Wet deposition of radon decay products

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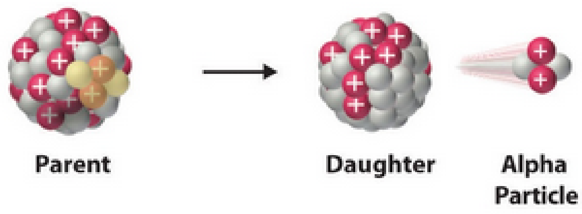
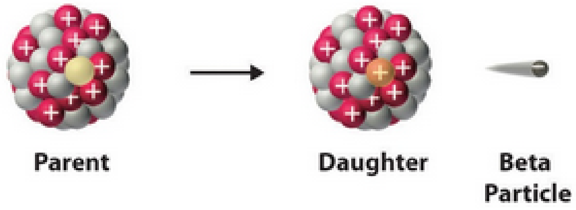
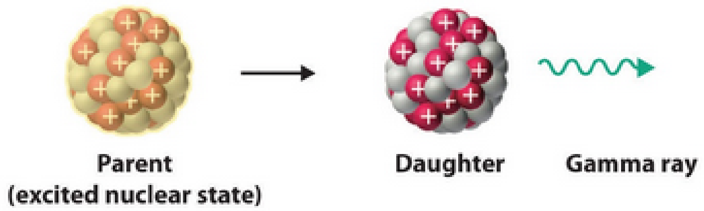
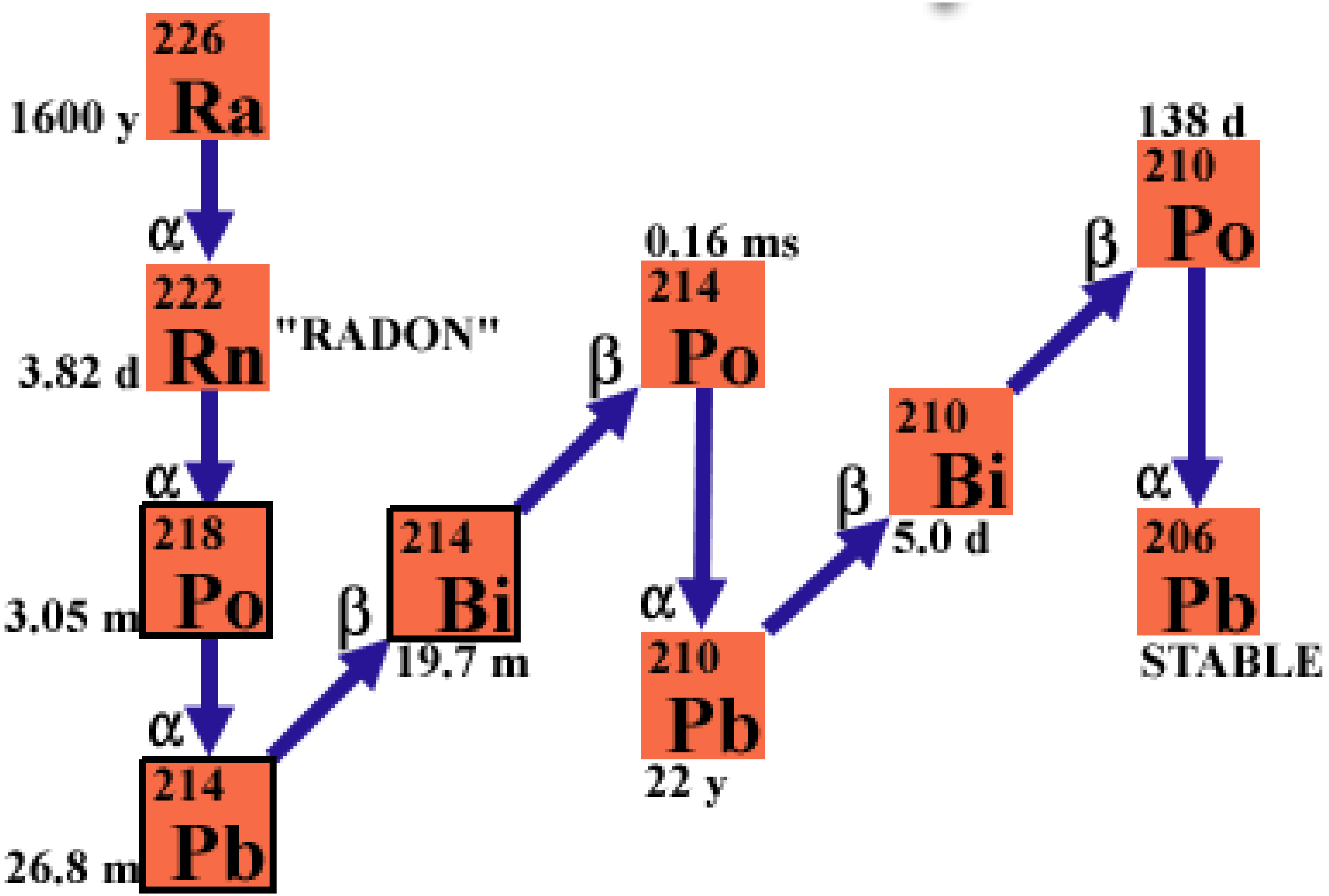
2021.03.18.

Radioactive decay

An unstable nucleus loses energy by radiation.
The material containing unstable nuclei is *radioactive*.

Decay Type	Radiation Emitted	Generic Equation	Model
Alpha decay	${}^{4}_{2}\alpha$	${}^{A}_{Z}X \longrightarrow {}^{A-4}_{Z-2}X' + {}^{4}_{2}\alpha$	
Beta decay	${}^{0}_{-1}\beta$	${}^{A}_{Z}X \longrightarrow {}^{A}_{Z+1}X' + {}^{0}_{-1}\beta$	
Gamma emission	${}^{0}_{0}\gamma$	${}^{A}_{Z}X^{*} \xrightarrow{\text{Relaxation}} {}^{A}_{Z}X' + {}^{0}_{0}\gamma$	

Decay diagram of radon



Natural radioactive sources

- Terrestrial radiation
- Cosmic radiation
- Internal radiation

How can the ^{226}Ra (decays to ^{222}Rn) get into the soil?

1.) The soil forming rock had 238U atoms in it, which decayed to ^{226}Ra, so the ^{226}Ra was “born” into the soil.

2.) During soil formation ^{226}Ra atoms got into the crystal structures.

3.) The ^{226}Ra precipitates on the surface of soil particles due to chemical reactions.

Natural radioactive sources

- Terrestrial radiation
- Cosmic radiation
- Internal radiation

How can the ^{226}Ra (decays to ^{222}Rn) get out of the soil?

1.) and 2.) : since radon is a noble gas it does not stick into the crystal structure because of interactions and can easily get to the intergranular space = **"emanation"**

(the radons decayed during diffusion can not get out to the intergranular space)

3.) was already in the intergranular space

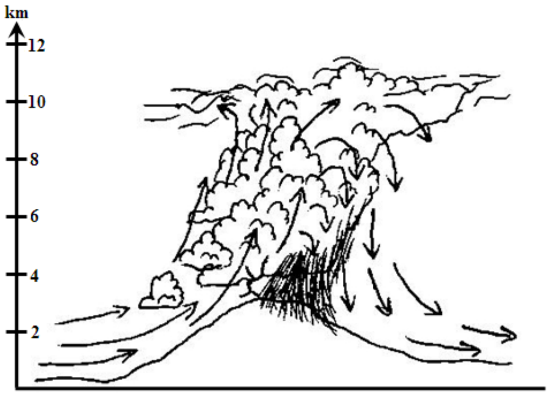
From the intergranular space radon can get to the atmosphere = **"exhalation"**

The troposphere

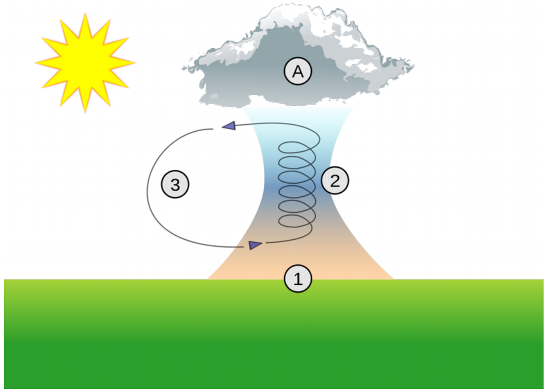
- Lowest layer of Earth's atmosphere
- Nearly all the weather conditions take place in it
- Contains 75% of the atmosphere's mass
- 99% of the total mass is water vapour and aerosols
- Its average hight above Hungary: 11 km (winter: 8-10 km, summer: 12-14 km)
- Significant horizontal and vertical convection
- Clouds are formed here

Vertical convections in the troposphere

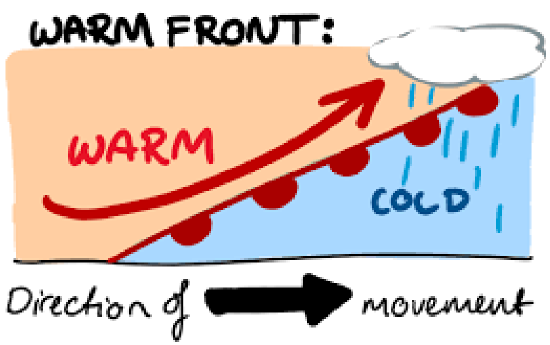
1.) Orographic effect:
because of a geographical obstacle (e.g. mountain)



2.) Termic convection:
different temperatures (and therefore different pressures) move the layers



3.) Fronts:
two air masses meet with different temperatures meet and slip on each other



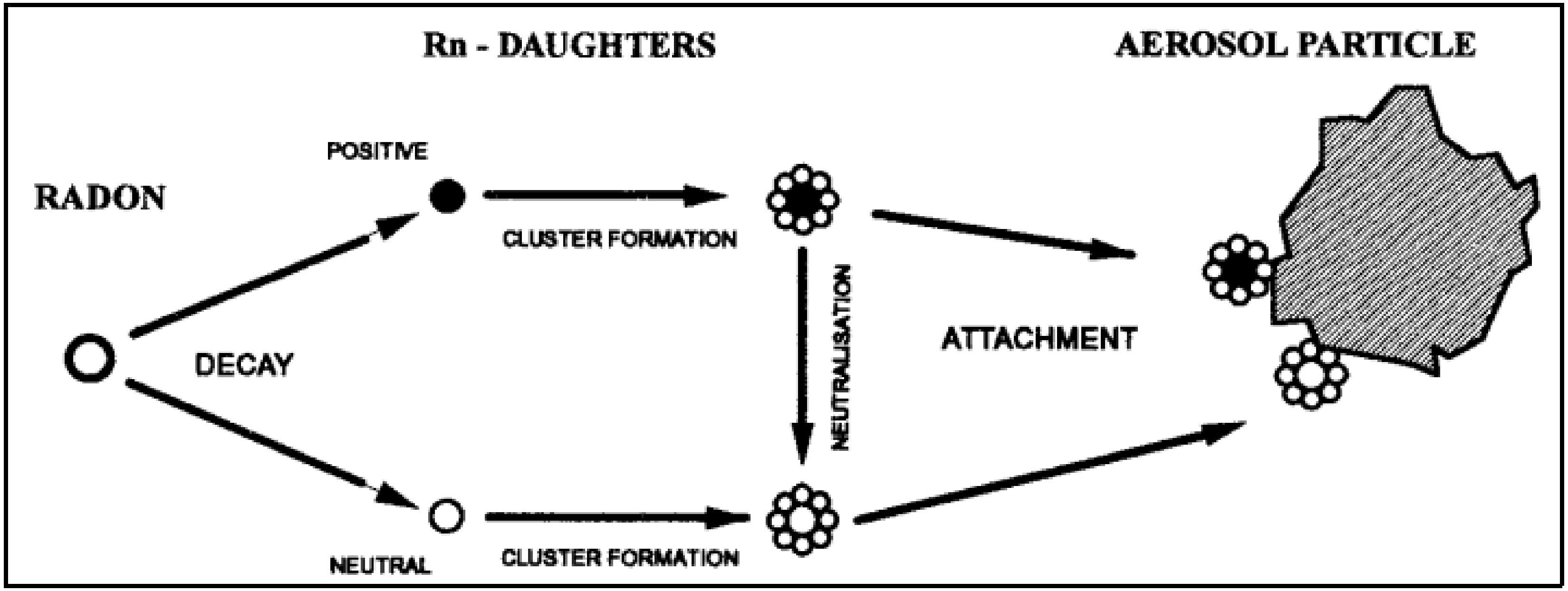
- as the upward going air cools it can contain less and less humidity
- when the relative humidity of the air reaches 100% it becomes supersaturated
- the extra humidity must precipitate and therefore we need condensation nuclei!

Remark: condensation can occur below 100% relative humidity depending on the aerosols size and material

Radon decay in the atmosphere

Radon has a relatively short half life (3.82 days)
→ a lot of its daughters will be present in the troposphere

The daughters are in solid state
→ they can attach to the aerosol particles



reaction time: < 1s

diameters: 0.5-5 nm

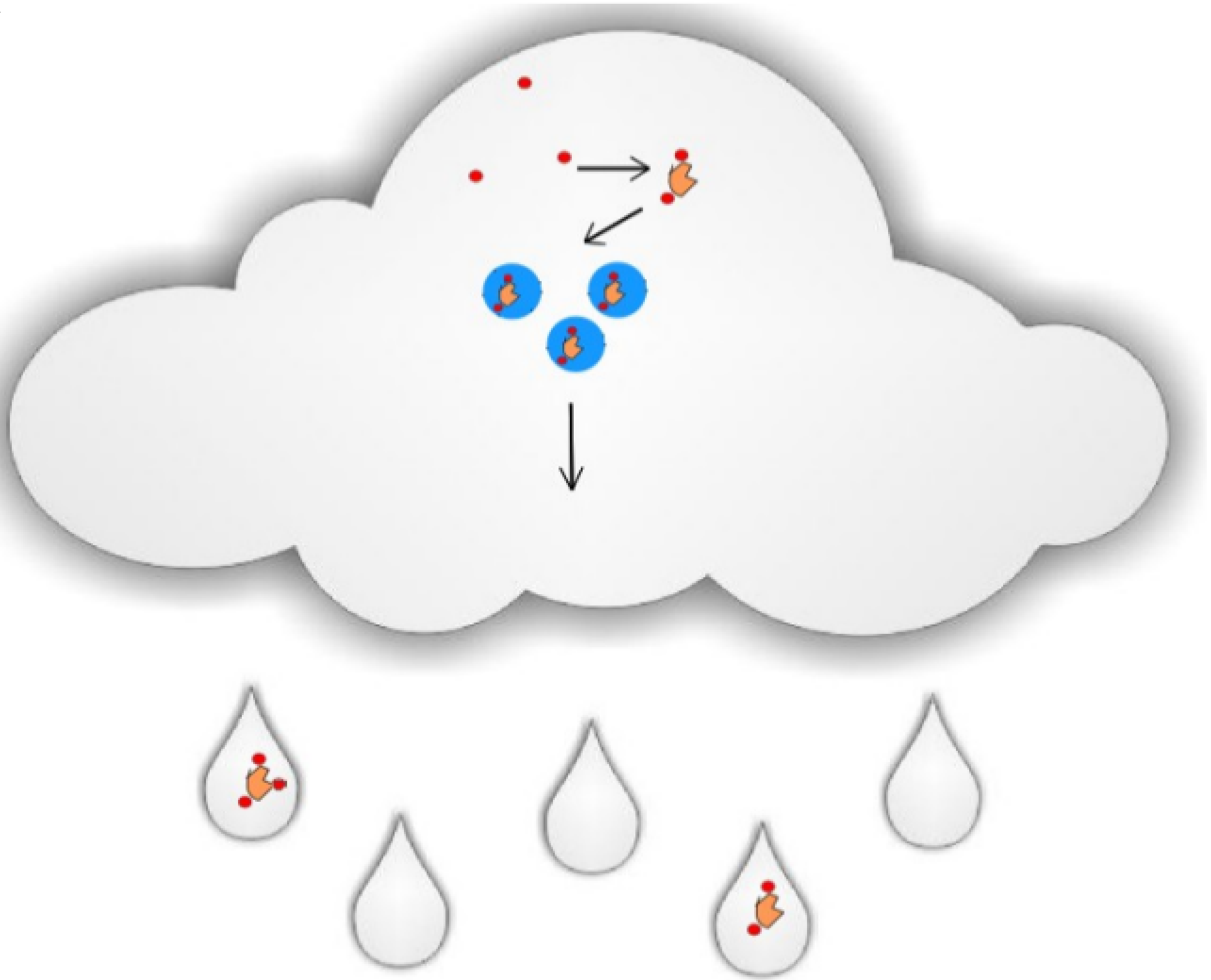
diameters: 0.015-0.5 µm

In-cloud scavenging

In order to condense into a rain droplet, water vapor requires the initial presence of an aerosol particle
(otherwise the small cluster of water molecules will spontaneously evaporate).

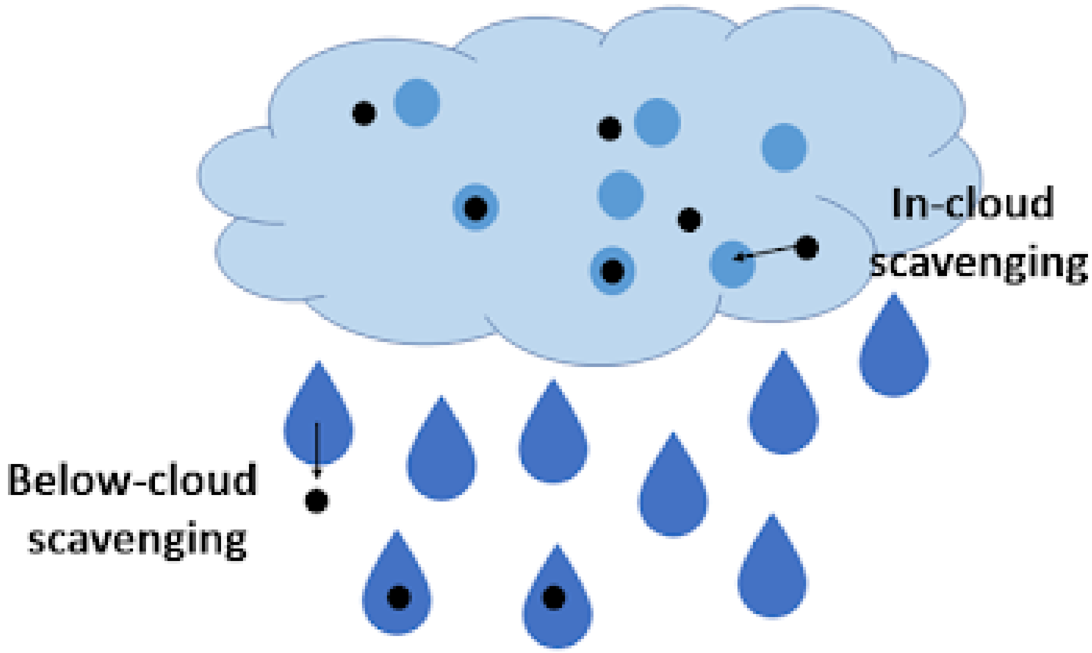
The formed cloud droplets contain the radioactive nuclei.

