



# Evaluation of the precision of radioactive deposition measurements

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## Abstract

Sampling and measurement of radioactive deposition (fallout) plays an important role in environmental radiation monitoring. It provides information on the existing environmental radioactive contamination levels and detects any changes in the activity concentrations of radionuclides deposited on the ground, and consequently enables the assessment and execution of actions for the protection of public. By its joint monitoring with airborne concentrations by active sampling and prevailing meteorological conditions, the migration of radionuclides through the environmental compartments, following a discharge to the atmosphere, can also be investigated.

To ensure the adequate fulfillment of the above objectives, the precision of the radioactive deposition measurements has been examined at the KFKI Campus in Budapest, in a series of experiments over several years. The obtained results are summarized in this work. We experimentally quantified the uncertainty arising from the deposition sampling process and evaluated its relation to the uncertainty associated to the analytical measurement, the gamma spectrometric analysis. Key sources of the uncertainty of the measurement process were identified which should be the target of reduction in order to enhance the overall performance of the method. In addition to the combined sampling of the total deposition, measurement data of separate sampling of wet and dry deposition are also available. These measurements facilitated the examination of the deposition processes, furthermore, the validation of dry and wet deposition models and model parameters.

## Measurements

As part of the environmental radiological monitoring system operated in the KFKI Campus (located at latitude, 47°29'20.89"N and longitude, 18°57'13.44"E) in Budapest, the sampling of deposition is carried out at 4 compound measurement stations (Stations 1,2,5,6), using identical stainless steel collectors with a surface area of 0.2 m<sup>2</sup>. Continuous sampling are performed, the routine sampling duration (i.e. the frequency with which the collected samples are taken off) varies from weekly to monthly periods at the different stations. In addition to the existing samplers for combined sampling of the total deposition, some of which are located at the same measurement station for parallel collection, a collector equipped with a rain sensor, hence capable of separate sampling of wet and dry deposition, has been installed on the site. The sampling is followed by subsequent off-line laboratory measurements, which is first preceded by sample preparation, during which non-volatile components in the liquid-form sample are concentrated by evaporation. Nuclide specific determination of activity concentrations of radionuclides deposited on the ground is possible by means of gamma spectrometry. High-purity germanium (HPGe) detectors with relative efficiencies of 20-40% are used, typically in iron-shielded low-background measuring chambers. Routine measurement time is 60 000 s.

Table 1: Summary of routine measurement conditions

Deposition sample type	Sample volume	Sampling frequency	Analysis	Counting time
Wet and dry (combined)	0.2 m <sup>2</sup> (collector surface area)	Weekly (Stat. 1,6)	Gamma spectrometry	60 000 s
Dry (separated)		Monthly (Stat. 1,2,5)		
Wet (separated)		Monthly (Stat. 1)		

## Evaluations

### DETERMINATION OF THE MEASURAND

The measurand is activity concentration ( $a$ ), in the measurement model of its determination  $n_n$  is the net peak area of the full-energy peak (FEP) attributed to the radionuclide of interest,  $t_m$  is the counting time,  $w$  is the conversion factor which is a function of other input quantities upon which the measurand depends, such as FEP efficiency ( $\epsilon$ ), emission probability ( $p_\gamma$ ), sample volume (collector surface area;  $q$ ), correction factors for decay during sampling (assuming continuous accumulation on the sample medium;  $K_s$ ), between sampling and measurement ( $K_d$ ), and during counting ( $K_m$ ):

$$a = \frac{n_n}{t_m} \cdot w = \frac{n_n}{t_m} \cdot \frac{1}{\epsilon \cdot p_\gamma \cdot q \cdot K_s \cdot K_d \cdot K_m}$$

When a FEP in the background is present at the energy of interest, its contribution is subtracted from  $n_n$ .  $\epsilon$  is obtained through an empirical efficiency calibration, using a radioactive source with the same geometry properties and approximately the same density and composition as the sample, so self-attenuation correction is neglected, but true coincidence summing correction is considered when the nuclides of interest have a cascade of successive photon emission.

