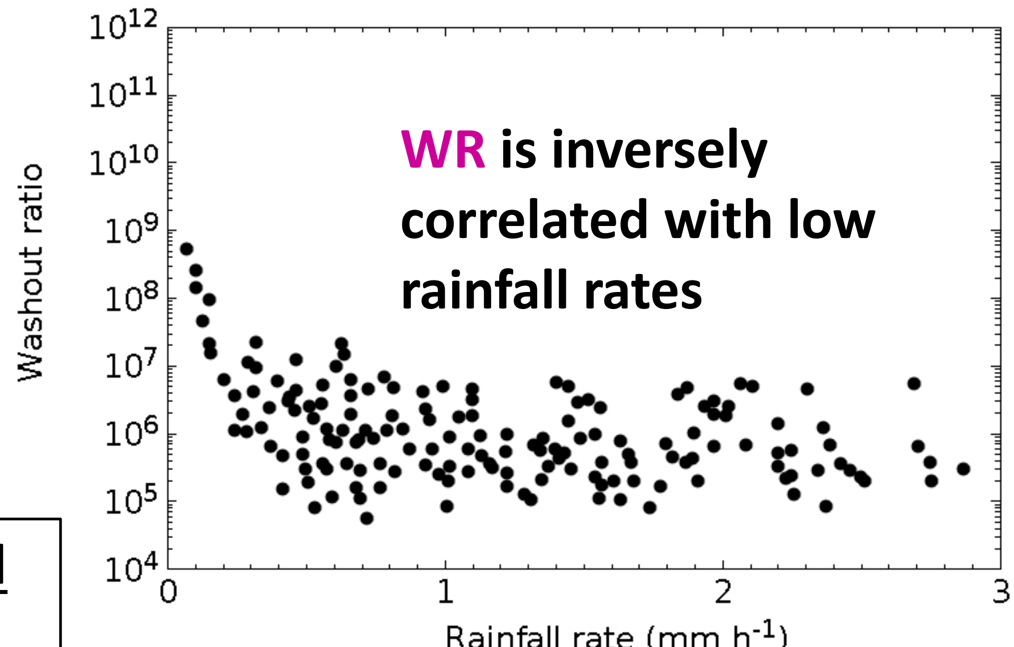
→ **WR** ↗ if rainfall rate ↗

^{222}Rn daughters are attached to sub-micron and micron aerosol particles

→ Low capture efficiency of aerosol particles by the falling raindrops

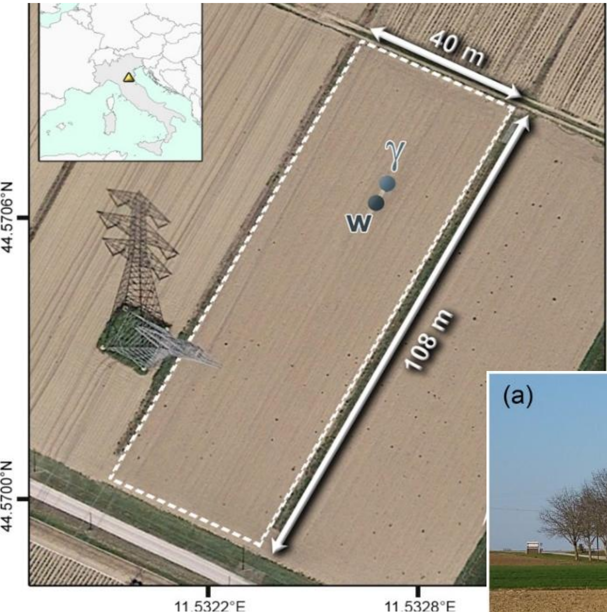
↓

<u>During a rain event , lead ^{214}Pb and bismuth ^{214}Bi are mainly removed from the atmosphere by rainout (in-cloud scavenging process)</u>



Data obtained from: "Wet deposition of ^{222}Rn progeny in Northern Finland measured with an automatic precipitations gamma analyser" J. Paatero, 2000

Experimental site and setup

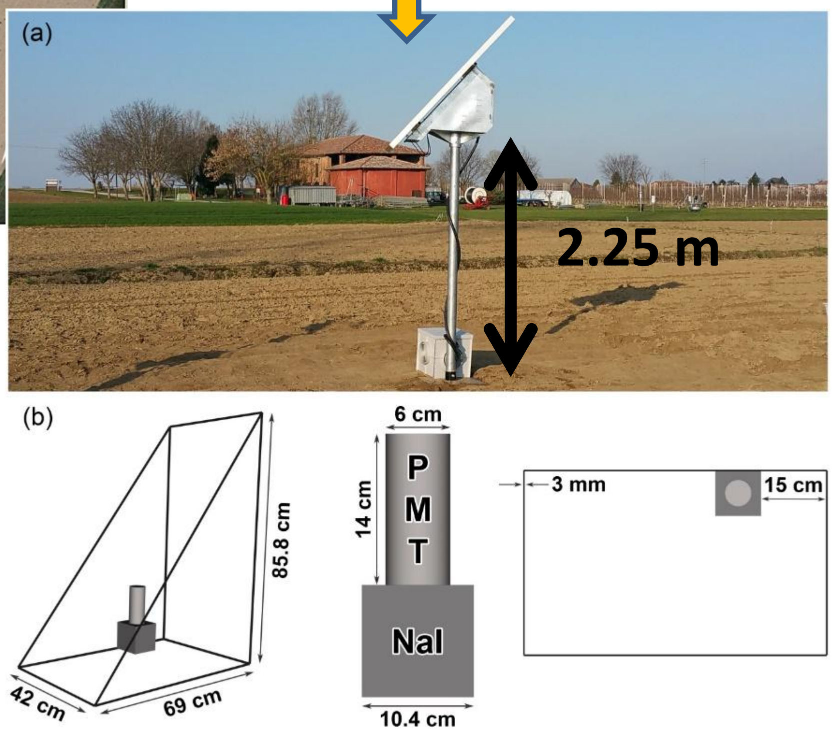


Experimental test field of the Acqua Campus of CER (Emilia-Romagna, Italy)