The cloud drops will grow until they are large enough (100μm) to overcome the updraft speed within the cloud and fall as rain.



Below-cloud scavenging

The second way of increasing the radioactivity of a raindrop:



- The falling raindrop can collide with an aerosol particle containing radioactive nuclei.
- But it rarely happens because of the different sizes: the aerosol particles try to avoid the bigger raindrops along aerodynamic flow lines.
- The below-cloud scavenging has a much less significant role in the increase of the raindrop radioactivity.

Summary of an article

Paatero, J. and Hatakka, J. ,
Wet deposition efficiency of short-lived radon-222 progeny in central Finland, Boreal Env. Res. 4: 285–293 (1999)

- The ^{214}Po concentration increases as the size of the raindrops decreases (the size of a raindrop does not change the radioactive nuclei content)
- Experimental evidence that the in-cloud scavenging in way more significant than the below-cloud scavenging
- The ^{214}Po amount correlates with the amount of precipitation
- The air's ^{222}Rn concentration has a seasonal fluctuation (e.g. in winter the snow blocks most of the radon exhalation)
- The ^{214}Po content of the precipitation does not have seasonal fluctuations (→ it's value must be determined by other meteorological processes)
- The precipitation's and the air's 222Rn and 214Po concentration depends on the wind direction

Model for wet deposition of radon progeny

Minato, S. , A Simple Rainout Model for Radon Daughters, J. Nucl. Radiochem. Sci., Vol. 8, No.1, pp. N1-N3 (2007)

Calculates radioactivities of short-lived radon daughters in rainwater as a function of rainfall rate for a given unit concentration of radon in the cloud air.

The model incorporates as semi-empirical formulas

- Collection for cloud droplets by raindrops
- Removal of raindrops from cloud
- Transport time of raindrops from the cloud base to the ground

Assumptions:

≈ ^{222}Rn and its daughters are uniformly distributed
≈ Equilibrium is established between radon and its progeny
≈ Cloud droplets are formed immediately and simultaneously
≈ Atmospheric conditions (e.g. temperature) changes suddenly
≈ Cloud droplets capture only ^{218}Po
≈ Activity ratio: RaA (^{218}Po) : RaB (^{214}Po) : RaC (^{214}Bi) = 1 : 0 : 0

Change in the number of radon daughters

$$\underset{\text{}}{\frac{dn_A}{dt}} = \overset{\text{I.}}{-\lambda_A n_A} \overset{\text{II.}}{- \Psi_C n_A} \overset{\text{III.}}{+ a_{Rn}}$$

I. the decay of the A isotope (^{218}Po)
II. the droplet becomes a raindrop with Ψ probability
III. Radon activity produces ^{218}Po

equilibrium
Ψ same for all

$$a_{Rn} = \lambda_A n_A + \psi_C n_A$$
$$\lambda_A n_A = \lambda_B n_B + \psi_C n_B$$
$$\lambda_B n_B = \lambda_C n_C + \psi_C n_C$$

$$a_A = \lambda_A n_A = \frac{\lambda_A}{\lambda_A + \psi_C} a_{Rn}$$
$$a_B = \lambda_B n_B = \frac{\lambda_B}{\lambda_B + \psi_C} a_A$$
$$a_C = \lambda_C n_C = \frac{\lambda_C}{\lambda_C + \psi_C} a_B$$

a: radioactivity per unit volume [Bq/m3]
n: number of cloud droplets containing X
λ: decay constants [1/s]

Change in the number of radon daughters

$$a_A = \lambda_A n_A = \frac{\lambda_A}{\lambda_A + \psi_C} a_{Rn}$$

$$a_B = \lambda_B n_B = \frac{\lambda_B}{\lambda_B + \psi_C} a_A$$

$$a_C = \lambda_C n_C = \frac{\lambda_C}{\lambda_C + \psi_C} a_B$$

Considering the number of raindrops: N

removal rate

$$\psi_C n_A = \lambda_A N_A + \psi_r N_A$$

$$\psi_C n_B + \lambda_A N_A = \lambda_B N_B + \psi_r N_B$$

$$\psi_C n_C + \lambda_B N_B = \lambda_C N_C + \psi_r N_C$$

Activity of raindrops per unit volume

$$A_A = \lambda_A N_A = \frac{\psi_C}{\lambda_A + \psi_r} a_A$$

$$A_B = \lambda_B N_B = \frac{\psi_C}{\lambda_B + \psi_r} a_B + \frac{\lambda_B}{\lambda_B + \psi_r} A_A$$

$$A_C = \lambda_C N_C = \frac{\psi_C}{\lambda_C + \psi_r} a_C + \frac{\lambda_C}{\lambda_C + \psi_r} A_B$$

Radon daughters under The cloud base

$$\frac{dA_A}{dt} = -\lambda_A A_A$$

$$\frac{dA_B}{dt} = \lambda_B (A_A - A_B)$$

$$\frac{dA_C}{dt} = \lambda_C (A_B - A_C)$$

Change in the number of radon daughters

Gamma dosimeters → since ^{218}Po produce only α-radiation, only B and C isotopes contribute to the dose of interest

→ we assume the signals of B and C are equal

The final radioactivity observed on ground level:

$$A_B + A_C = A_{B0} e^{-\lambda_B t} + A_{A0} \frac{\lambda_B}{\lambda_B - \lambda_A} \left(e^{-\lambda_A t} - e^{-\lambda_B t} \right) + A_{C0} e^{-\lambda_C t} + A_{B0} \frac{\lambda_C}{\lambda_C - \lambda_B} \left(e^{-\lambda_B t} - e^{-\lambda_C t} \right)$$

$$+ A_{A0} \frac{\lambda_C \lambda_B \left[(\lambda_C - \lambda_B) e^{-\lambda_A t} - (\lambda_C - \lambda_A) e^{-\lambda_B t} + (\lambda_B - \lambda_A) e^{-\lambda_C t} \right]}{(\lambda_B - \lambda_A)(\lambda_C - \lambda_A)(\lambda_C - \lambda_B)}$$

Activity of rainwater from that:

$$\alpha_B + \alpha_C = \frac{\rho}{L_r} (A_B + A_C)$$

(Bq/m^3 to Bq/ml)

Change in the number of radon daughters

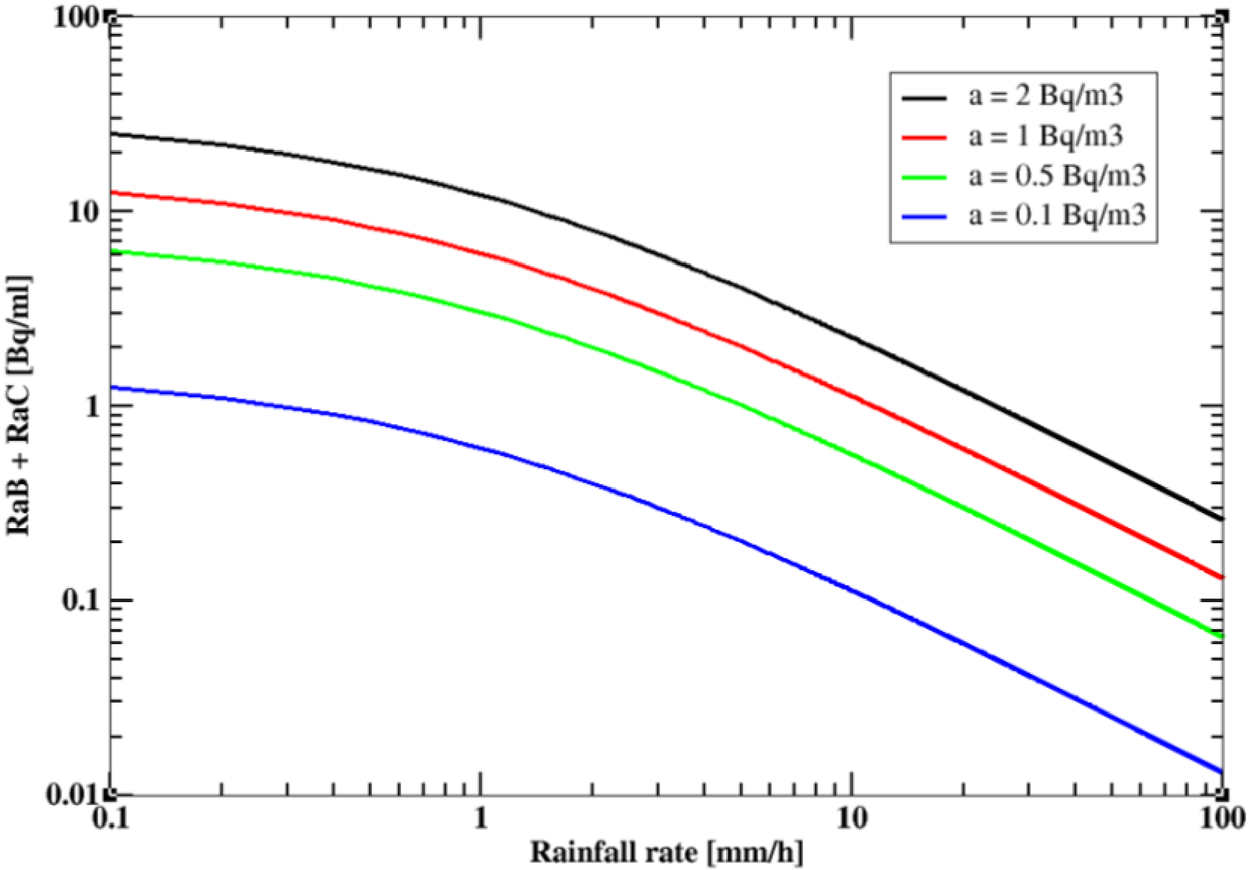
$$\alpha_B + \alpha_C = \frac{\rho}{L_r}\left(A_B + A_C\right)$$

L_r: water content of raindrops [g/m^3]
ρ: density of rainwater [g/mL]

The unknown values in this final equation can be calculated by using empirical rules considering...

- The rainfall rate
- The probability of the removal of one cloud droplet
- The mean radius of cloud droplet
- The depth of the cloud
- The average velocity of raindrops

Result of the model's final equation



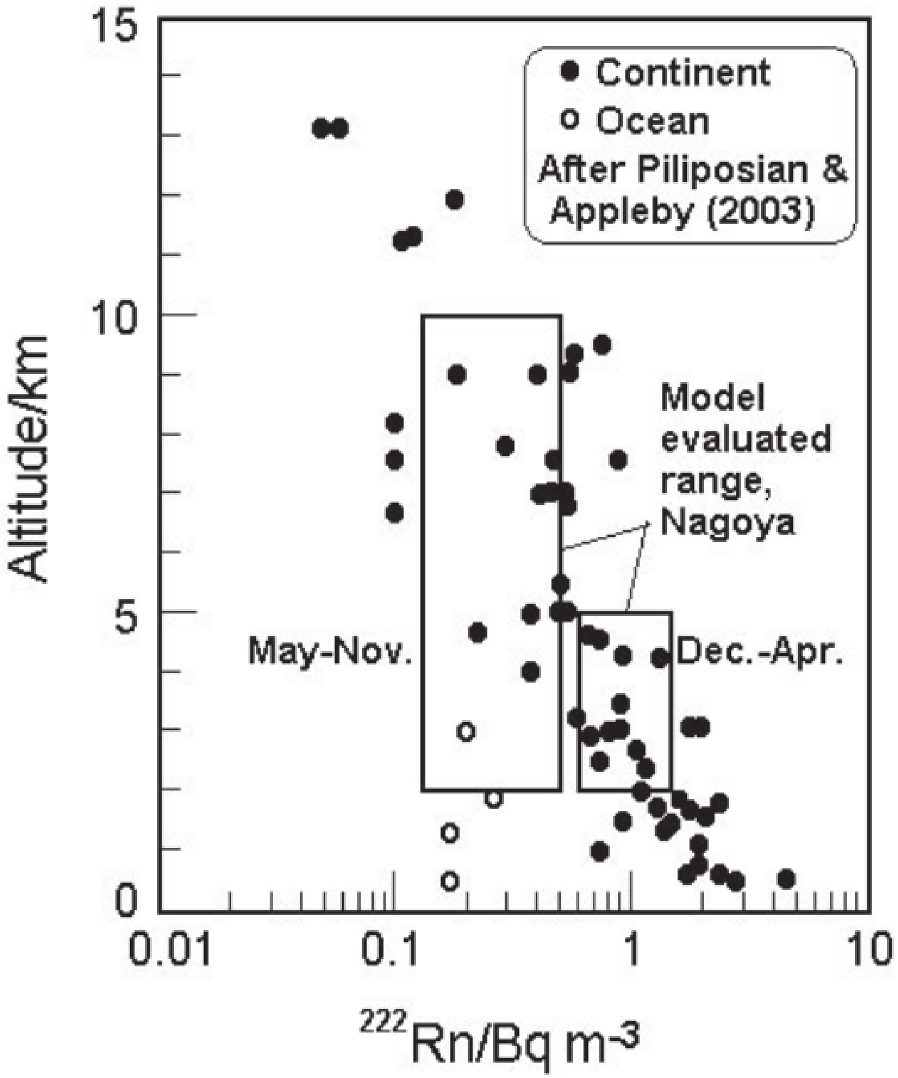
Different colors: given concentrations of radon in clouds

- The increase of rainfall rate does not cause an increase in the radioactivity at ground level
- Even the smallest amount of precipitation is capable of depositing most of the airborne progeny, further precipitation dilutes it
- Agrees with the previous studies that the below-cloud processes are less important then the in-cloud ones
- An increase of Rn activity in unit volume of cloud air produces a higher activity at ground level

Result of the model’s final equation

Using the model radon concentration data measured in cloud forming height in Nagoya was estimated
→ the results confirmed the model

Data measured by Piliposian and Appleby above North-America, Eurasia and Hawaii was estimated by the model
→ when the environmental circumstances were similar, the two datasets were comparable
→ between May and November the estimations were not that precise, meaning there must be other important factors which are not included in the model.



Data from the ELTE measuring station

Gamma dose rate detector

- 3 meters above ground
- Saves data in 10 minutes intervals
- The unit of measure is Sv/h
- Measuring range:
  10 nSv/h – 10 Sv/h
- Error: under 1 Sv/h: ±10%, above 1 Sv/h: ±15%

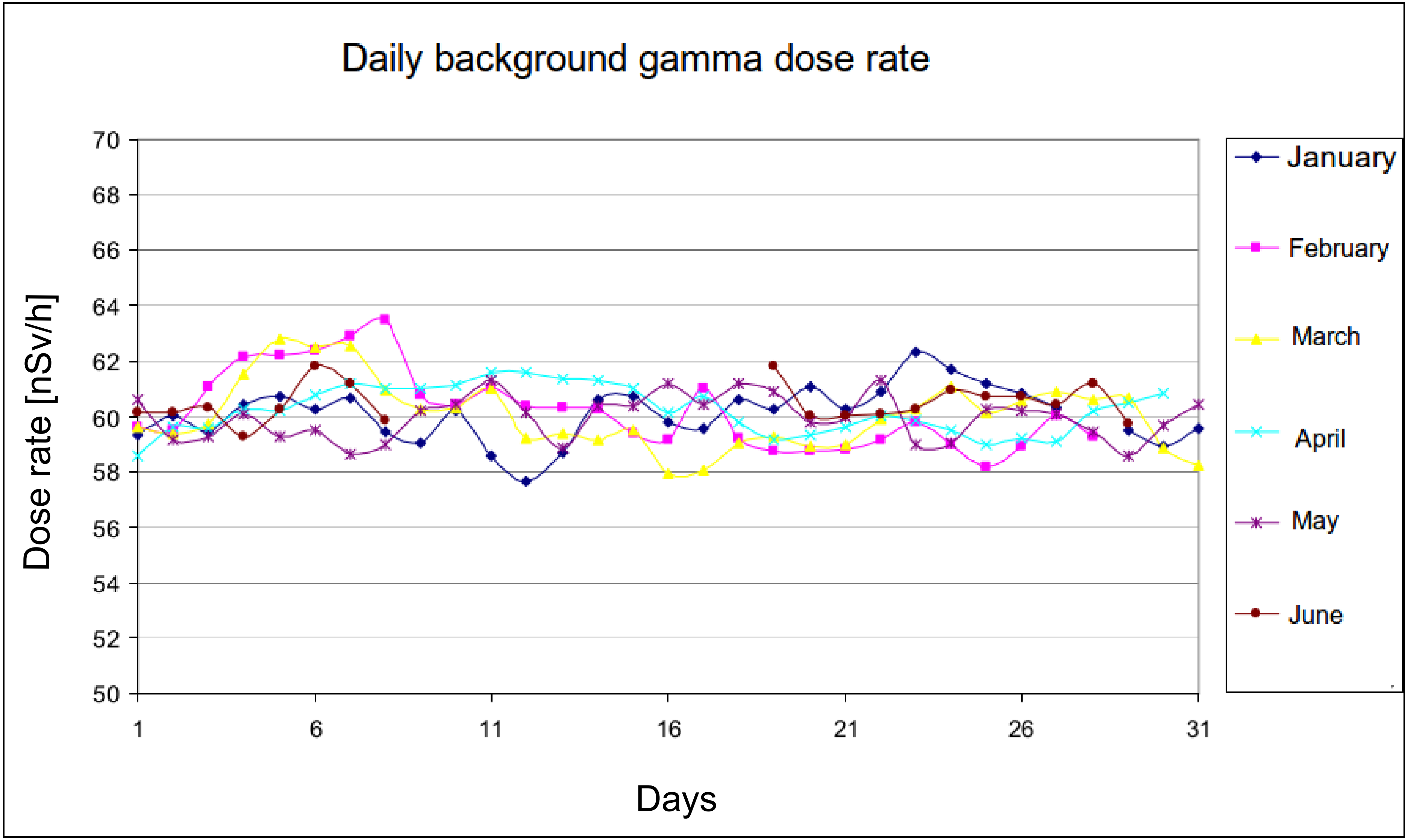
Meteorological measuring station

- Saves data in 10 minutes intervals
- Measures soil temperature, temperature, precipitation, global radiation, humidity, air pressure, wind
- We only use the precipitation