## Evaluations (cont.)

### UNCERTAINTY EVALUATION

Sampling uncertainty was estimated empirically through parallel sample collections performed at the same location. Empirical estimation of the analytical (i.e. measurement) uncertainty was carried out by using replicates of gamma spectrometric measurements of the same samples. To perform these calculations, guide [1] was followed. Additionally, the analytical uncertainty was estimated by combining the uncertainties of the quantities upon which the measurand depends, according to an appropriate model. As such modelling approaches, both uncertainty propagation (i.e. GUM uncertainty framework) and stochastic approach (i.e. Monte Carlo Method-based propagation of distributions) were used. The formulae used for the calculations are presented in [2].

### DETERMINATION OF CHARACTERISTIC LIMITS

Characteristic limits, such as the decision threshold and detection limit were determined by applying [3], both uncertainty propagation-based and stochastic (Monte Carlo Method-based; hereinafter referred to as MCM-based) approaches.

## Results

The results are illustrated through the isotope of <sup>7</sup>Be, since this radionuclide is permanently present in radioactive deposition.

### MEASURED ACTIVITY CONCENTRATIONS

Fig 1. shows the results of the comparative analysis of samples taken at the same location over the same period. Although the average values show a good agreement, there are noticeable differences in some months, which justify quantifying and accounting for sampling uncertainty.

### SAMPLING AND ANALYTICAL UNCERTAINTY

It was shown (see Table 2) that sampling uncertainties are in most cases substantially larger than the empirically estimated analytical uncertainty, and their contribution dominates the overall uncertainty. This type of uncertainty can be reduced in statistical term by increasing the number of sampling replicates.

Table 2: Sampling and analytical uncertainties

Uncertainty	Typical relative uncertainty range (%)		Typical relative contribution to the overall uncertainty (%)
	Min	Max	
Sampling	0.13	79.5	81.8
Analytical	0.36	4.61	18.2

Table 3 shows the typical values of the analytical uncertainty estimated by the modelling approach using uncertainty propagation and that of the input quantities. The analytical uncertainty estimated that way typically exceeded the uncertainty obtained empirically, while the relative value of the latter remained below 5.0%, the former had an average of about 20%, occasionally exceeding 50% for low-level activities.

Table 3: Typical values of analytical uncertainty and the quantities affecting it under routine conditions

Quantity	Typical relative uncertainty range (%)		Typical relative contribution to analytical uncertainty (%)	
	Min	Max	Min	Max
$n_n$	1.90	64.5	23.2	99.6
$t_m$	0.01	0.29	0.00	
$\epsilon$	2.50	3.09	0.22	57.9
$p_\gamma$	0.38		0.01	1.08
$q$	1.67	2.89	0.20	17.8
$K_s$	0.01	0.03	0.00	
$K_d$	0.02	0.07	0.00	
$K_m$	0.00	0.01	0.00	
$a$	3.69	65.6	100	

The most dominant contributor to the analytical uncertainty in most cases is the net peak area, its uncertainty can typically be even above 50% when small number of counts are detected in case of low activities. By reducing its uncertainty the analytical uncertainty can effectively be decreased. As an example, by extending the counting time by a factor of 4, a reduction in analytical uncertainty of up to 40% was achieved.