Period of data acquisition :

4th April – 2nd November 2017

Gamma station (γ) **Agro-metereological station (w)**

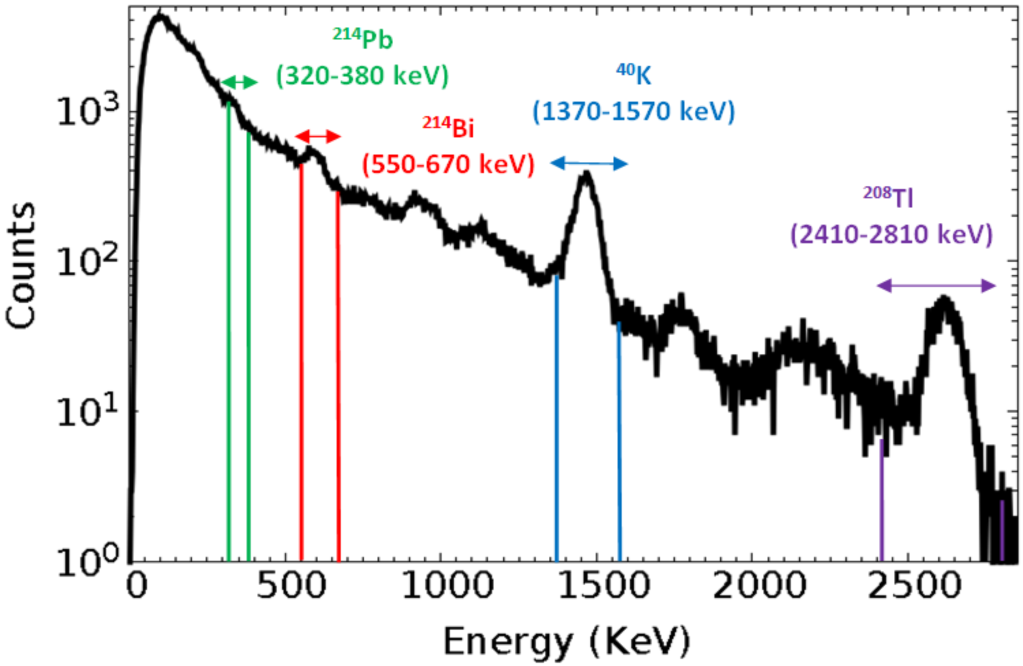


Single time-referenced dataset with a temporal resolution of 15 min

Apparent activity of ^{214}Bi (cps)

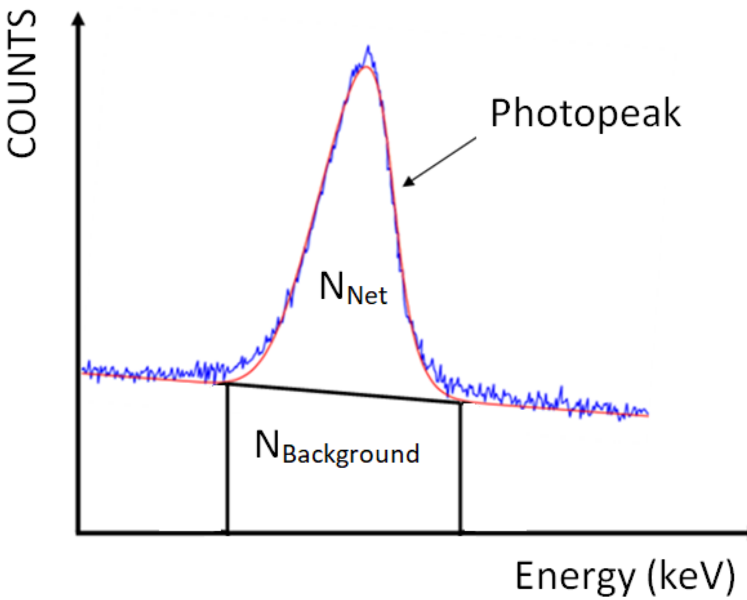
Rainfall rate (mm/s)

Apparent gamma activity of ^{214}Bi



The gamma-emitting daughters of ^{238}U and ^{232}Th and ^{40}K gamma decays are the main contributors to the environmental gamma spectrum

Gamma events of interest in the range 550-670 KeV (^{214}Bi photopeak)



Apparent activity of ^{214}Bi : count rate n_{rate}, i.e. counts per second (cps) recorded by the NaI(TI) detector

$$n_{rate} = \frac{N_{net}}{T} = \frac{N_{total}}{T} - \frac{N_{background}}{T}$$

Acquisition time T = 900 s (15 min)

Period of analysis of the rain-induced gamma activity of ^{214}Bi on the ground

$^{214}Pb\ (t_{1/2} = 26.7\ \text{min}) \xrightarrow{\beta,\ \gamma} {}^{214}Bi\ (t_{1/2} = 19.9\ \text{min})$