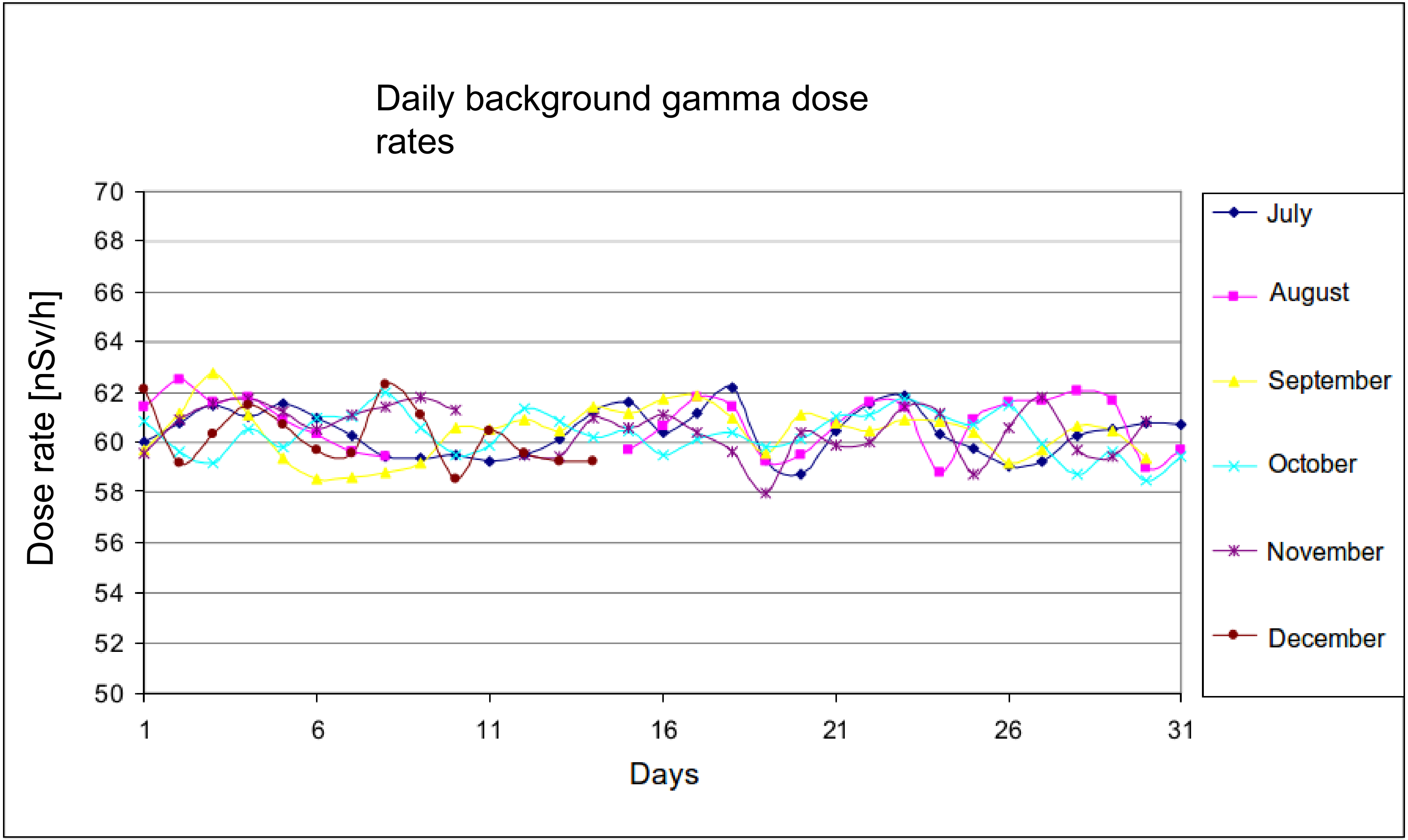
Data analysis and results

- Time of the measurements: 2009. 01. 01 – 2009. 12. 14.
- Average gamma dose rate: 60,7 nSv/h
- Highest value: 91 nSv/h
- Lowest value: 51,6 nSv/h
- Precipitation summed for the whole year: 360,5 mm
- Most intensive rain event: 14,6 mm in 10 minutes (June 28.)

Daily background gamma dose rates



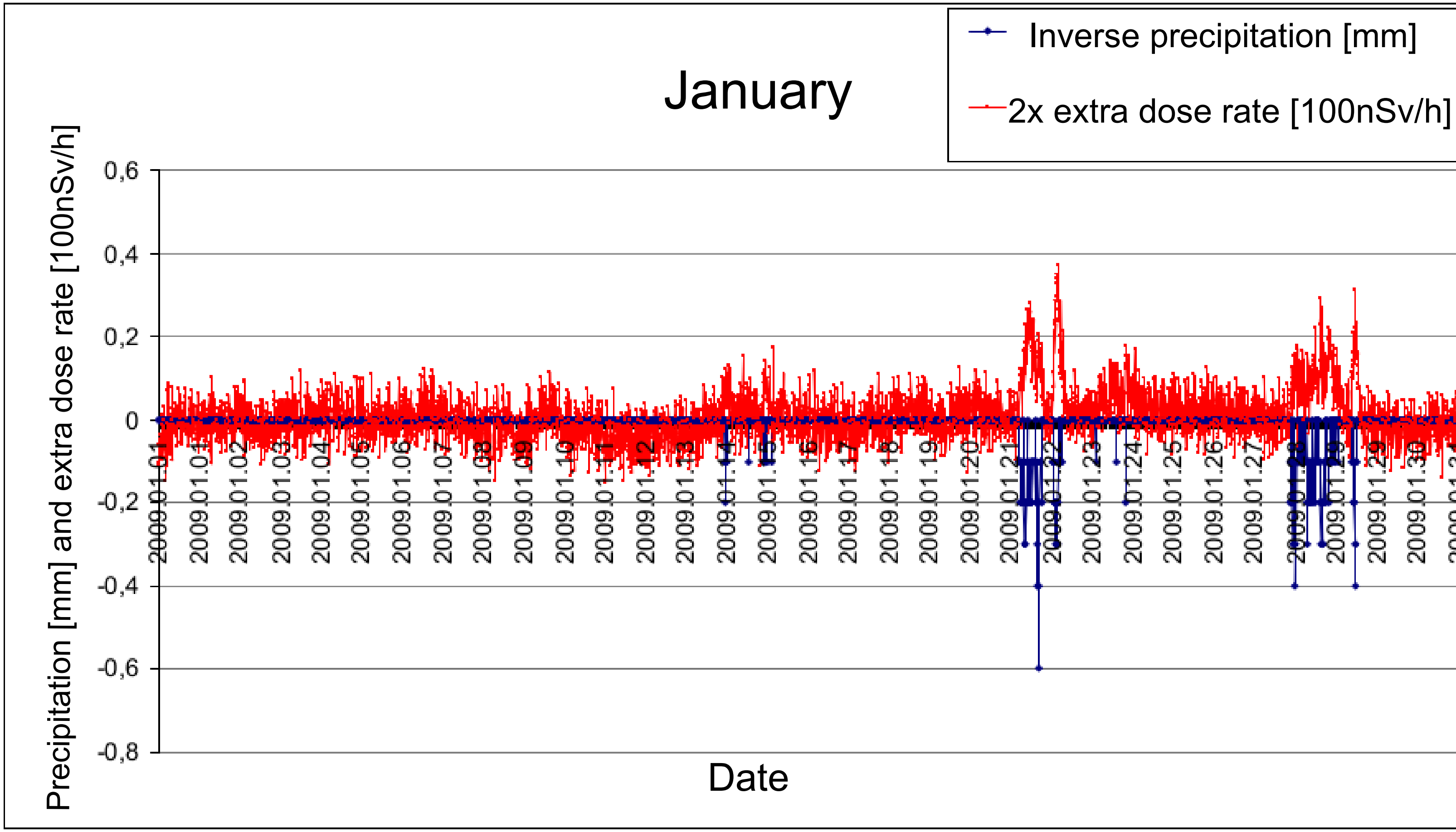
Daily background gamma dose rates



Monthly gamma dose rates

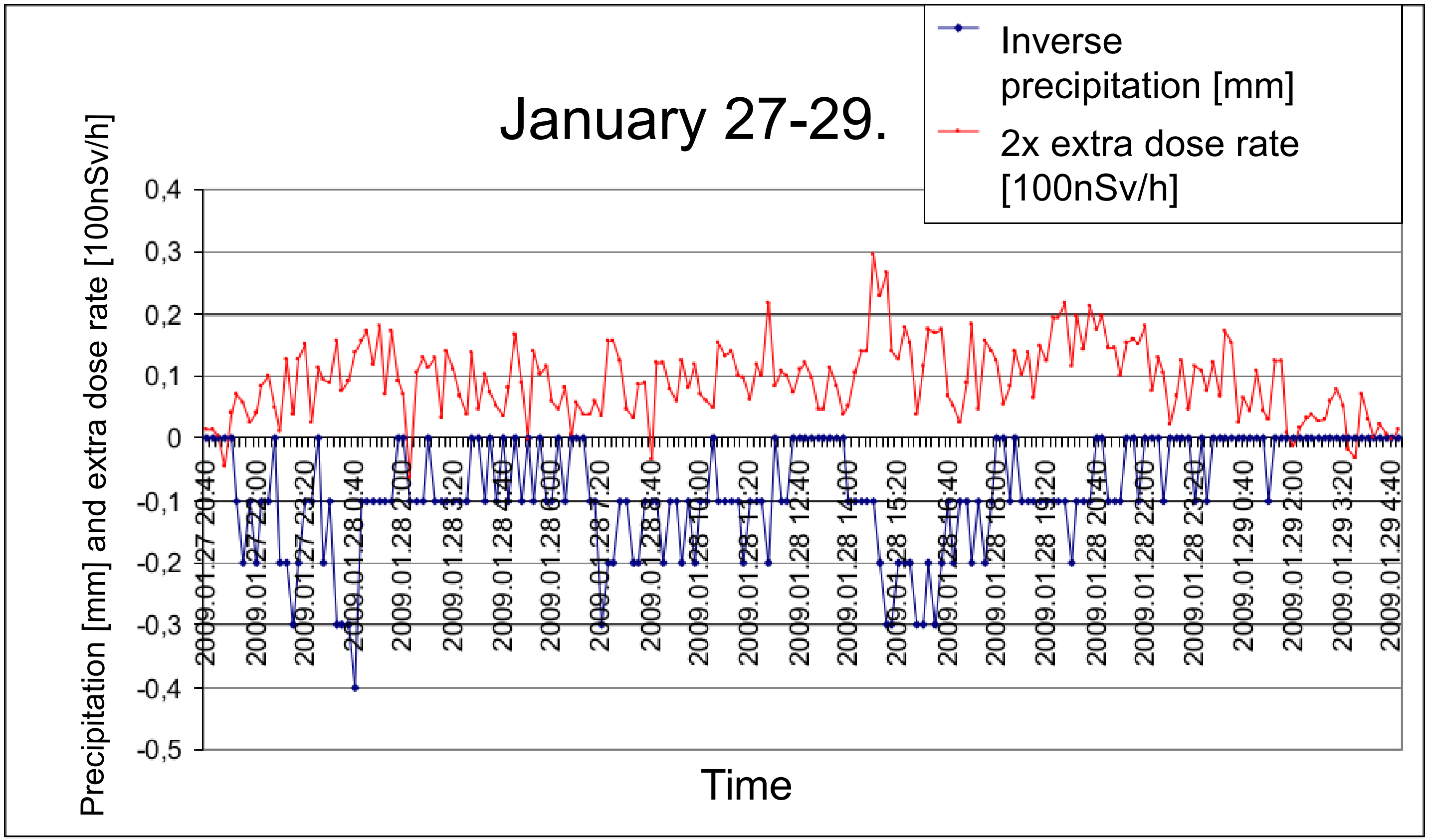
- Comparison of the precipitation and extra dose rate data
- The background gamma dose rate is subtracted
- The extra dose rate data is 2x multiplied for good visual comparison

Monthly gamma dose rates - January



Big precipitation event between January the 27th and 29th!

Biggest precipitation event in January

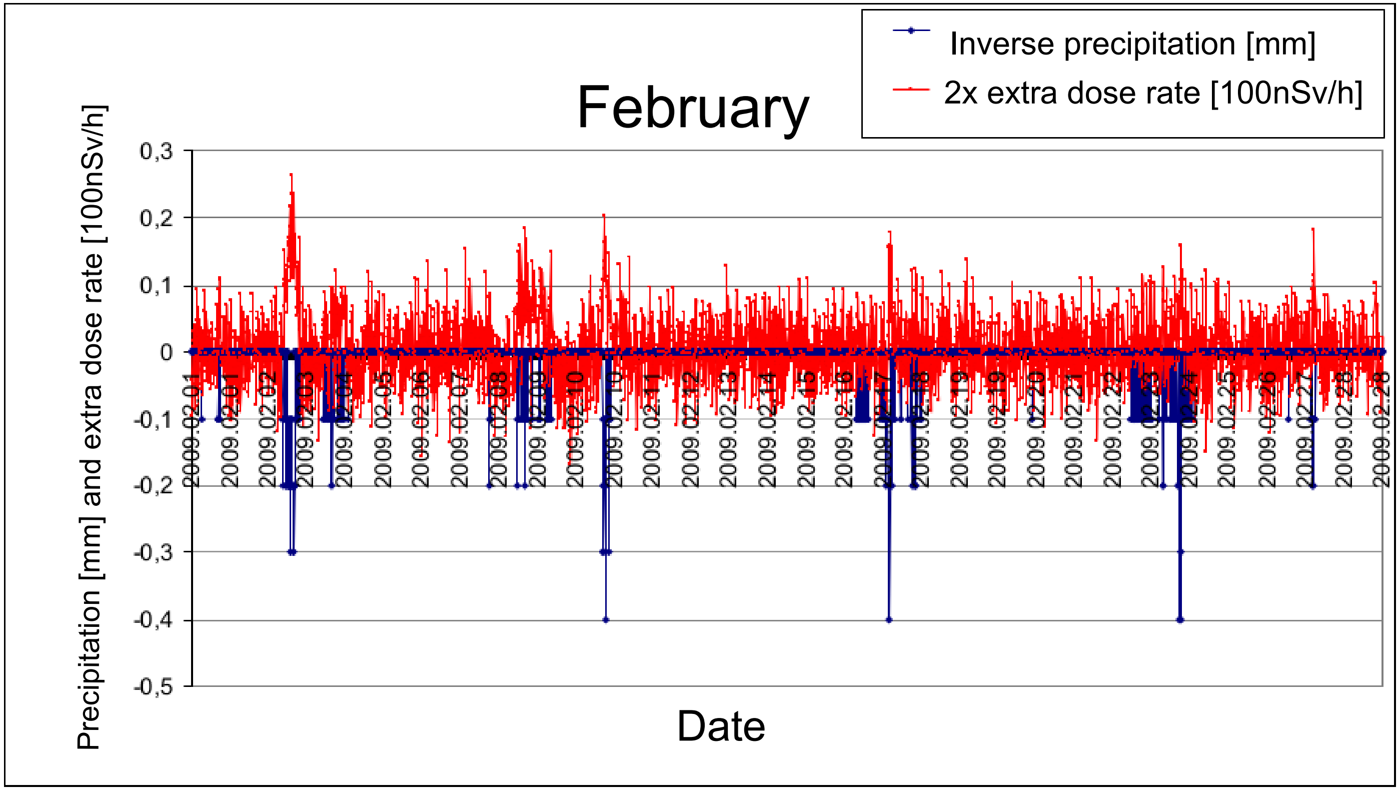


- 28 hours
- 16,5 mm precipitation (snow)
- Average: 65,4 nSv/h
- Maximum: 75 nSv/h
- 1,5 hours after snowing: 60 nSv/h
- Used background dose rate: from the previous and following day

From the rate of the decrease we see that it can depend on the amount of precipitation.

Monthly gamma dose rates - February

35,4 mm precipitation in total.

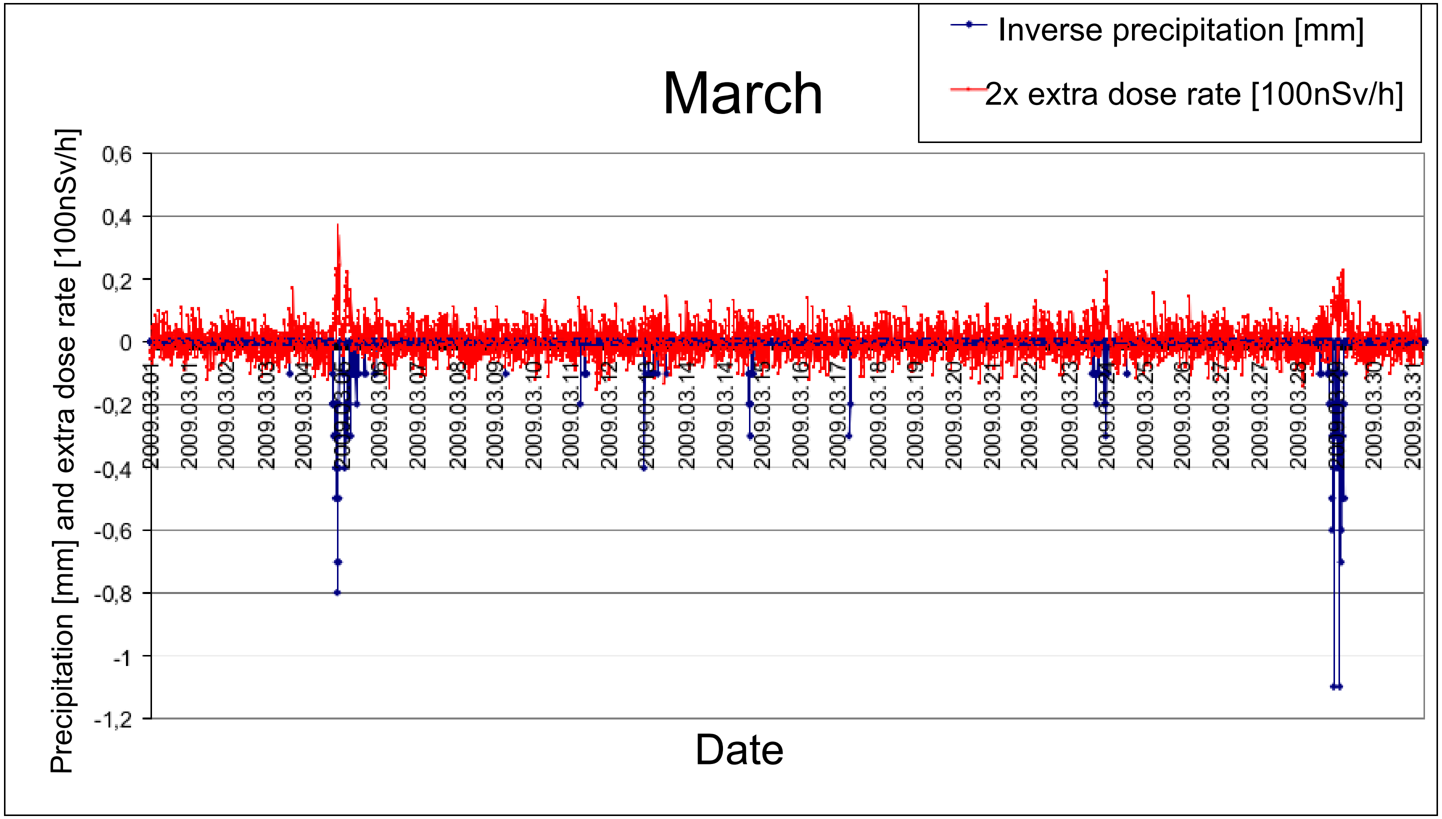


02.10.