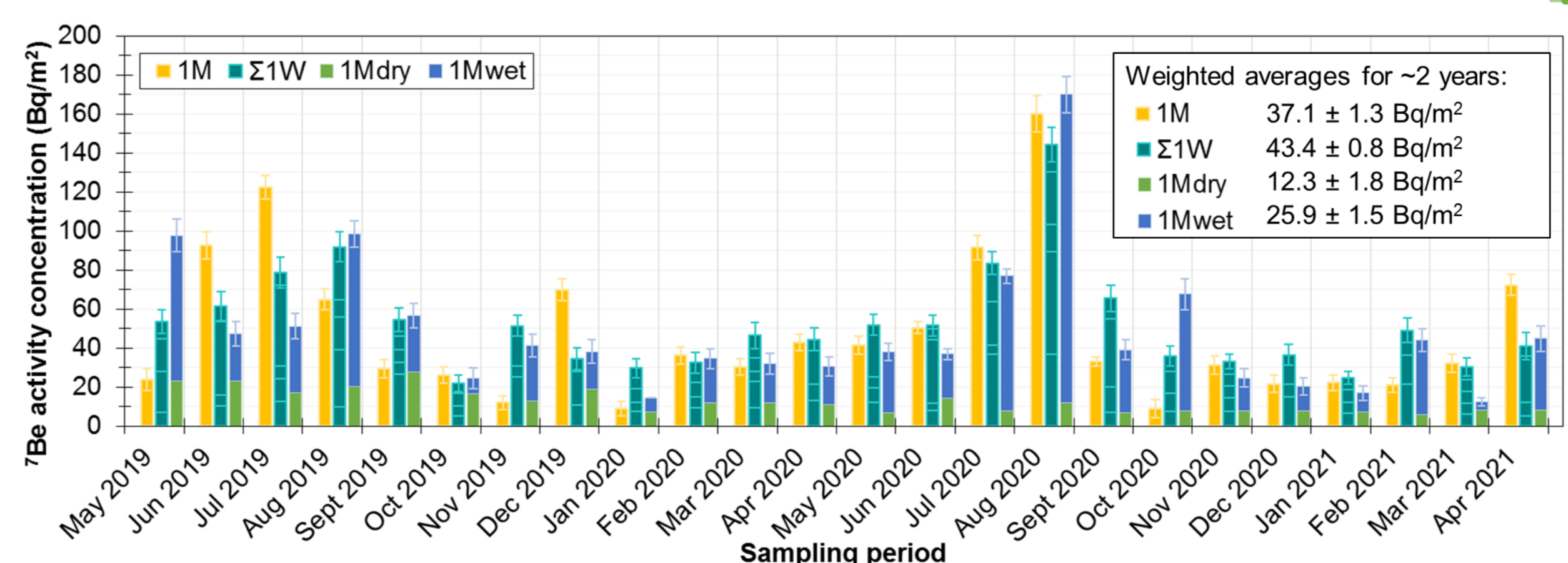


Figure 1: Comparison of <sup>7</sup>Be activity concentrations of deposition samples collected in parallel at Station 1, from a measurement series of ~2 years

## Results (cont.)

Since the distribution of the net peak area can be assumed as Gaussian and it is linearly related to the measurand, even its highest uncertainty and contribution that resulted in its sole domination of the analytical uncertainty, did not prevent the applicability of the uncertainty propagation-based evaluation of uncertainty and characteristic limits. Consideration of the maximum possible uncertainties for the quantities in a reciprocal function with the activity concentration ( $t_m$  and those incorporated in conversion factor  $w$ ) and increasing their relative contribution to be dominant over the net peak area still did not cause any non-compliance with the conditions for the uncertainty propagation and did not require the use of MCM-based propagation of distributions. The relative difference between the results obtained with uncertainty propagation and the stochastic method was less than 1% (see Fig 2).