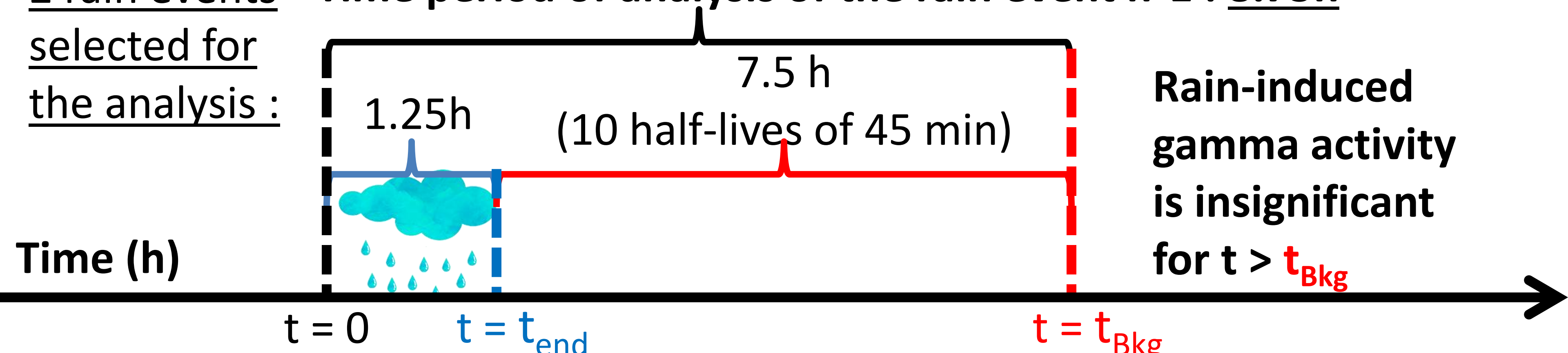
^{214}Pb and ^{214}Bi have similar half-lives

In rainwater, ^{214}Pb and ^{214}Bi are present together

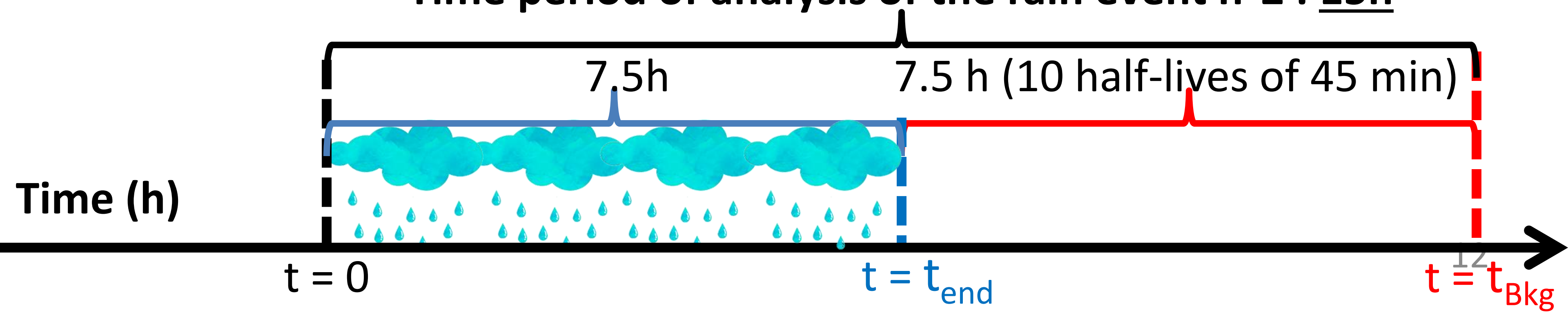
→ **In rainwater, the half-life of system ^{214}Pb - ^{214}Bi is <u>45 min</u>**

<u>2 rain events selected for the analysis :</u>

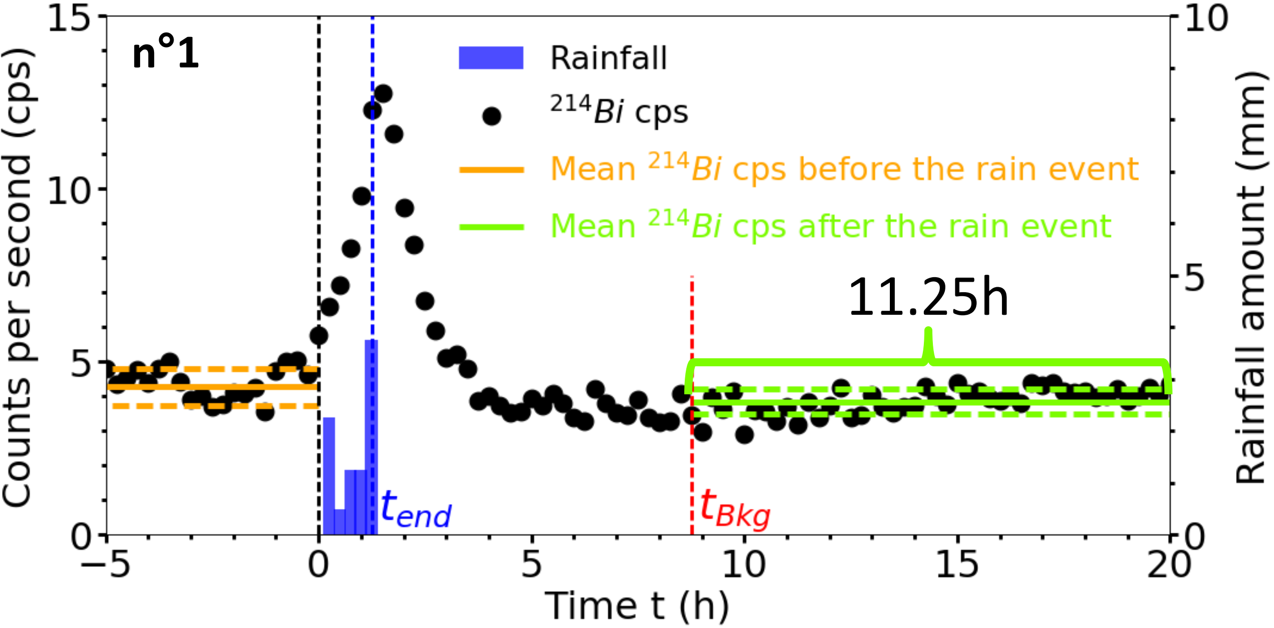
Time period of analysis of the rain event n°1 : <u>8.75h</u>