- In 1,9 hours
- 2,6 mm precipitation
- Dose rate increase from 62,6 nSv/h to 71,2 nSv/h
- 3 hours after the rain: decreased to 61,3 nSv/h

From the rate of the decrease we see that it can depend on the amount of precipitation.

Monthly gamma dose rates - March

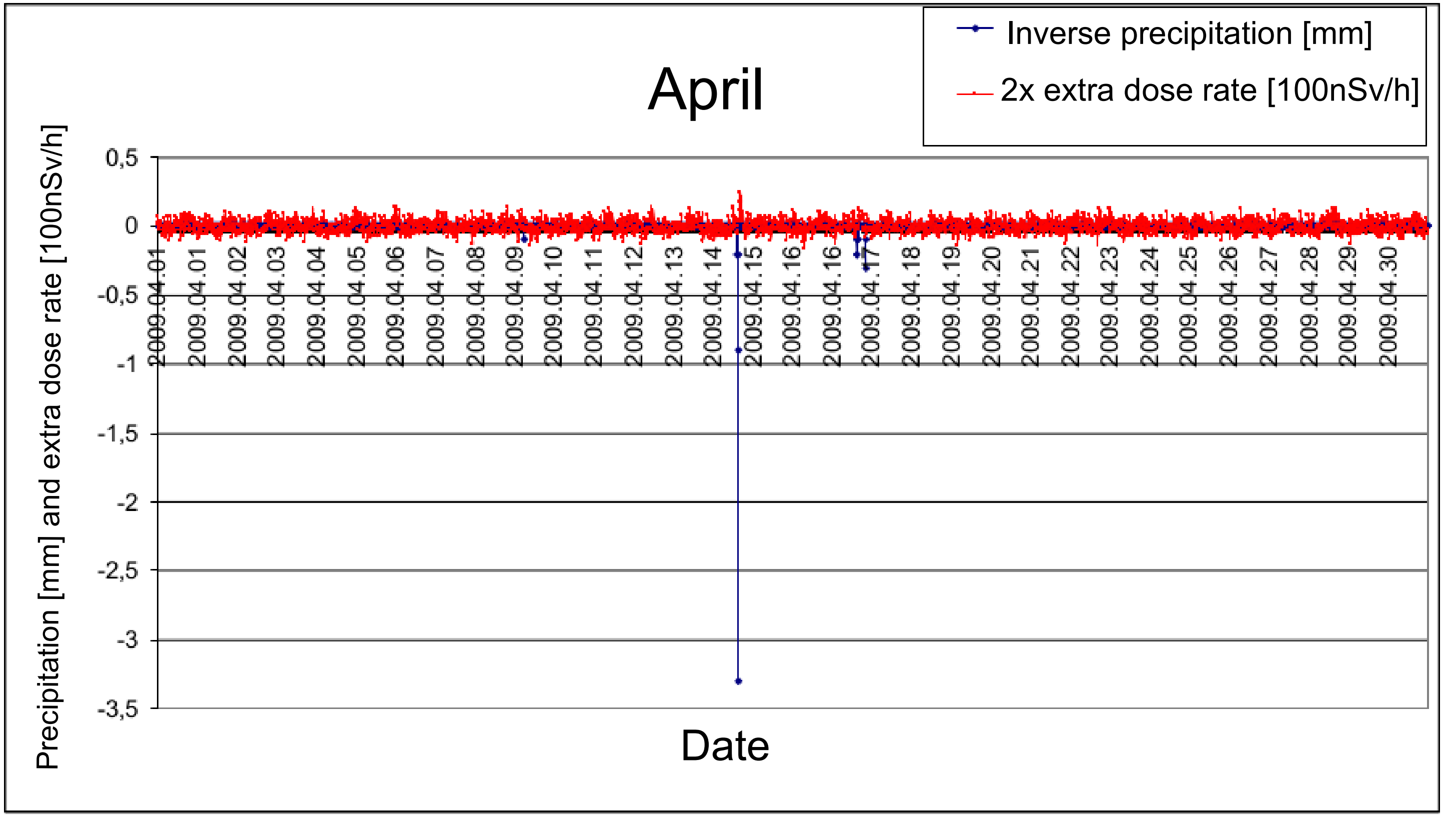


03.29.

- In 3 hours
- 6,7 mm precipitation
- Average dose rate increased by 8,62 nSv/h
- There was a 10 minutes measurement recorded 1,1mm
- In that 10 minutes 71,1 nSv/h was recorded (14 nSv/h increase!)

Monthly gamma dose rates - April

Really dry month, only 5,8mm produced by 3 precipitation events.

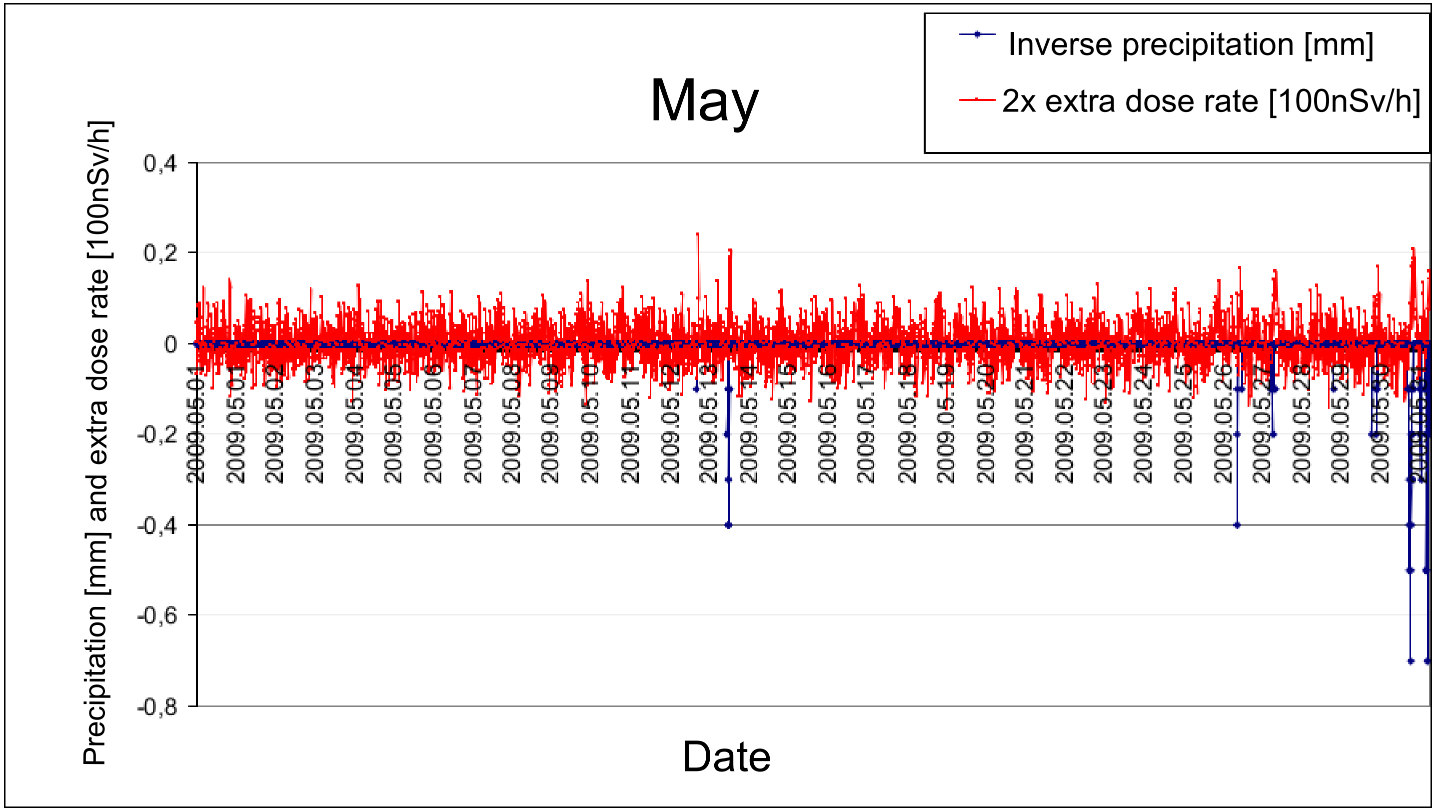


04. 14.

- In 30 minutes
- 4,4 mm precipitation
- Average dose rate increased only by 3 nSv/h

Monthly gamma dose rates - May

Dry month, only 13,4 mm.

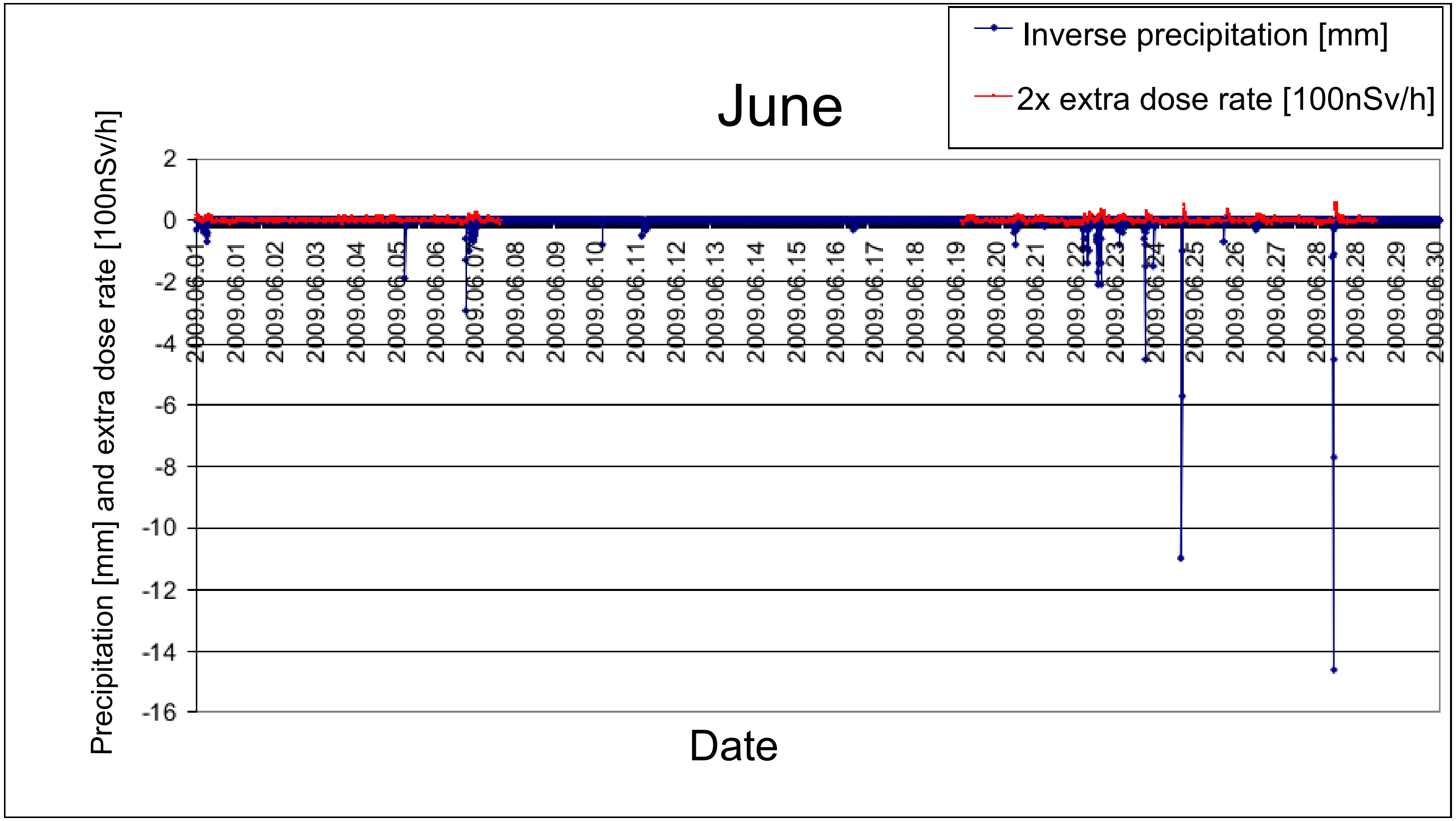


05. 31.

- Biggest precipitation event
- Didn't increase the dose rate dramatically

Monthly gamma dose rates - June

The detectors did not work between the 8th and 19th of June.

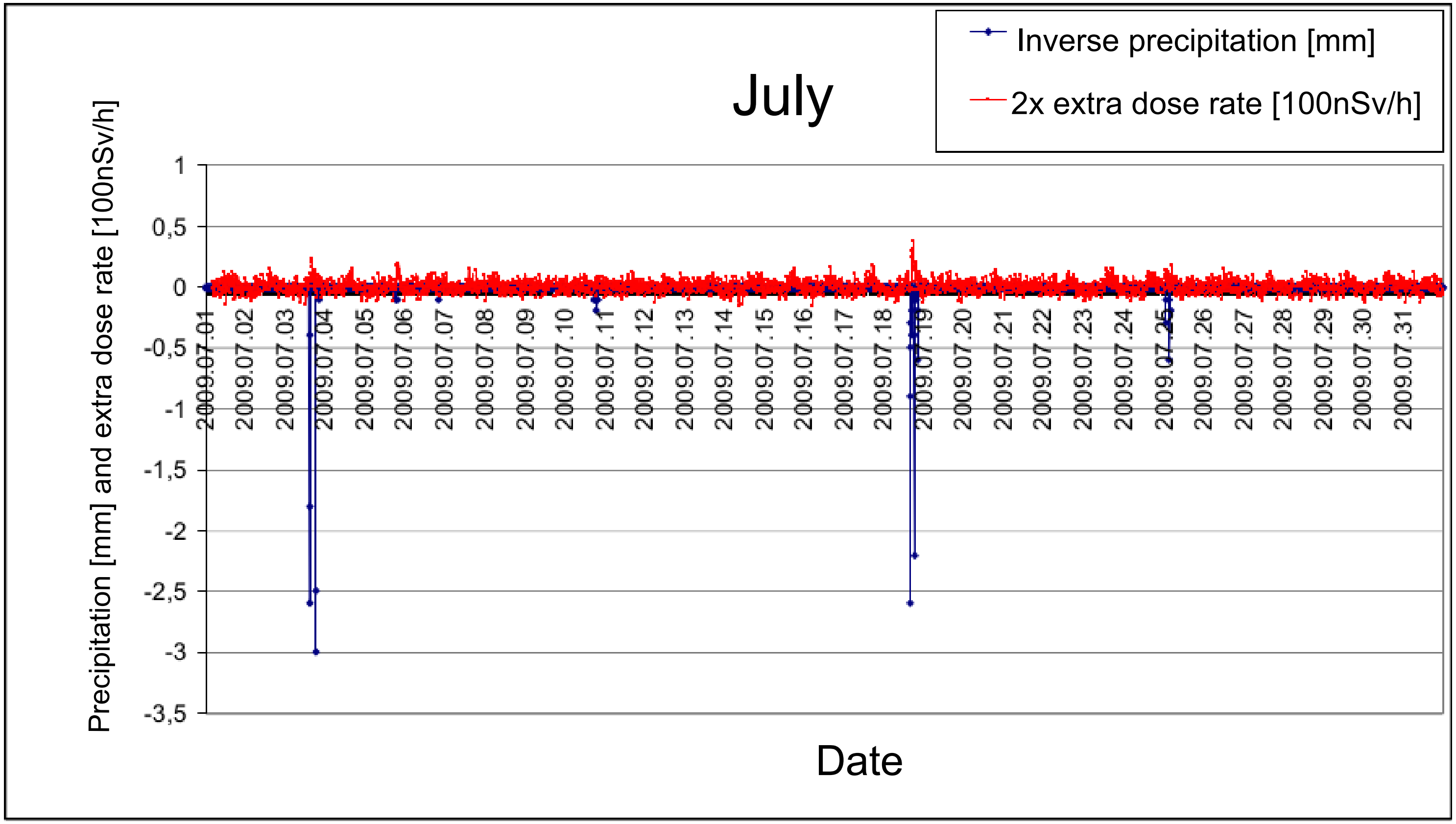


06. 28.

- Biggest precipitation in 2009 considering 10 minutes time frames (14,6mm)
- Summed up: 29,7mm rain
- The increase in the dose rate also the highest: 91 nSv/h
- Even 2 hours after the rain event there was still a 2,5nSv/h extra dose rate

Monthly gamma dose rates - July

2 bigger rain events and several little ones (total: 21,6mm).



07. 03.