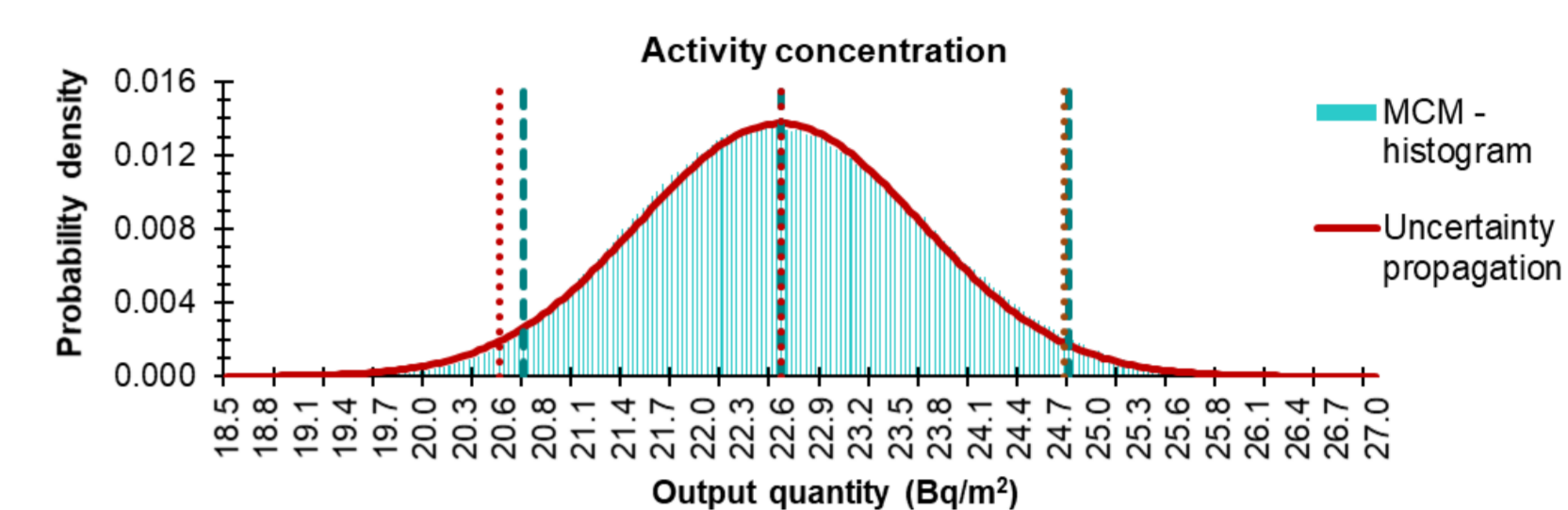


Figure 2: Probability density functions for the measurand obtained by uncertainty propagation and stochastic (MCM-based) approaches

### CHARACTERISTIC LIMITS

Typical detection limit for <sup>7</sup>Be, under routine measurement conditions, is 5.2-7.9 Bq/m<sup>2</sup>, depending on the used detector's efficiency. There are several solutions to reduce this value, e.g.

- by quadrupling the routine measurement time, characteristic limits could be reduced approximately to their half,
- by combining the samples taken in the same period by joint evaporation and measurement, an increase in the collecting surface corresponding to the sample number and an equivalent reduction in characteristic limits is achievable.

For a more detailed analysis of reduction methods, see [4].

### INPUT DATA FOR DEPOSITION MODEL VALIDATIONS

The rain sensor of the collector for separate sampling of wet and dry deposition is equipped with an event counter which gives the duration of the open state of the wet deposition collector. Precipitation amounts (with a 10-minute resolution) are measured by a precipitation meter located next to the sampling point. There occurred significant differences in the duration of rainfall events obtained by the two instruments (as in Fig 3), which were also contributed to by data losses of precipitation meter. Therefore, there were differences in the time integrated wet deposition activity concentration values calculated based on these, in contrast to the dry deposition, which showed good agreement (see Fig 4). The deposition velocity was given as the ratio of the time integrated dry deposition activity concentration and the air activity concentration. Due to the lack of the reliably available precipitation data, its calculation was performed using data from rainfall-free periods in order to neglect the potentially inaccurately estimated depletion of airborne activities due to wet deposition. Average deposition velocities of 8.7×10<sup>-4</sup> and 9.3×10<sup>-4</sup> m/s were determined which are in good agreement with the values for aerosols given in international recommendations.