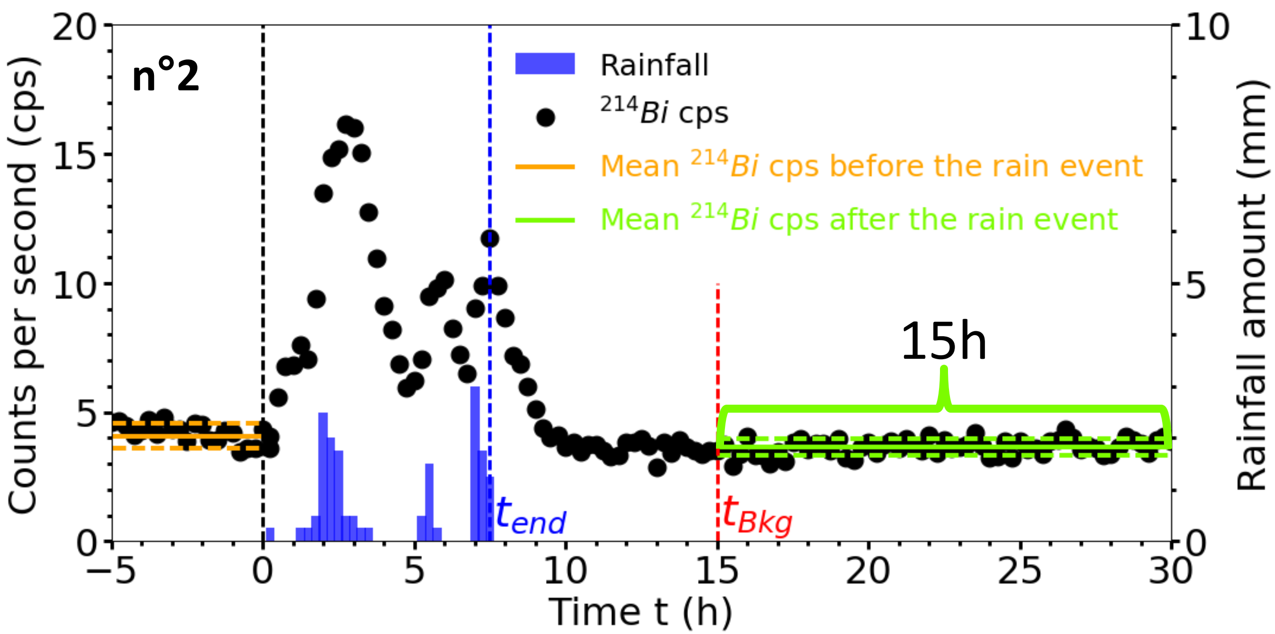


Time period of analysis of the rain event n°2 : <u>15h</u>



Data acquired during the rain events



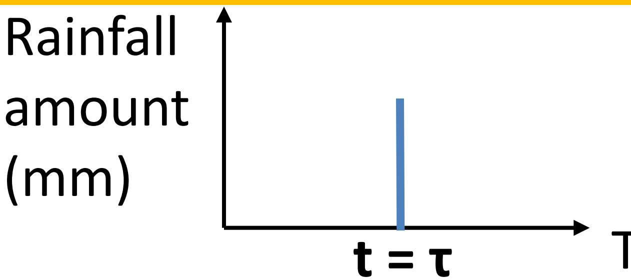


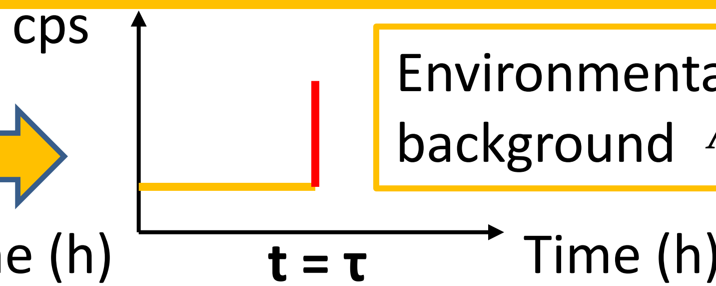
Rain events	Mean ^{214}Bi cps	Mean ^{214}Bi cps
n°1	4.28 ± 0.54	3.85 ± 0.37
n°2	4.08 ± 0.47	3.65 ± 0.30

Mean ^{214}Bi cps before the rain events has been evaluated over a period of 20 h

Rainfall bins = 'Dirac delta' rain impulses recorded every 15 min by the rain gauge during each rain event