- 1) 4,8mm rain
- 1) 5,13 nSv/h extra dose rate
- 2) 5,5mm rain
- 2) no extra dose rate

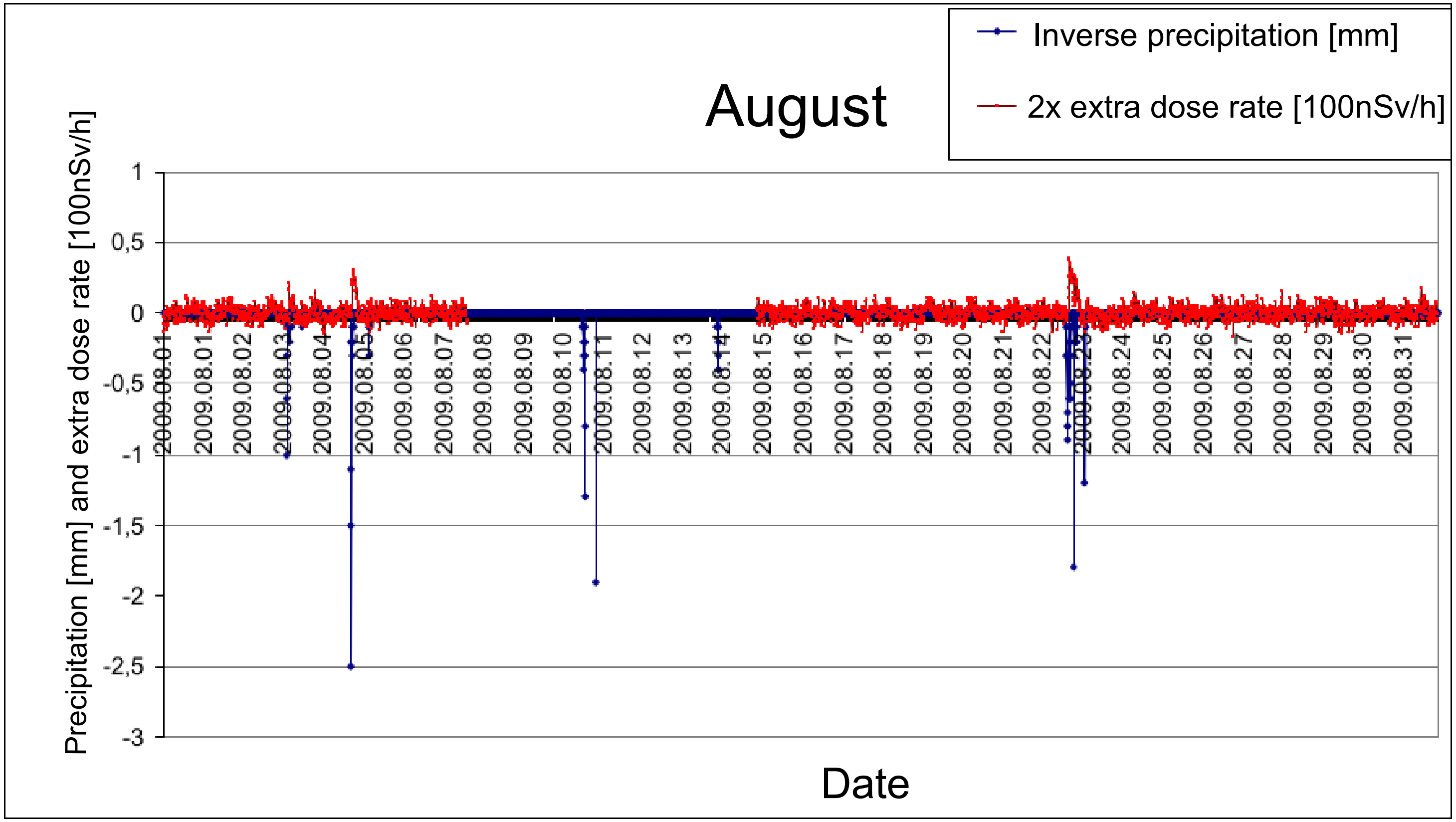
07. 18.

- 8,6mm rain
- 12 nSv/h extra dose rate

Biggest value: 79,2 nSv/h

Monthly gamma dose rates - August

The detectors did not work between the 8th and 15th of August.



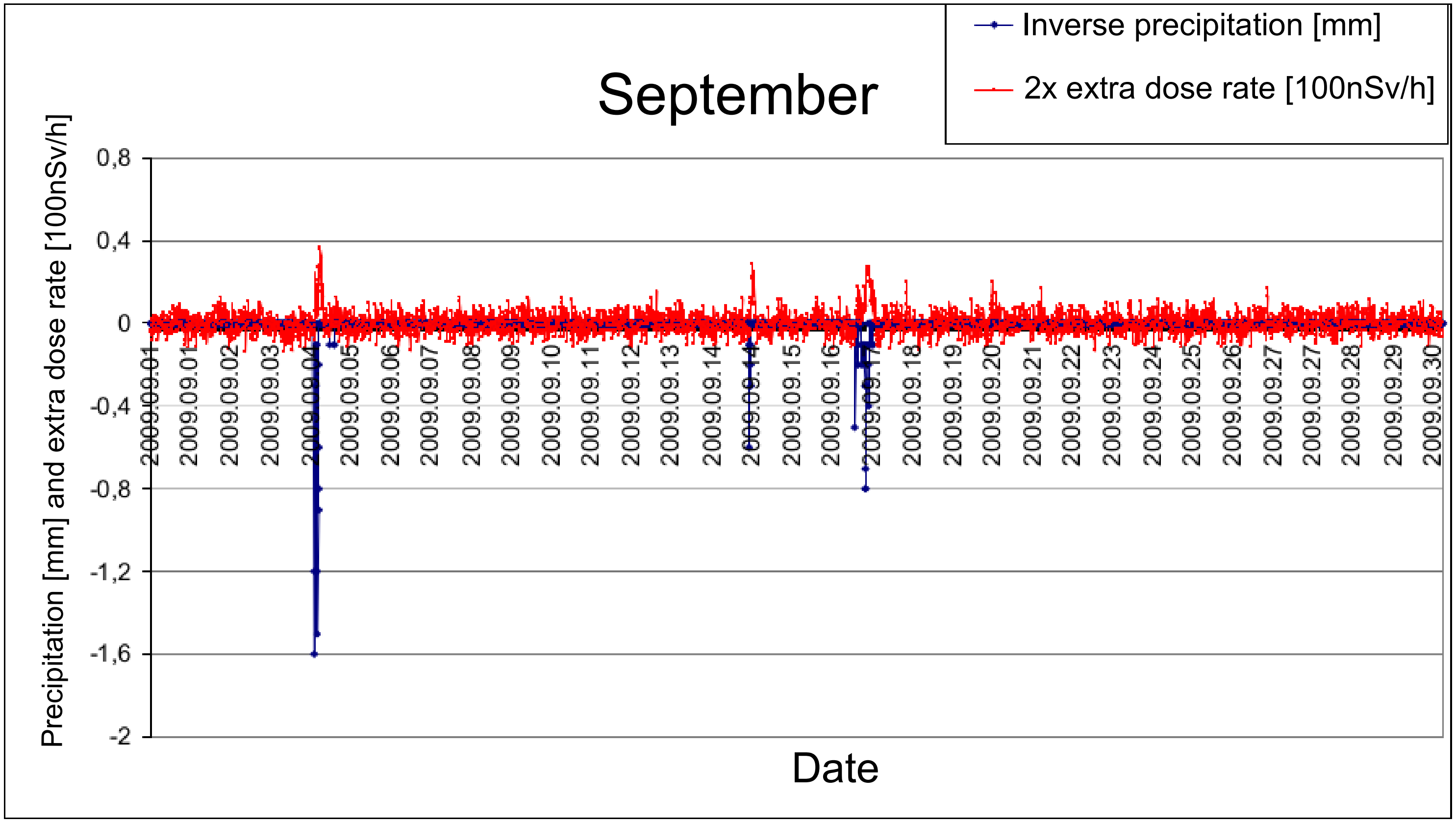
08. 23.

- 9,8mm rain
- In 6 hours
- Peak at 80,7 nSv/h

The other events have rain peaks as well.

Monthly gamma dose rates - September

September was the third most dry month after April and May in 2009 (18mm in total).

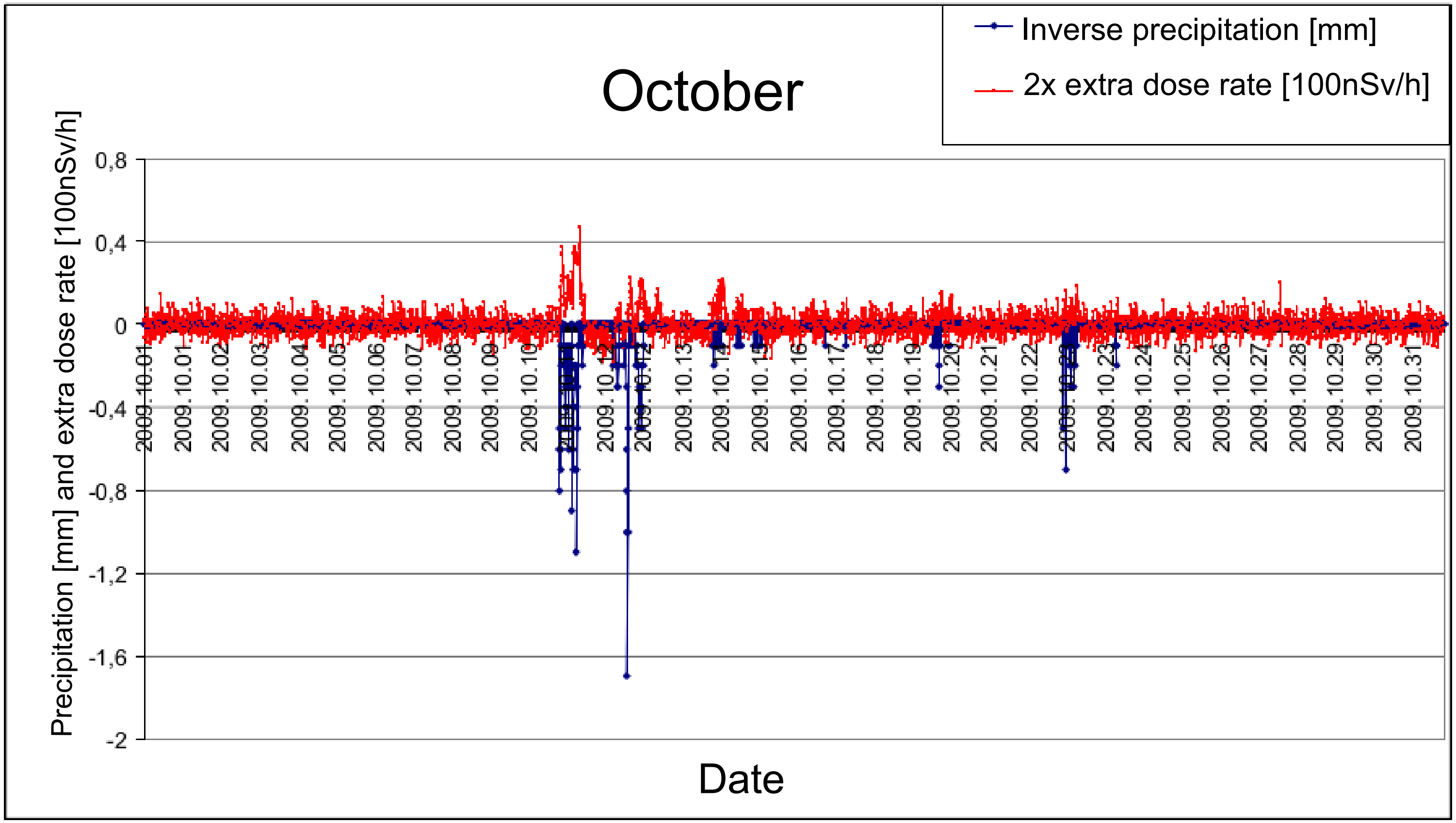


09. 05.

- Only 0,2mm rain
- Still had a peak

All the events have rain peaks.

Monthly gamma dose rates - October

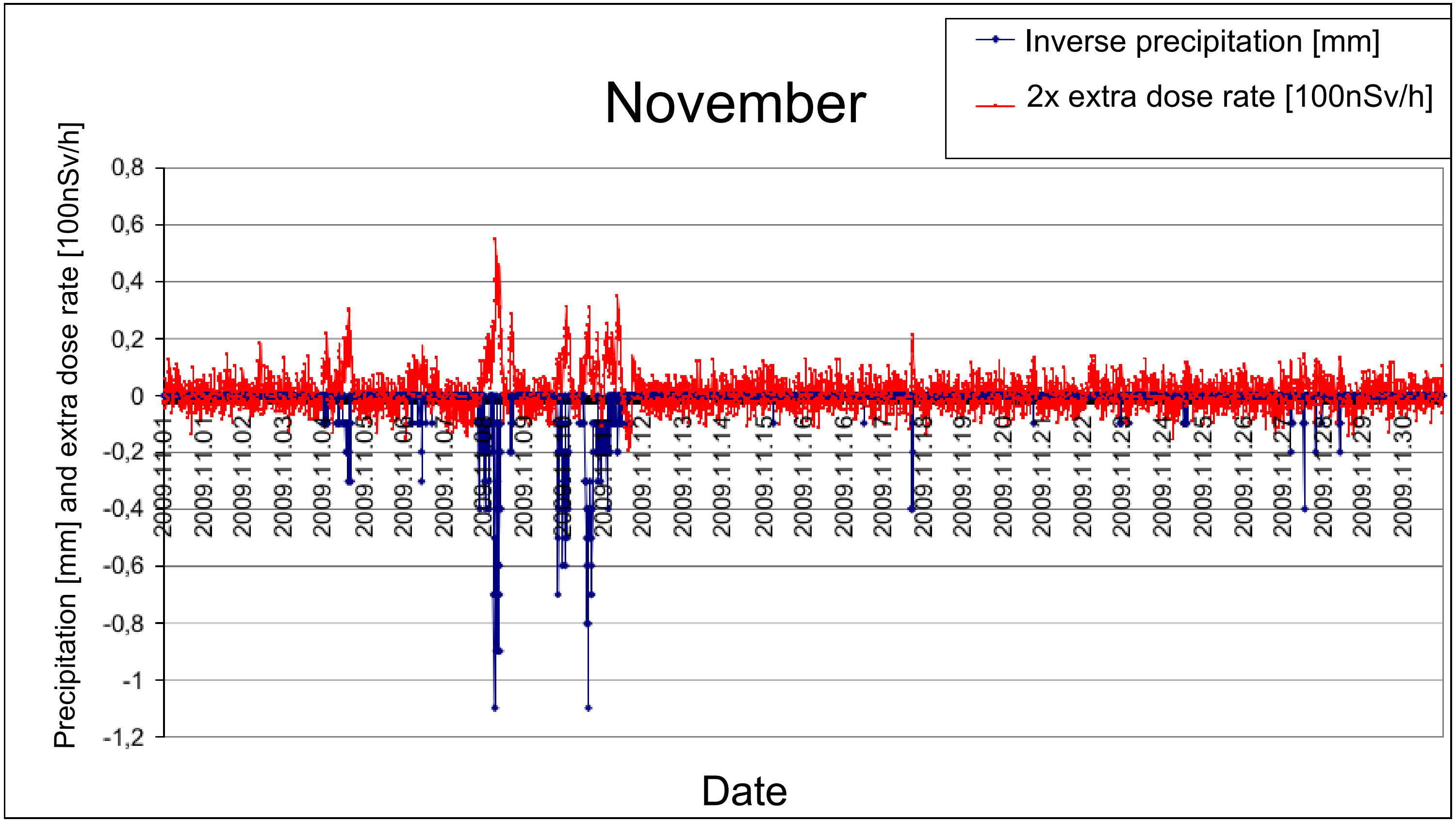


10. 10.

- It rained for 13 hours
- Total precipitation: 20,5mm
- It stopped for 40mins, that can be seen also on the dose rates.
- Dose rate maximum: 86,2 nSv/h

Monthly gamma dose rates - November

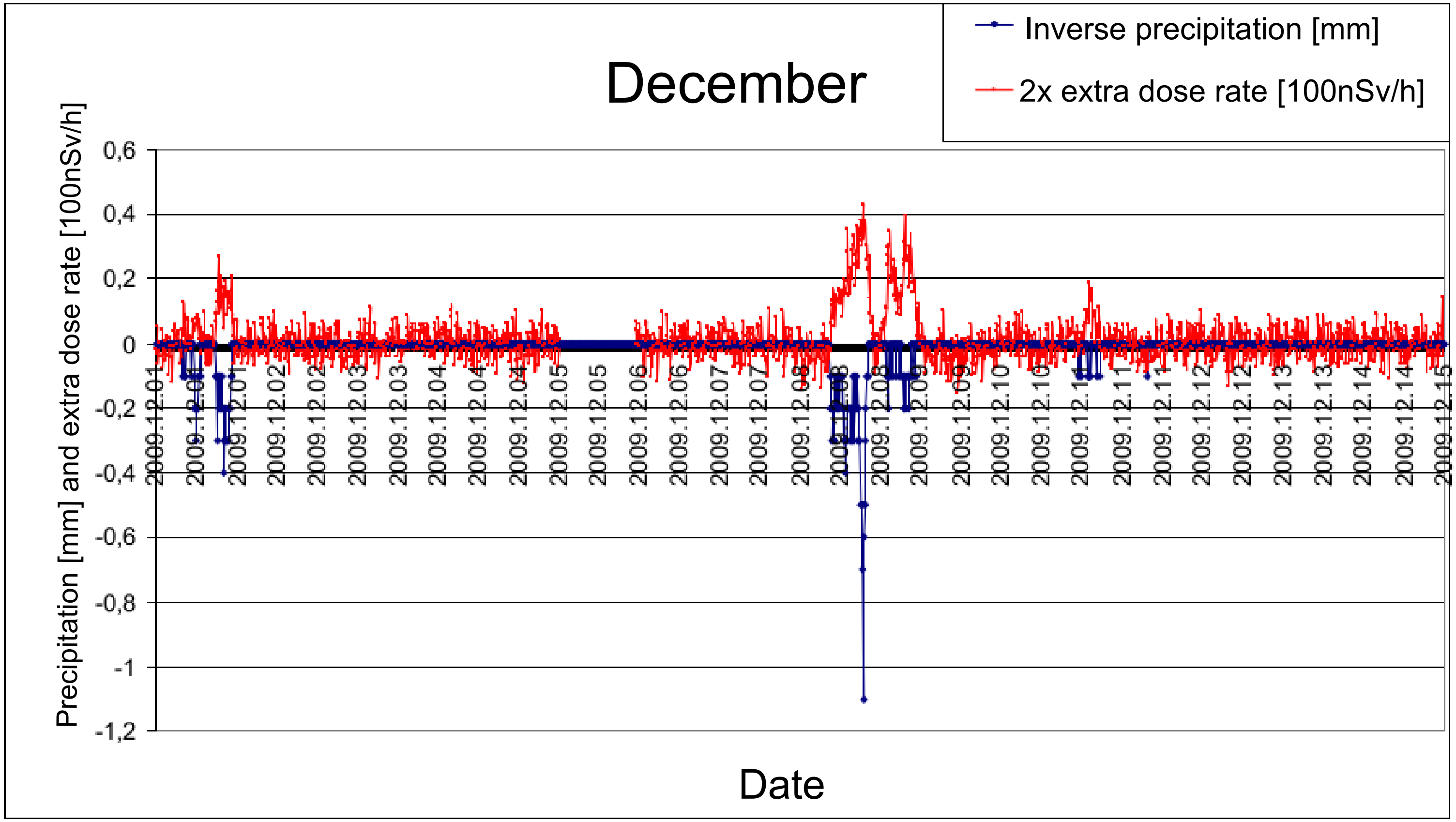
November had the highest background dose rate (61,23 nSv/h).