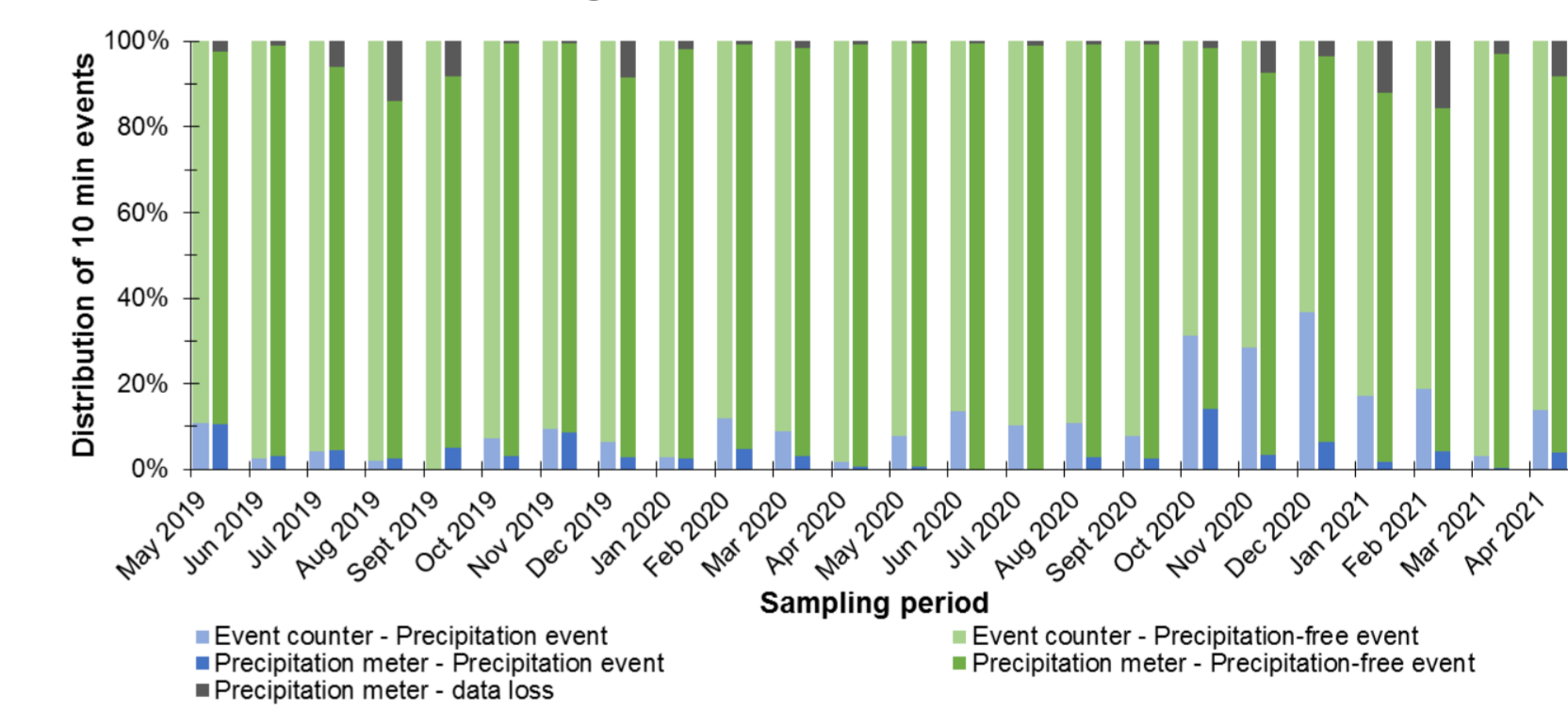


Figure 3: Percentage distribution of 10-minute resolution events (precipitation and precipitation-free) by month