Expected cps due to a single 'Dirac delta' rain impulse at **t = τ**





Environmental background A_{Bkg}

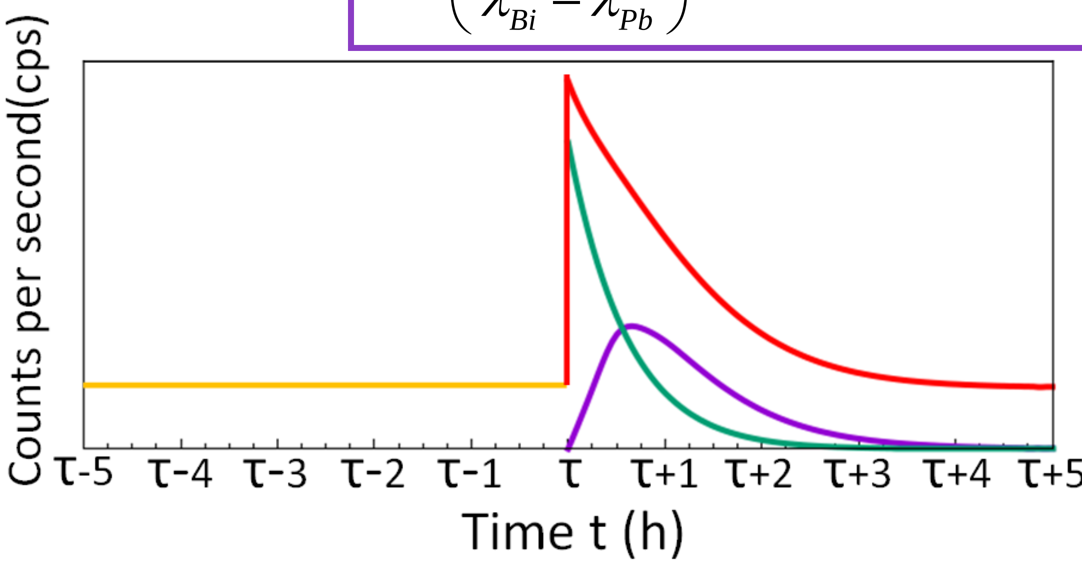
Expected cps $A_{Bi}^{\tau}(t=\tau)$

How evolve the cps for **t ≥ τ ?**

Since $^{214}Pb \xrightarrow{\beta,\gamma} {}^{214}Bi$ ⇒ $A_{Bi}^{\tau}(t)$ has a source term and decay term

$$A_{Bi}^{\tau}(t) = A_{Rain}(t) + A_{Bkg} = A_{Rain}^{source}(t) + A_{Rain}^{decay}(t) + A_{Bkg}$$

$$= A_{Pb}^{\tau} \cdot \left(\frac{\lambda_{Bi}}{\lambda_{Bi} - \lambda_{Pb}} \right) \cdot \left(e^{-\lambda_{Pb}(t-\tau)} - e^{-\lambda_{Bi}(t-\tau)} \right) + A_{Bi}^{\tau} \cdot e^{-\lambda_{Bi}(t-\tau)} + A_{Bkg}$$



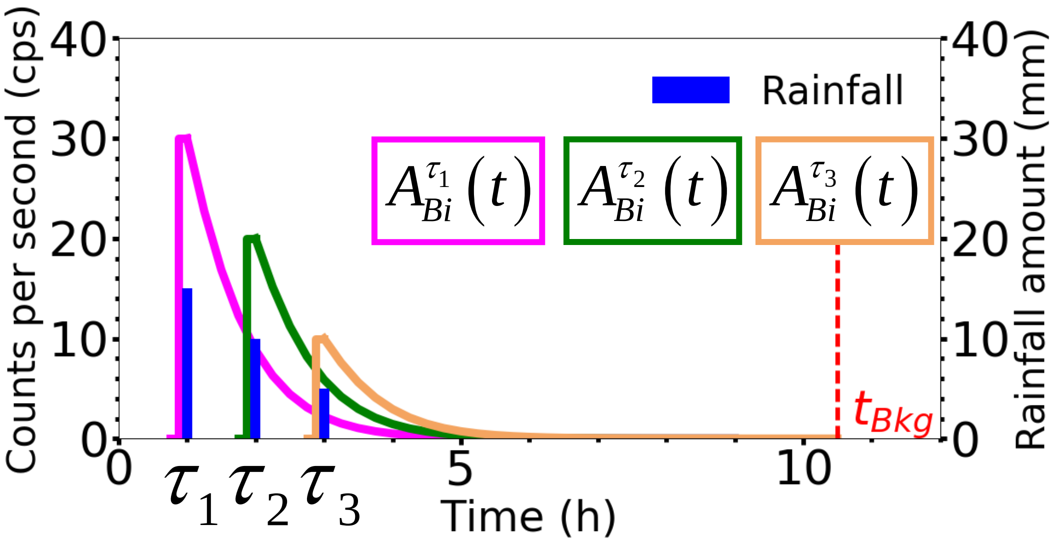
$A_{Bi}^{\tau}(t)$: expected cps due to an impulse of rain at **t = τ**