- Total precipitation: 67,2 mm
- Lots of precipitation events
- Small events had small/zero effect on the dose rate

Monthly gamma dose rates - December

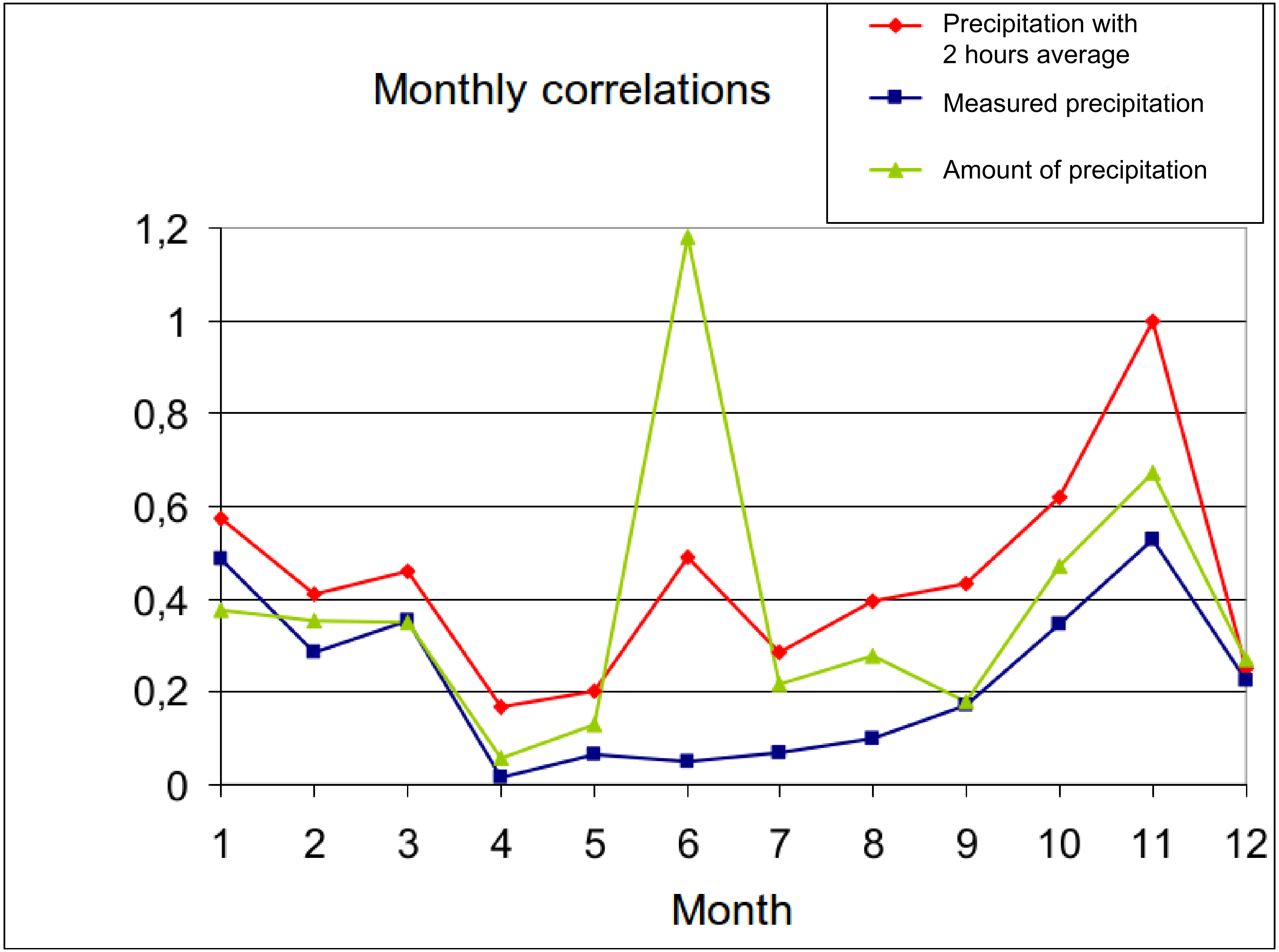
The detectors provide data only until the 15th of December.



12. 08.

- It rained for 12 hours
- Total precipitation: 15,4mm
- Maximum: 1,1mm/10mins 83,8 nSv/h

Monthly correlations

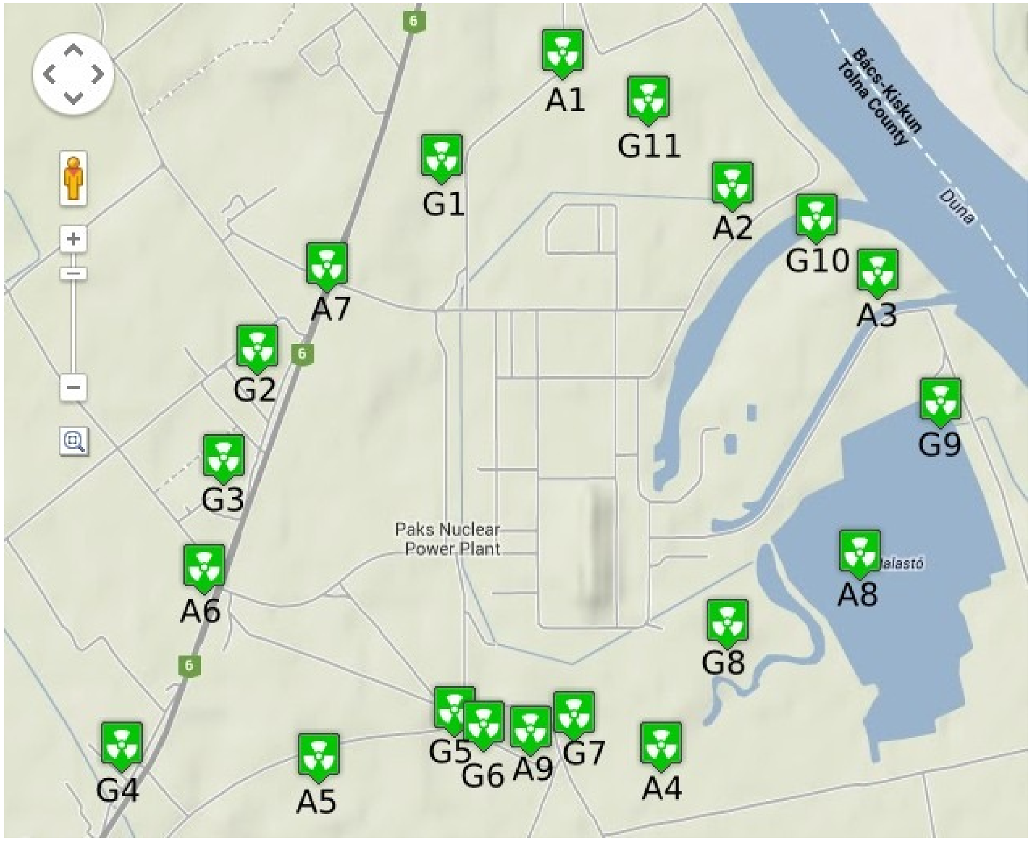


The months with higher precipitation amount (January, June, October, November) have a correlation above 0,5.

In the dry months (April, May, July, August, September) other factors have relatively bigger effects and therefore the correlation is lower.

Data from the Paks measuring system

Measured for safety reasons,
To distinguish natural and artificial effects.



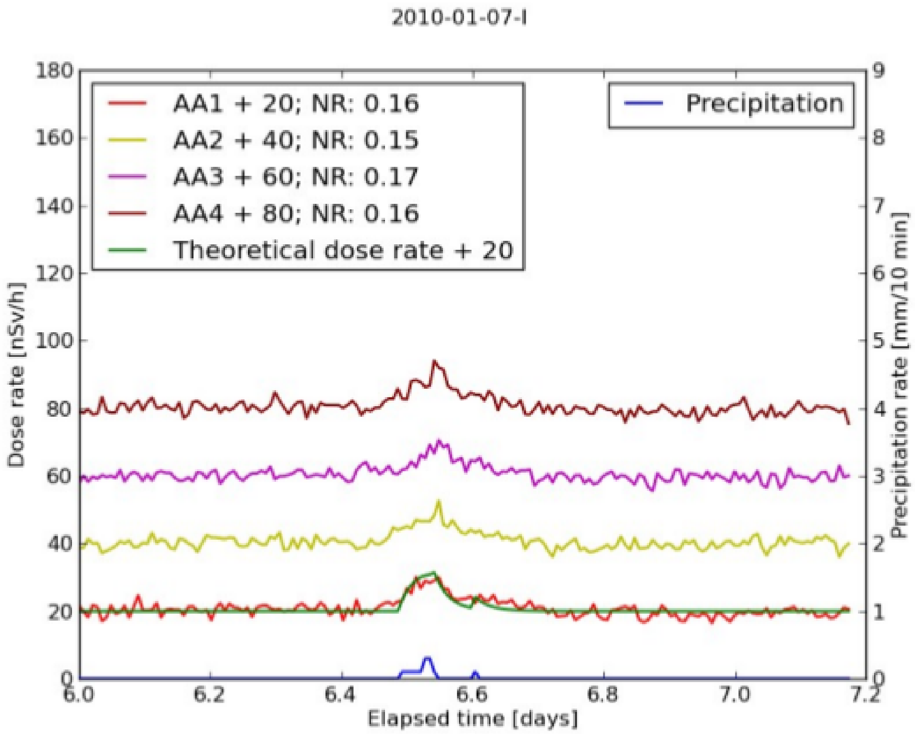
Gamma dose rate detectors

- 20 pieces
- Saves data in 10 minutes intervals
- The unit of measure is Sv/h
- Measuring range: 10 nSv/h – 10 Sv/h
- Measurements in 2010 ans 2011

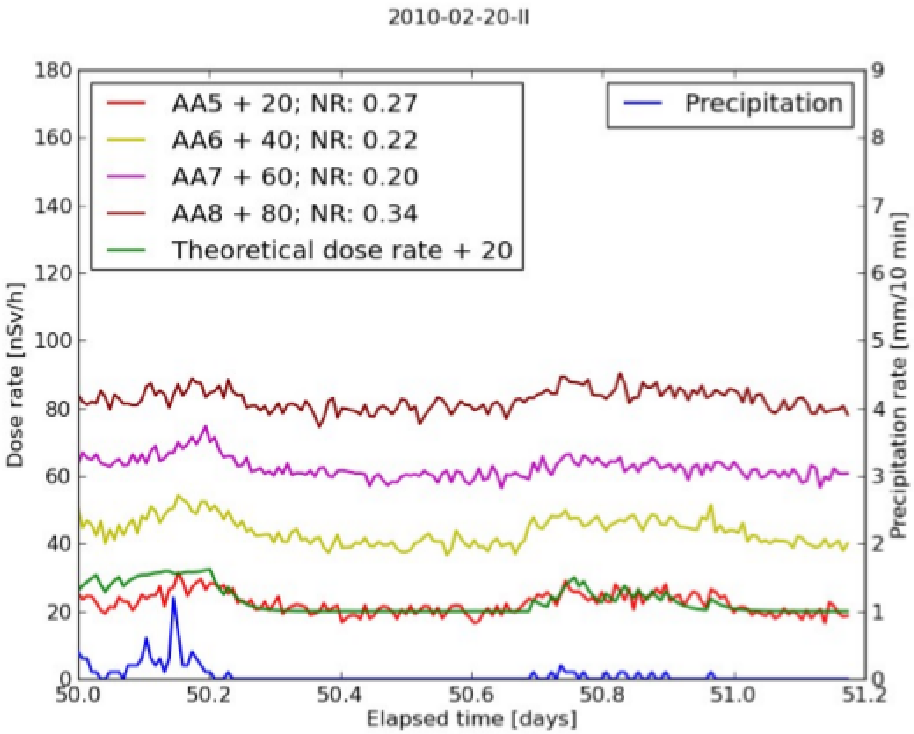
Meteorological parameters

- Saves data in 10 minutes intervals
- Measures temperature, precipitation, solar irradiance, velocity and direction of wind, atmospheric pressure
- We only use the precipitation

2010 Winter

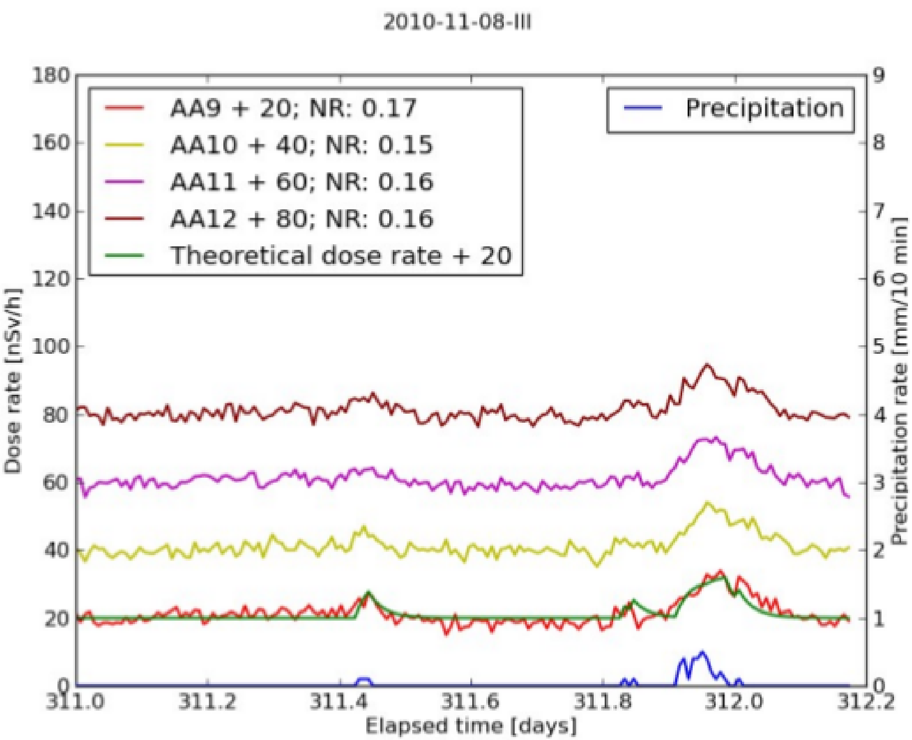


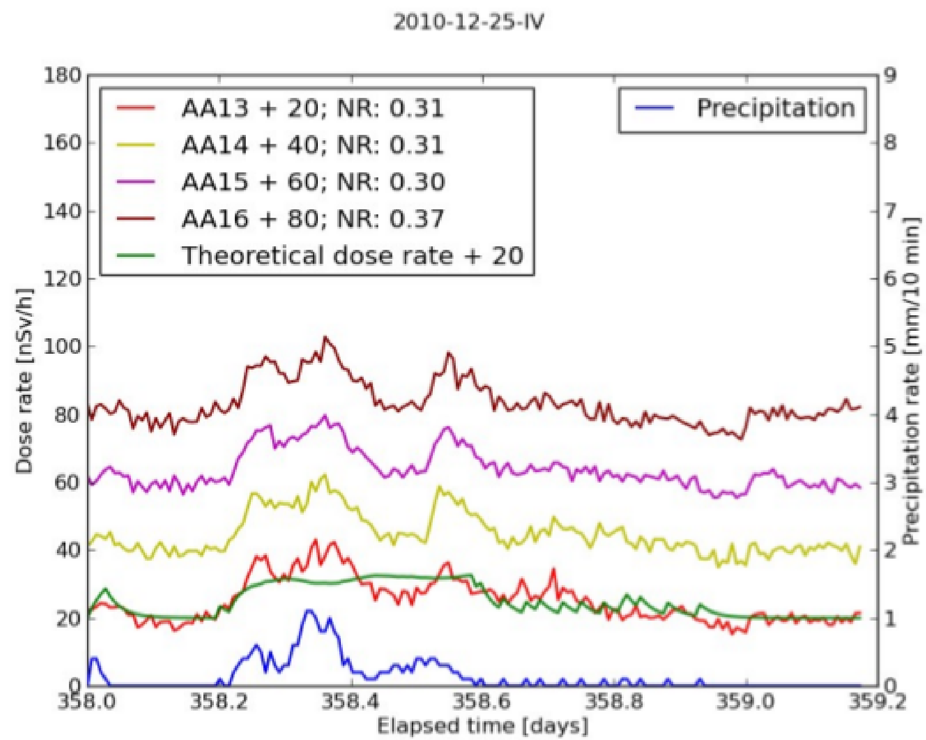
2010-01-07-I
AA1 + 20; NR: 0.16
AA2 + 40; NR: 0.15
AA3 + 60; NR: 0.17
AA4 + 80; NR: 0.16
Theoretical dose rate + 20
Precipitation
Dose rate [nSv/h]
Precipitation rate [mm/10 min]
Elapsed time [days]



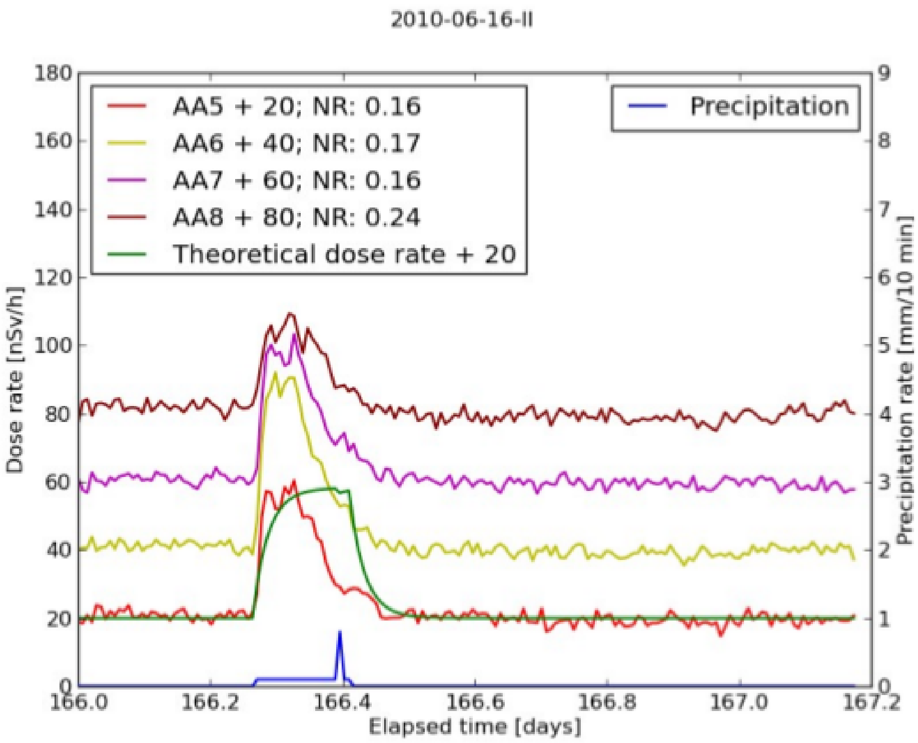
2010-02-20-II
AA5 + 20; NR: 0.27
AA6 + 40; NR: 0.22
AA7 + 60; NR: 0.20
AA8 + 80; NR: 0.34
Theoretical dose rate + 20
Precipitation
Dose rate [nSv/h]
Precipitation rate [mm/10 min]
Elapsed time [days]