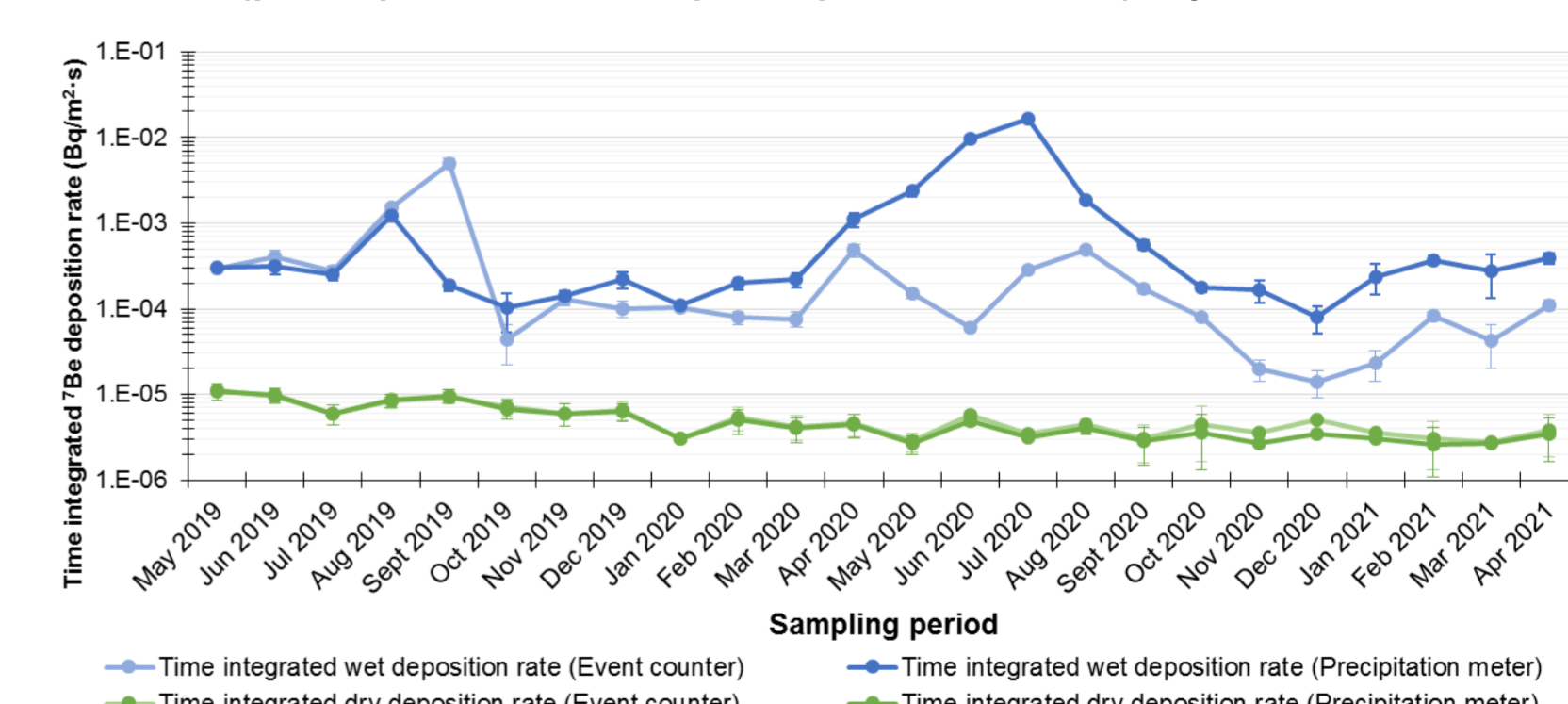


Figure 4: Time integrated dry and wet deposition rates derived from data of the event counter and precipitation rate

#### Notations:

- 1M: monthly combined (wet + dry) deposition;
- Σ1W: monthly summation of weekly combined (wet + dry) deposition;
- 1M<sub>dry</sub>: monthly dry deposition,
- 1M<sub>wet</sub>: monthly wet deposition;
- error bars indicate the analytical uncertainty (determined via uncertainty propagation).

## References

[1] Ramsey, M.H., Ellison, S.L.R., Roston, P. (eds.), Eurachem/EUROLAB/CITAC/Nordtest/AMC Guide, Measurement uncertainty arising from sampling: A guide to methods and approaches, Second edition, Eurachem (2019), ISBN 978-0-948926-35-8, <https://eurachem.org/>  
[2] Jakab, D., Pázmándi, T., Zagvyai, P., Effects of the uncertainty contributions on the methods used for measurement uncertainty evaluation, Applied Radiation and Isotopes, Vol. 173, 2021, 109704, <https://doi.org/10.1016/j.apradiso.2021.109704>  
[3] ISO 11929-3:2019, Determination of the characteristic limits (decision threshold, detection limit and limits of the coverage interval) for measurements of ionizing radiation -- Fundamentals and application  
[4] Jakab, D., Endrődi, Zs., Pázmándi, T., Zagvyai, P., Methods for improving the detection capabilities of environmental radioactive measurements in the light of increased atmospheric radioactivity levels in 2020, IRPA15, Seoul, South Korea, 18.01-05.02.2021