λ_{Pb} , λ_{Bi}: decay constants (s^{-1}) of ^{214}Pb and ^{214}Bi respectively

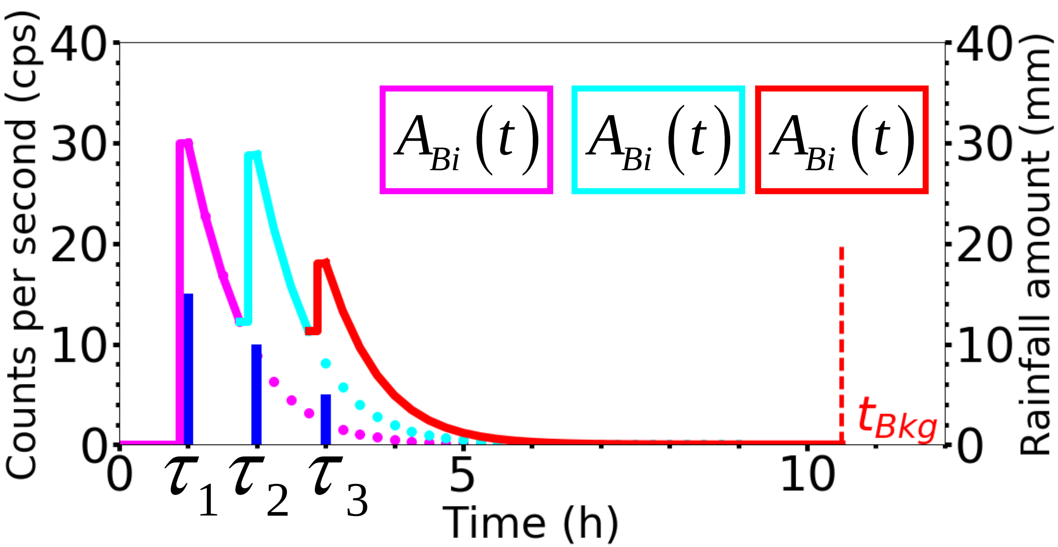
A_{Pb}^{τ}, A_{Bi}^{τ} : activities of ^{214}Pb and ^{214}Bi at **t = τ**

Expected cps due to multiple 'Dirac delta' rain impulses

Let's consider a rain event composed by multiple 'Dirac delta' rain impulses:



Expected cps due to each individual 'Dirac delta' rain impulse



Total expected cps $A_{Bi}(t)$:

For $\tau_1 \leq t < \tau_2$

$$A_{Bi}(t) = A_{Bi}^{\tau_1}(t)$$

For $\tau_2 \leq t < \tau_3$

$$A_{Bi}(t) = A_{Bi}^{\tau_1}(t) + A_{Bi}^{\tau_2}(t)$$

For $t \geq \tau_3$

$$A_{Bi}(t) = A_{Bi}^{\tau_1}(t) + A_{Bi}^{\tau_2}(t) + A_{Bi}^{\tau_3}(t)$$

Mathematical formalism to analyze the cps due to multiple 'Dirac delta' rain impulses

How to analyze the cps by considering an environmental parameter measured on the field during a rain event ?

Considering:

$G_{Pb}(\tau)$ $G_{Bi}(\tau)$: activities of ^{214}Pb and ^{214}Bi respectively per mm of rainwater (**cps mm^{-1}**) deposited on the ground at **t = τ**

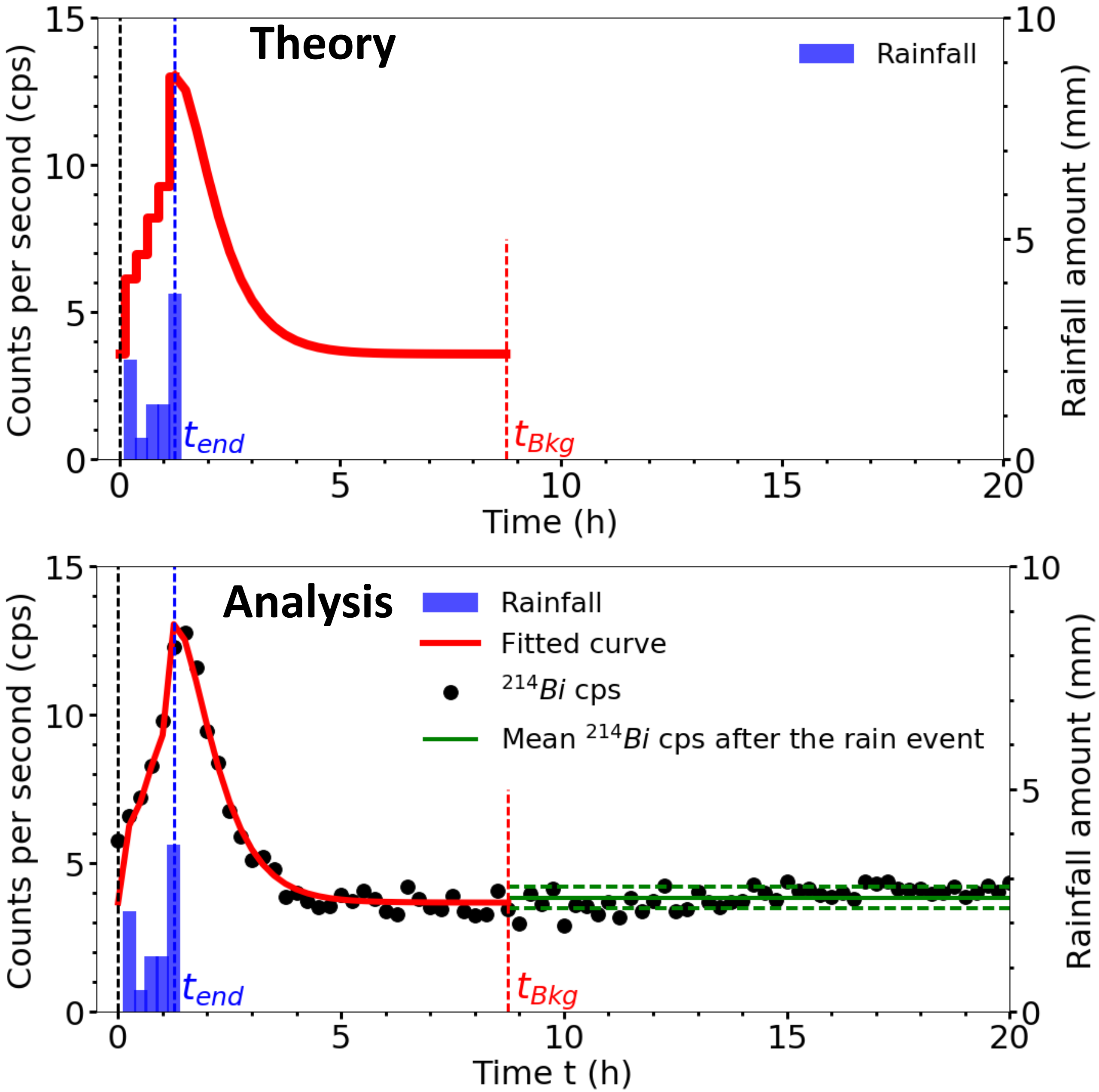
$f(\tau)$: rainfall rate (**mm s^{-1}**) recorded at **t = τ**