2010 Winter

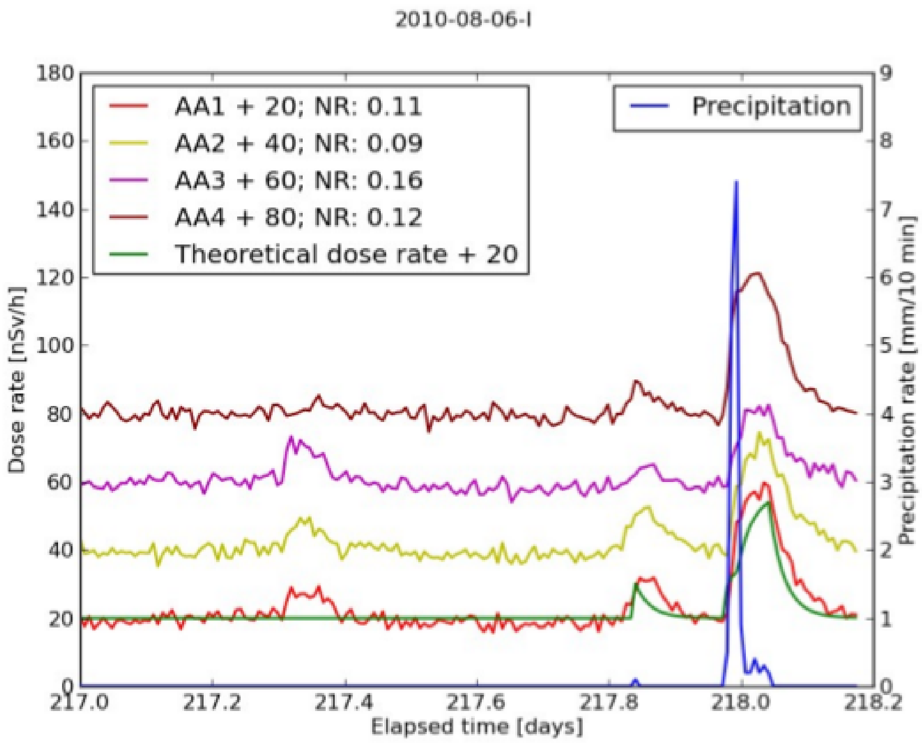

2010-11-08-III
AA9 + 20; NR: 0.17
AA10 + 40; NR: 0.15
AA11 + 60; NR: 0.16
AA12 + 80; NR: 0.16
Theoretical dose rate + 20
Precipitation
Dose rate [nSv/h]
Precipitation rate [mm/10 min]
Elapsed time [days]
311.0
311.2
311.4
311.6
311.8
312.0
312.2


2010-12-25-IV
AA13 + 20; NR: 0.31
AA14 + 40; NR: 0.31
AA15 + 60; NR: 0.30
AA16 + 80; NR: 0.37
Theoretical dose rate + 20
Precipitation
Dose rate [nSv/h]
Precipitation rate [mm/10 min]
Elapsed time [days]
358.0
358.2
358.4
358.6
358.8
359.0
359.2

2010 Summer

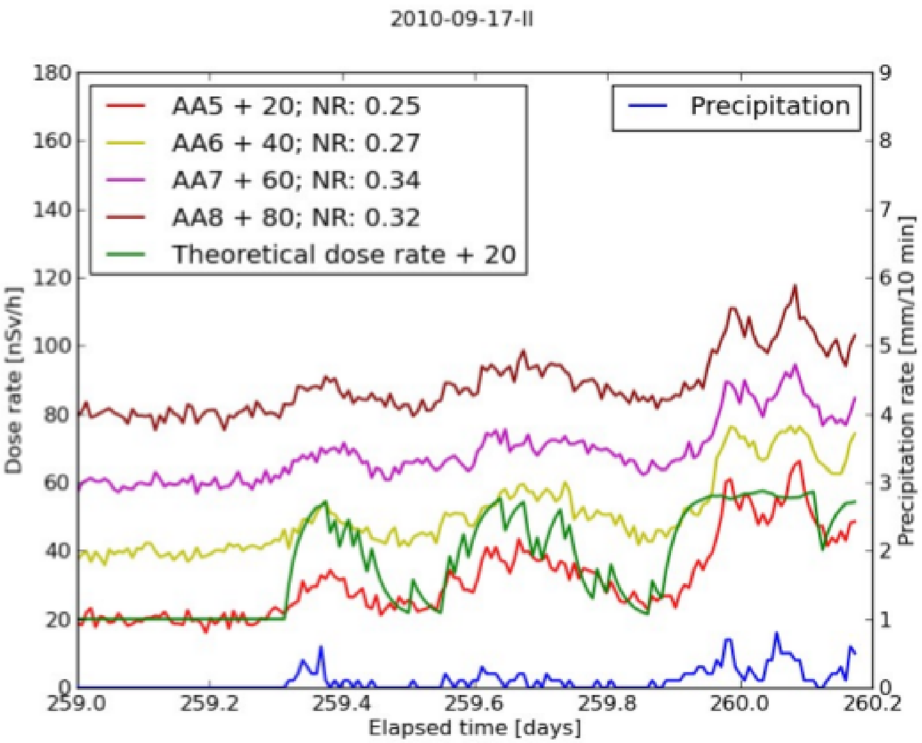


2010-06-16-II
AA5 + 20; NR: 0.16
AA6 + 40; NR: 0.17
AA7 + 60; NR: 0.16
AA8 + 80; NR: 0.24
Theoretical dose rate + 20
Precipitation
Dose rate [nSv/h]
Precipitation rate [mm/10 min]
Elapsed time [days]

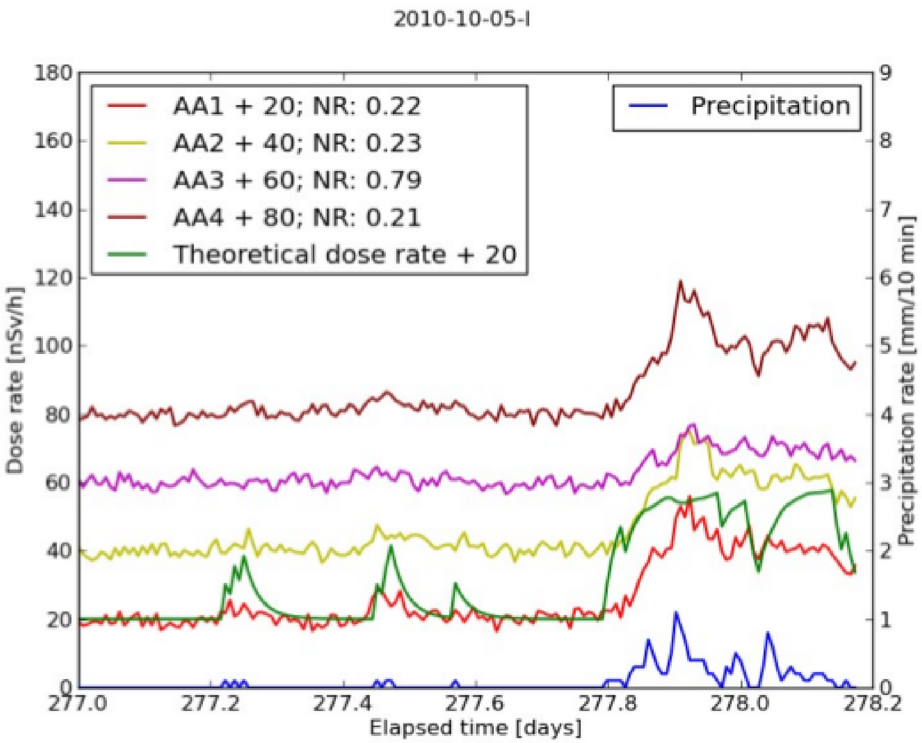


2010-08-06-I
AA1 + 20; NR: 0.11
AA2 + 40; NR: 0.09
AA3 + 60; NR: 0.16
AA4 + 80; NR: 0.12
Theoretical dose rate + 20
Precipitation
Dose rate [nSv/h]
Precipitation rate [mm/10 min]
Elapsed time [days]

2010 Autumn

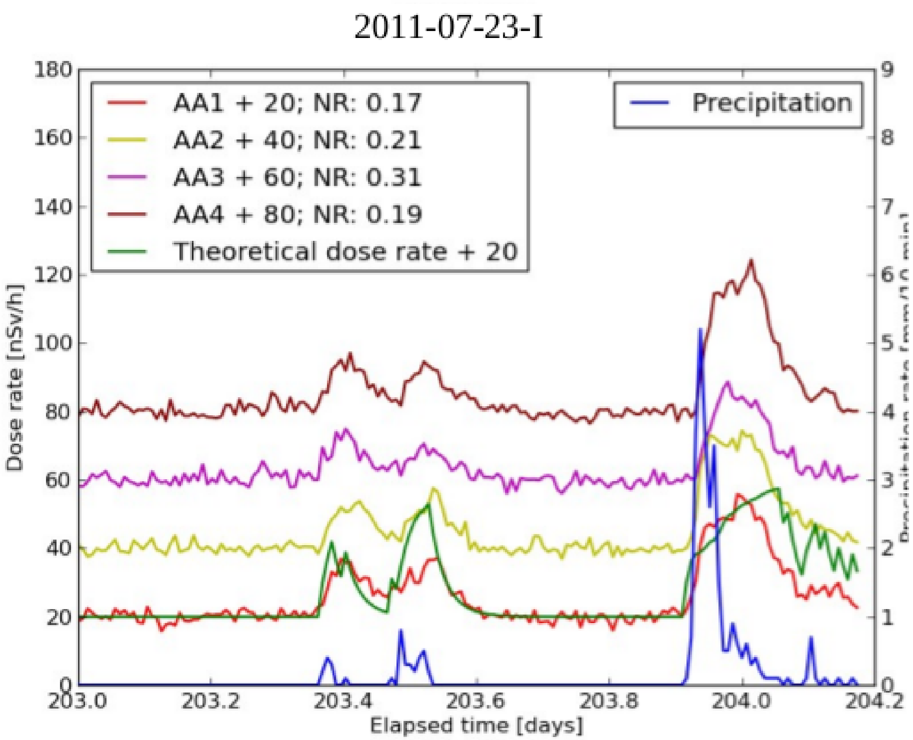


2010-09-17-II
AA5 + 20; NR: 0.25
AA6 + 40; NR: 0.27
AA7 + 60; NR: 0.34
AA8 + 80; NR: 0.32
Theoretical dose rate + 20
Precipitation
Dose rate [nSv/h]
Precipitation rate [mm/10 min]
Elapsed time [days]

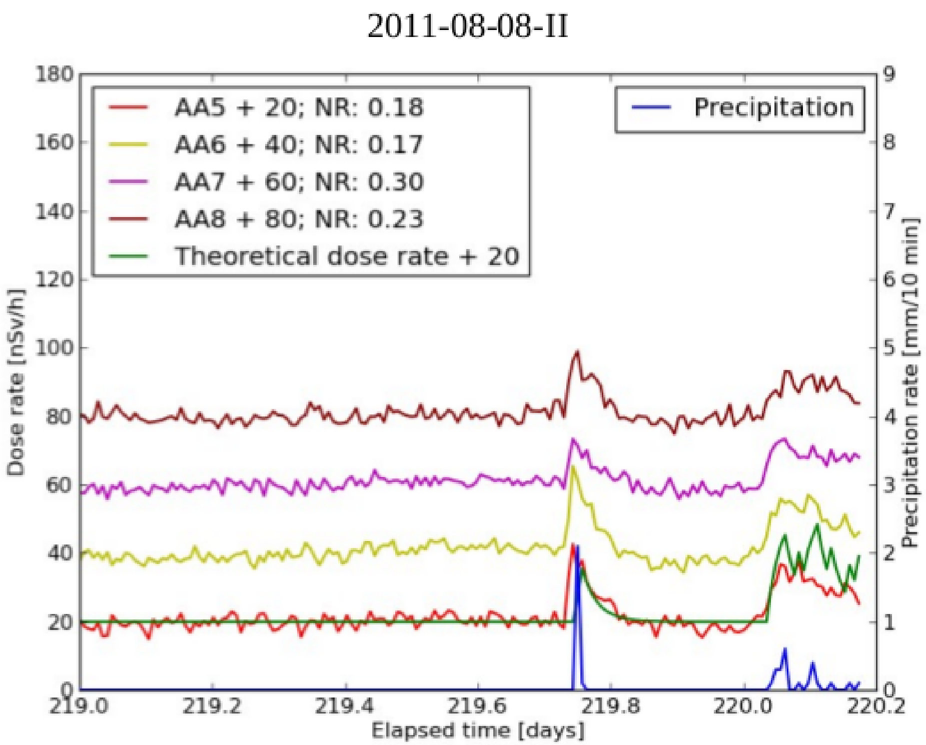


2010-10-05-I
AA1 + 20; NR: 0.22
AA2 + 40; NR: 0.23
AA3 + 60; NR: 0.79
AA4 + 80; NR: 0.21
Theoretical dose rate + 20
Precipitation
Dose rate [nSv/h]
Precipitation rate [mm/10 min]
Elapsed time [days]

2011 Summer



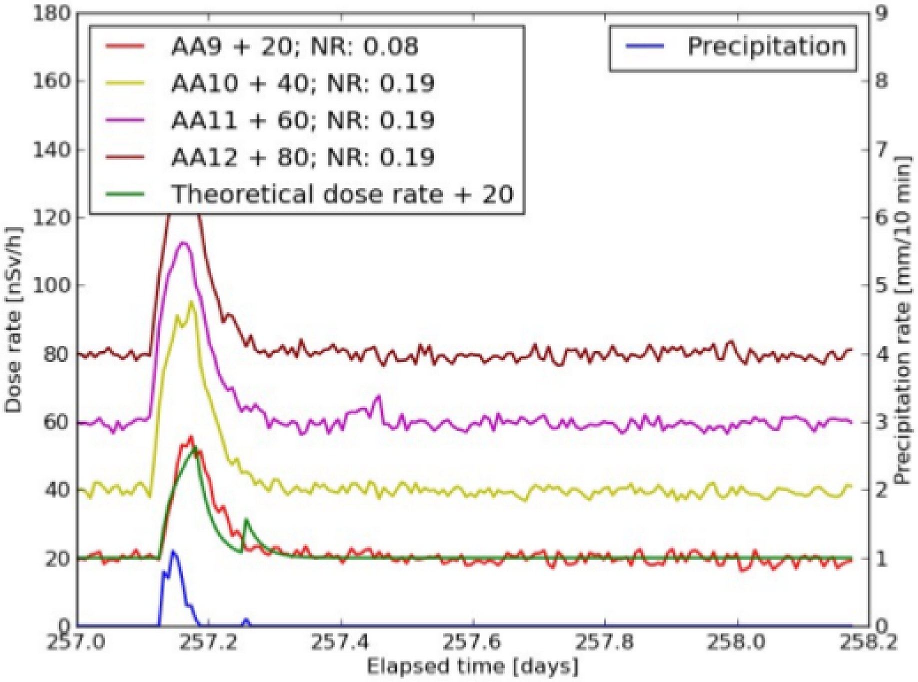
2011-07-23-I
AA1 + 20; NR: 0.17
AA2 + 40; NR: 0.21
AA3 + 60; NR: 0.31
AA4 + 80; NR: 0.19
Theoretical dose rate + 20
Precipitation
Dose rate [nSv/h]
Elapsed time [days]



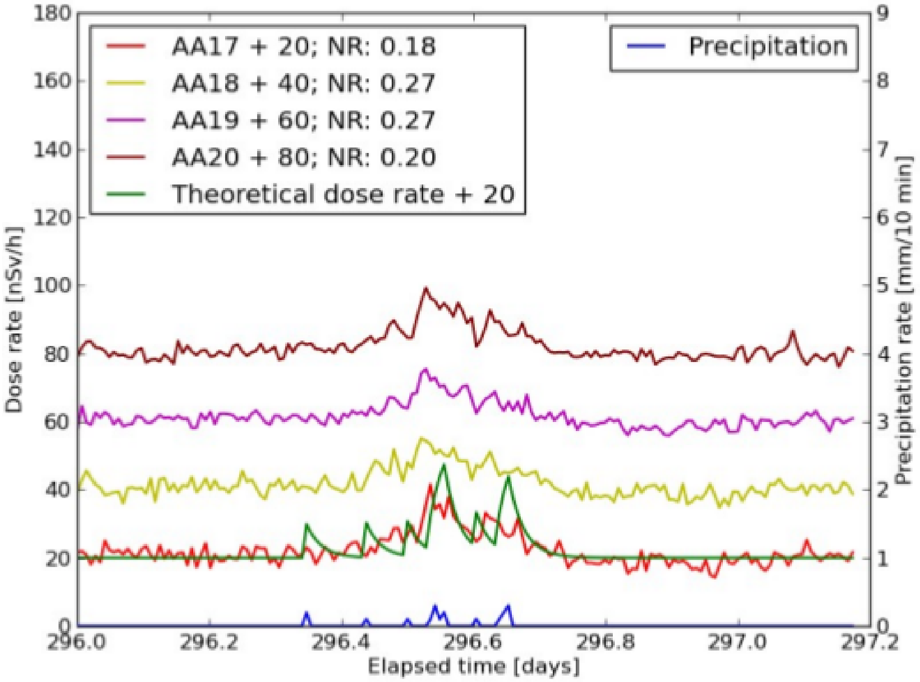
2011-08-08-II
AA5 + 20; NR: 0.18
AA6 + 40; NR: 0.17
AA7 + 60; NR: 0.30
AA8 + 80; NR: 0.23
Theoretical dose rate + 20
Precipitation
Dose rate [nSv/h]
Precipitation rate [mm/10 min]
Elapsed time [days]