Expected count rate due to a multiple 'Dirac delta' rain impulses:

$$A_{Bi}(t) = \int_0^t f(\tau) \cdot \left[G_{Pb}(\tau) \cdot \left(\frac{\lambda_{Bi}}{\lambda_{Bi} - \lambda_{Pb}} \right) \cdot \left(e^{-\lambda_{Pb}(t-\tau)} - e^{-\lambda_{Bi}(t-\tau)}\right) + G_{Bi}(\tau) \cdot e^{-\lambda_{Bi}(t-\tau)} \right] \cdot d\tau + A_{Bkg}$$

For simplicity G_{Pb} and G_{Bi} are assumed to be constant during a rain event

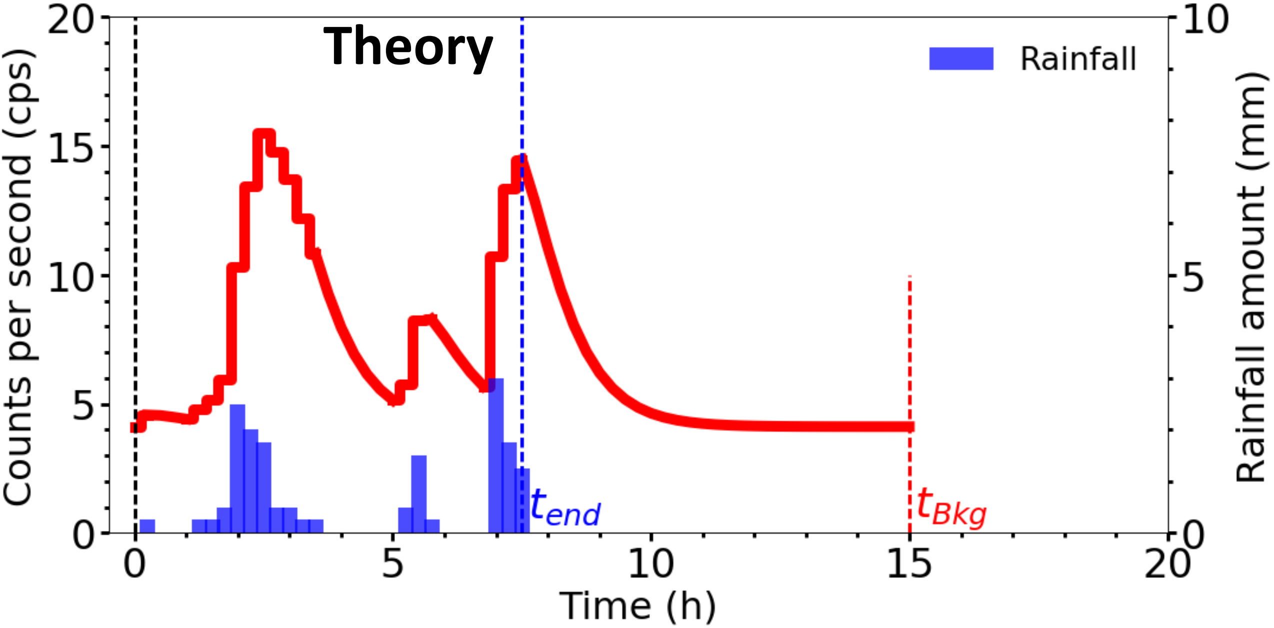
Analysis of the rain event n°1

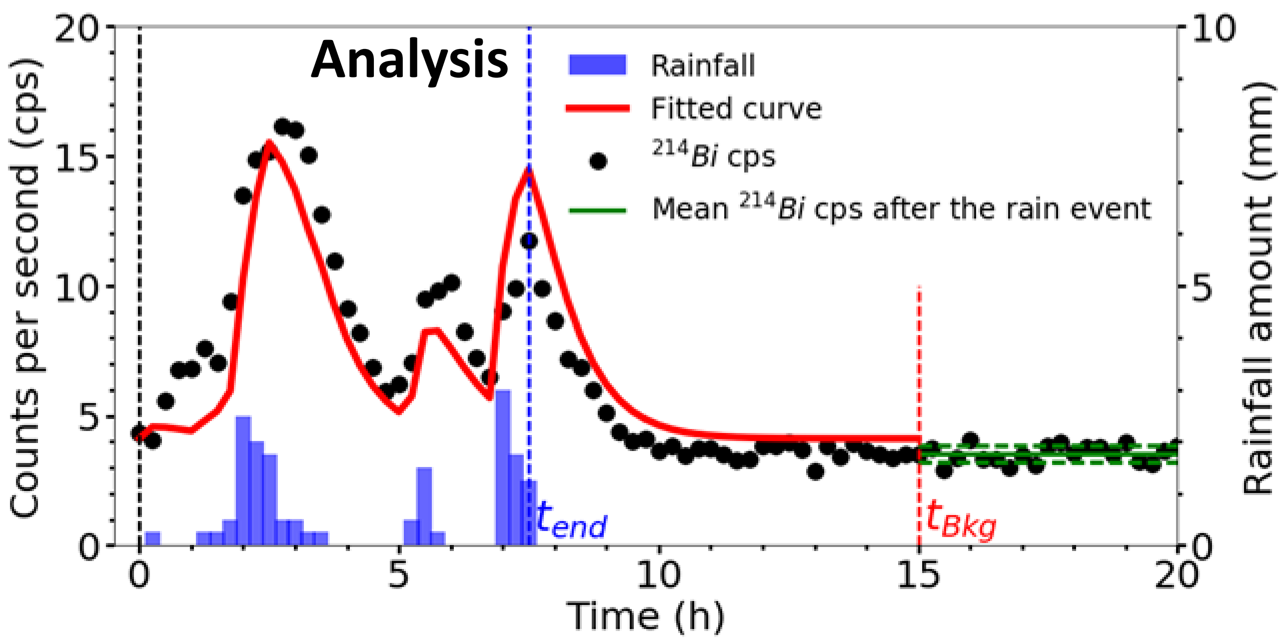


Chi-squared goodness of fit test

Fit parameters	Values
G_{Pb} (cps mm^{-1})	1.75 ± 0.08
G_{Bi} (cps mm^{-1})	1.15 ± 0.06
A_{Bkg} (cps)	3.58 ± 0.07
χ^2 / ndf	3.24

Analysis of the rain event n°2





Chi-squared goodness of fit test

Fit parameters	Values
G_{Pb} (cps mm^{-1})	2.00 ± 0.31
G_{Bi} (cps mm^{-1})	1.82 ± 0.25
A_{Bkg} (cps)	4.12 ± 0.29
χ^2 / ndf	64.80

Summary of the analyzed rain events

	Rain events				
Date	11^{th} July	6^{th} August	24^{th} September (**rain event n°1**)	18^{th} April (**rain event n°2**)	18^{th} September
Rain event duration (s)	3600	6300	4500	27000	75600
Rainfall rate (mm s^{-1})	63.89×10^{-4}	58.57×10^{-4}	20.00×10^{-4}	4.07×10^{-4}	2.38×10^{-4}
G_{Pb} (cps mm^{-1})	1.12 ± 0.03	1.64 ± 0.05	1.75 ± 0.08	2.00 ± 0.31	4.02 ± 0.38
G_{Bi} (cps mm^{-1})	0.71 ± 0.03	1.00 ± 0.04	1.15 ± 0.07	1.82 ± 0.25	1.45 ± 0.34
A_{Bkg} (cps)	3.48 ± 0.08	3.35 ± 0.09	3.58 ± 0.07	4.12 ± 0.24	4.10 ± 0.19
χ^2 / ndf	5.45	6.11	3.24	64.80	65.34

- G_{Pb} and G_{Bi} , activities of ^{214}Pb and ^{214}Bi per mm of rainwater respectively, and the rainfall rate are inversely correlated according to the analyzed events.
- The environmental background tend to be greater during the events with low rainfall rates than the ones with high rainfall rates.
- The uncertainties of the fit parameters obtained from the long events are greater than ones obtained from the short events.

Conclusion

The atmospheric radon is mainly originated from one meter below the soil surface due to its short half-life and the soil moisture content.

Since the atmospheric radon daughters ^{214}Pb and ^{214}Bi are mainly attached to sub-micron and micron aerosol particles, their scavenging from the atmosphere during rain events is mainly due to the rainout process (in-cloud scavenging process).

No enhancement of the rain-induced ^{214}Bi activity on the ground is due to a difference of ^{222}Rn concentrations at ground level.

The variation of the rain-induced ^{214}Bi activity during and after a short rain event is well described by a mathematical model based on the assumption that the variation in time of the activity of ^{214}Bi is due to the variation of the rainfall rate in time.

The observed discrepancies between the mathematical model and the recorded ^{214}Bi activity may be due to the variability of the activities of ^{214}Pb and ^{214}Bi per mm of rainwater deposited on the ground during long rain events.

Thank you for your attention !

Perspectives

Based on the mathematical formalism studied in the framework of this thesis…

- the variability of the activities of ^{214}Pb and ^{214}Bi per mm of rainwater during a long rain event can be investigated by assuming that a long rain event is a sum of multiple shorts rain events.

- large datasets obtained by performing measurements at different sites can be analyzed in order to understand the variability of the activities of ^{214}Pb and ^{214}Bi per mm of rainwater due to the environmental parameters such as the wind direction, air temperature and season

- This would lead to many interesting applications such as the calculation of the rainfall rate of a rain event from the variation of the rain-induced activity measured on the ground.