2011 Autumn

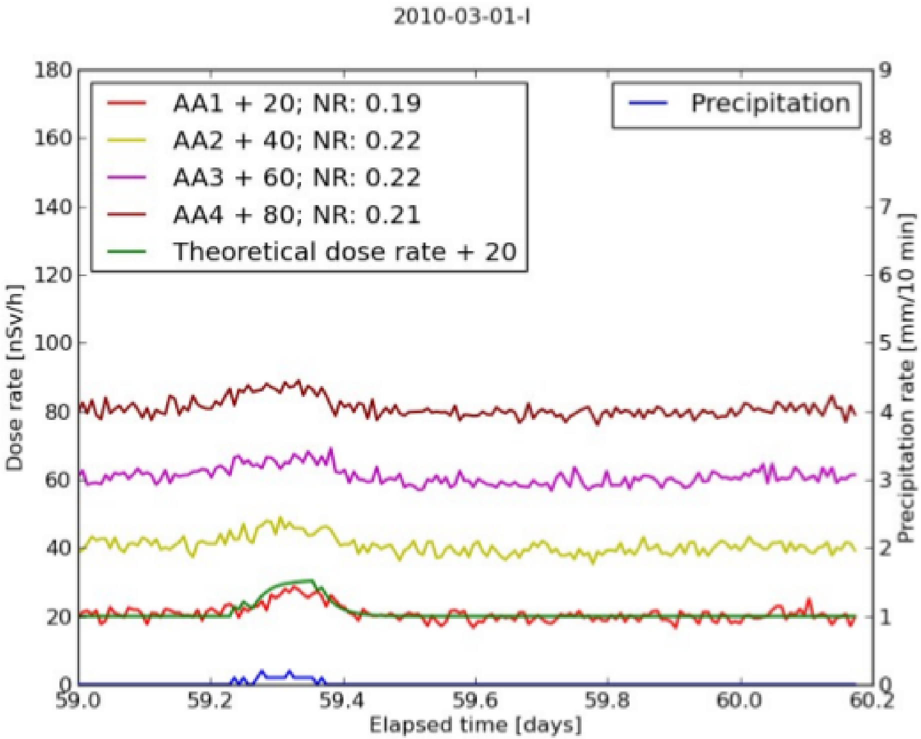
2011-09-15-III

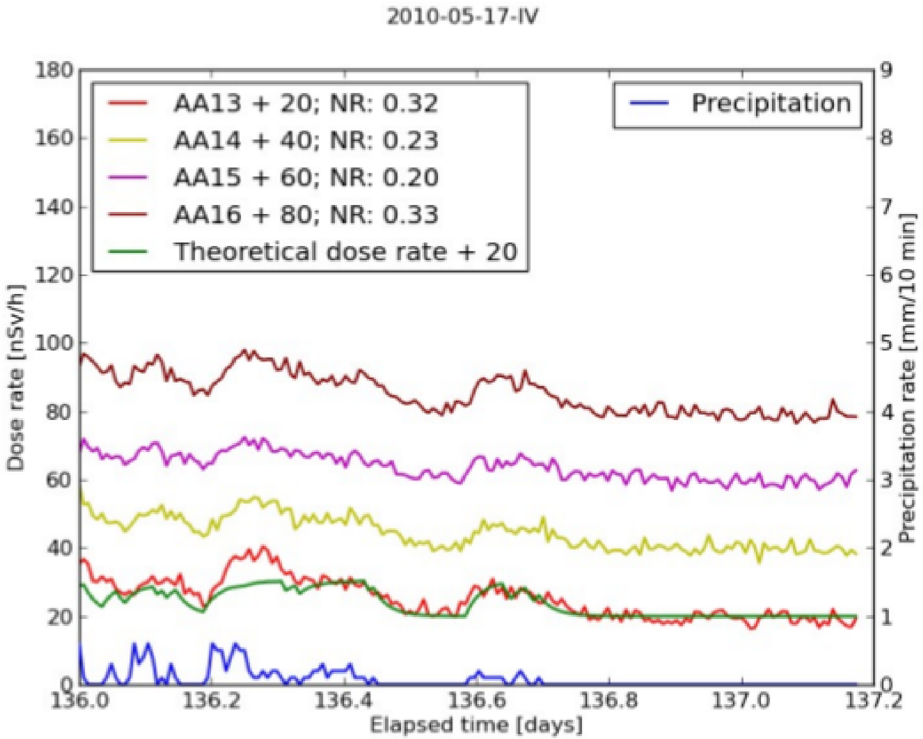


2011-10-24-V

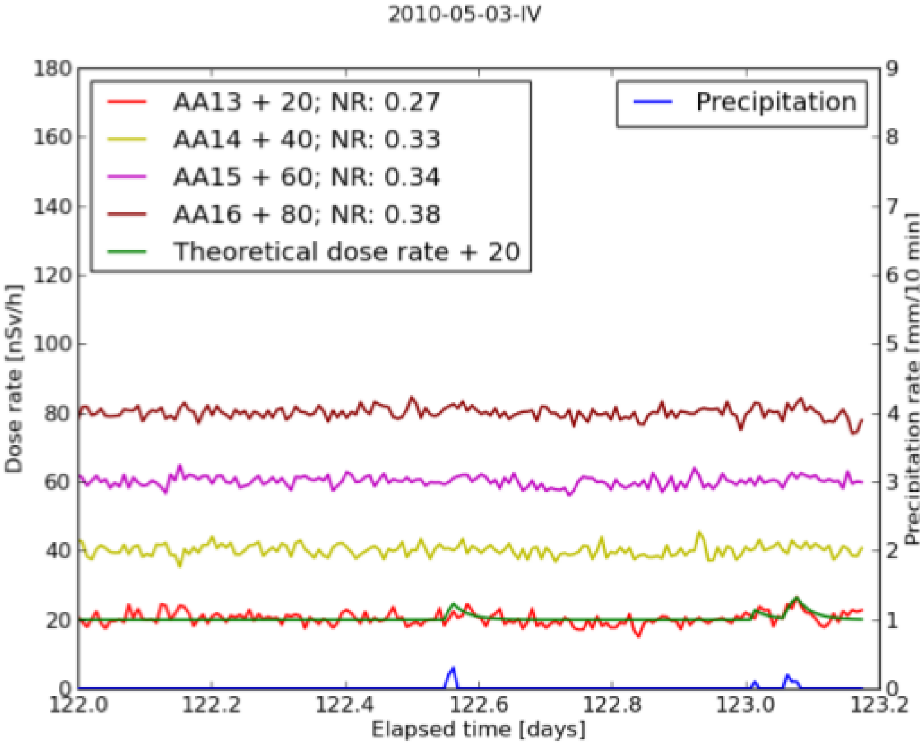


2010 Spring

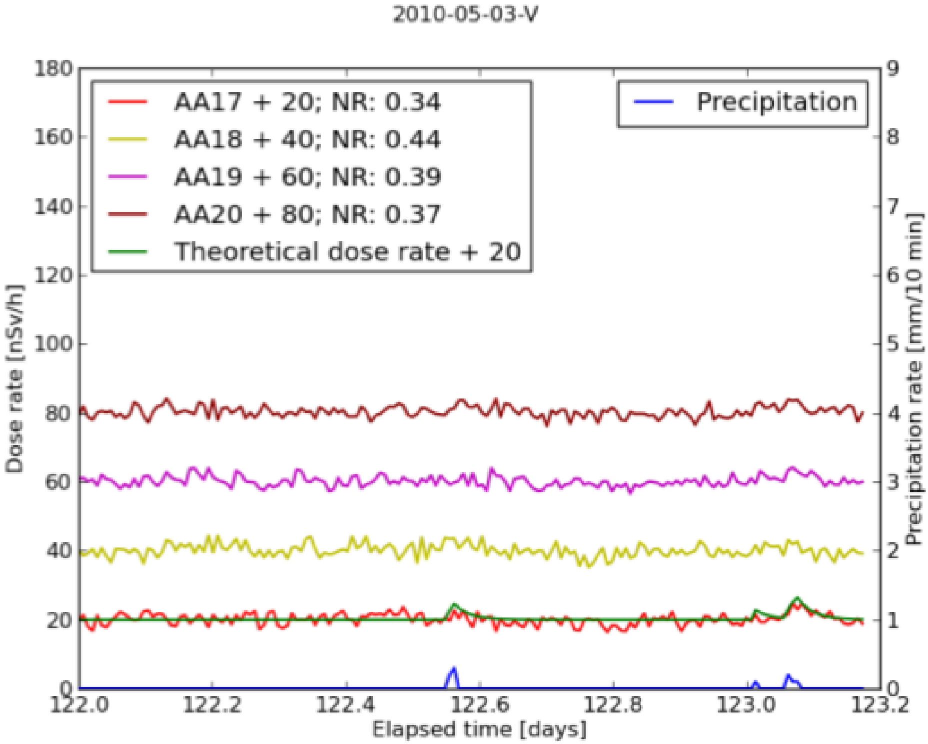

2010-03-01-I
AA1 + 20; NR: 0.19
AA2 + 40; NR: 0.22
AA3 + 60; NR: 0.22
AA4 + 80; NR: 0.21
Theoretical dose rate + 20
Precipitation
Dose rate [nSv/h]
Precipitation rate [mm/10 min]
Elapsed time [days]
59.0
59.2
59.4
59.6
59.8
60.0
60.2
0
20
40
60
80
100
120
140
160
180
0
1
2
3
4
5
6
7
8
9


2010-05-17-IV
AA13 + 20; NR: 0.32
AA14 + 40; NR: 0.23
AA15 + 60; NR: 0.20
AA16 + 80; NR: 0.33
Theoretical dose rate + 20
Precipitation
Dose rate [nSv/h]
Precipitation rate [mm/10 min]
Elapsed time [days]
136.0
136.2
136.4
136.6
136.8
137.0
137.2
0
20
40
60
80
100
120
140
160
180
0
1
2
3
4
5
6
7
8
9

2010 Spring

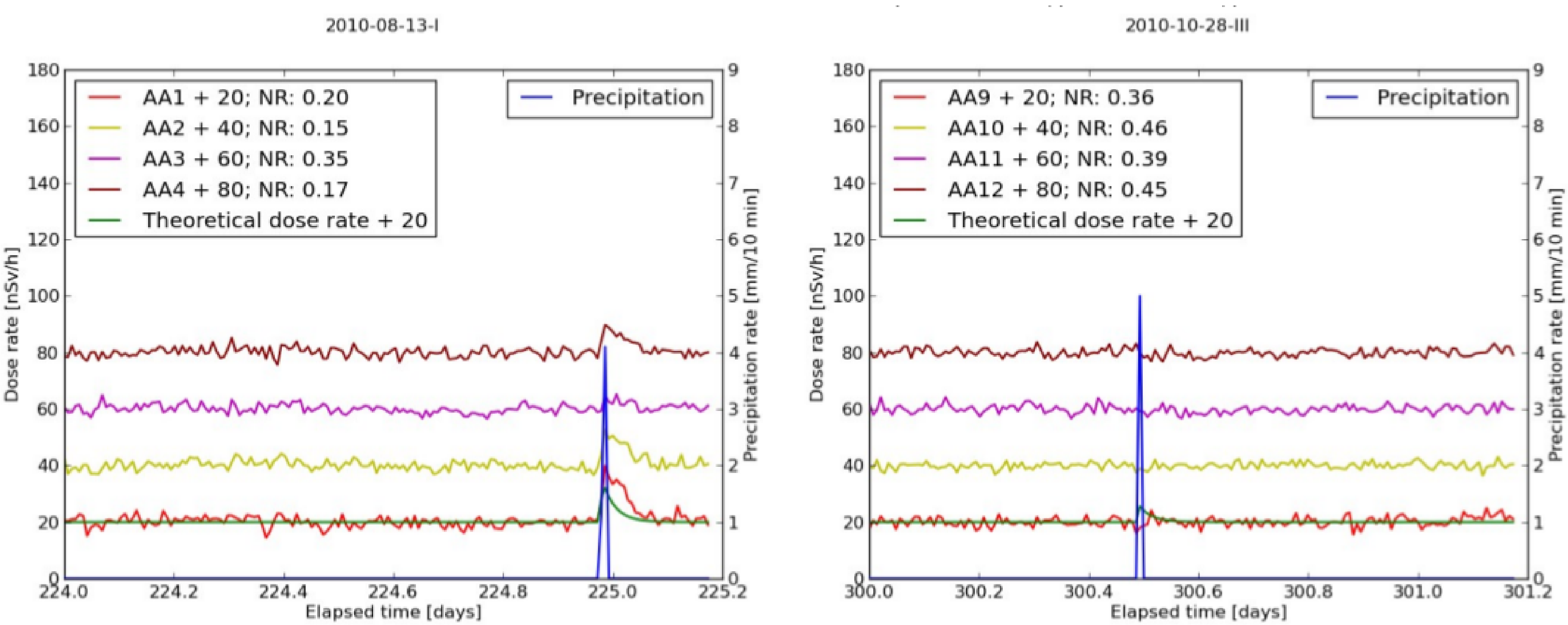


2010-05-03-IV
AA13 + 20; NR: 0.27
AA14 + 40; NR: 0.33
AA15 + 60; NR: 0.34
AA16 + 80; NR: 0.38
Theoretical dose rate + 20
Precipitation
Dose rate [nSv/h]
Precipitation rate [mm/10 min]
Elapsed time [days]

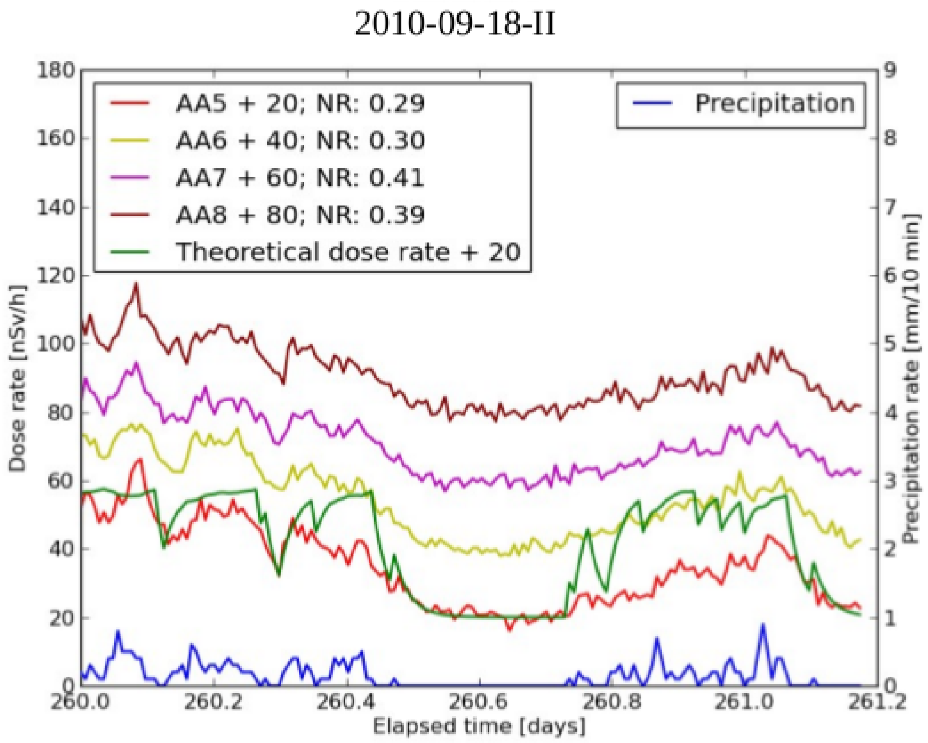


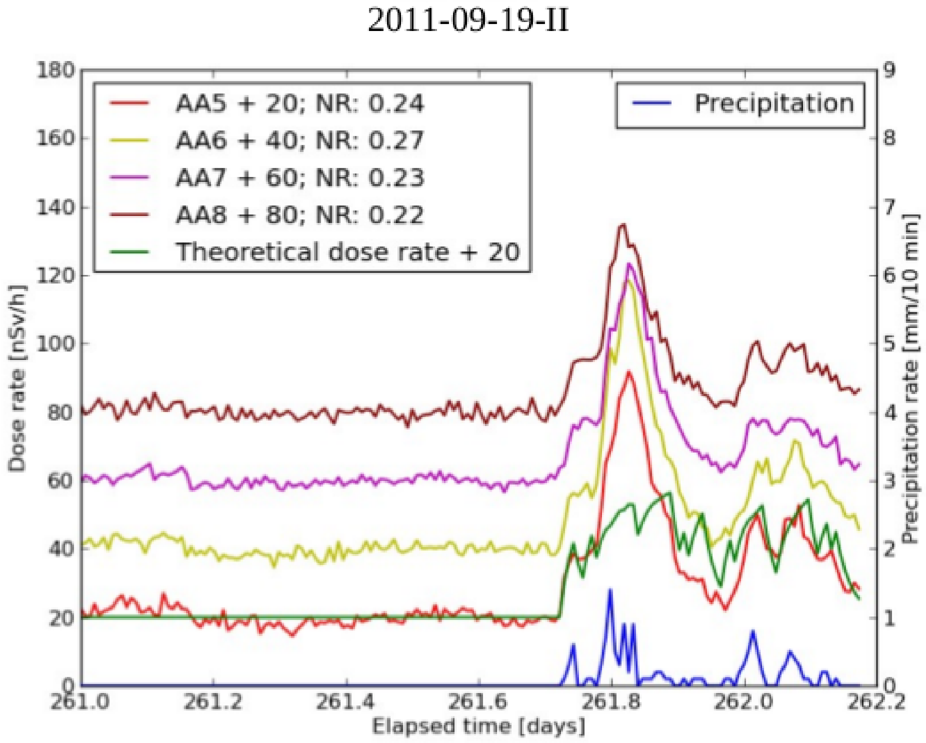
2010-05-03-V
AA17 + 20; NR: 0.34
AA18 + 40; NR: 0.44
AA19 + 60; NR: 0.39
AA20 + 80; NR: 0.37
Theoretical dose rate + 20
Precipitation
Dose rate [nSv/h]
Precipitation rate [mm/10 min]
Elapsed time [days]

2010 Summer + Autumn



2010, 2011 Autumn





Summary

The semi-empirical rain-out model successfully described the ground level gamma dose rates produced by the radon daughter elements.
Different parameter sets for different times of the year are required due to the seasonal change in the radon-exhalation and different environmental circumstances.

Winter	low activity concentration of radon in cloud air	low cloud depth
Summer and Autumn	highest radioactivity	high cloud depth
Spring	lowest radioactive concentrations	cloud depth between the winter and summer depth

These considerations allowed the model to give good results for the majority of rainfall cases.

Full coverage could not be achieved, but that was expected considering the complexity of the atmospheric processes.

With this parametrization we can estimate the radioactive dose rate increase provided by rainfall events.

Thank you for your attention!

Acknowledgements to **Mayer Petra** and **Bakos Katinka** who wrote their Bsc thesis about this topic and provided me a reliable source to get data and plots from.