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INPUT VALUES FOR MODEL VALIDATION OF DRY AND WET DEPOSITION MODELS BASED ON THE ENVIRONMENTAL MEASUREMENTS AFTER THE RU-106 RELEASE IN THE FALL OF 2017

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Abstract: Anthropogenic ^{106}Ru has been detected in the environment over the period from late September to early October 2017 by several European environmental radiological monitoring networks. The temporal distribution and spatial localization of the contaminated plume was studied with backward trajectory simulations, which were performed with the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) developed by the National Oceanic and Atmospheric Administration (NOAA). The transport and dispersion of air parcels was studied to interpret the ^{106}Ru measurements at ground-level and also to locate the possible origin region of the contaminated air masses. On the basis of the inverse dispersion modelling, the travel time from the release at the source location and residence time of the contaminated plume over a certain area were also estimated.
Following the release of ^{106}Ru to the atmosphere, several measurements of various environmental compartments (airborne particulates, deposition, terrestrial indicators) were performed both on a national and international basis. Based on the ^{106}Ru deposition measured in the environment, assessment of the values of dry deposition velocity and scavenging coefficient was performed with account taken of the occurrence, duration and intensity of precipitation during the radioactive plume residence. The assessed parameters of deposition and wash out were compared to the default values used by operational models. The comparison between the measured and calculated ^{106}Ru deposition provided input values, which can be used for the validation of the dry and wet deposition models.

Key words: *Ruthenium-106, dry and wet deposition model, deposition velocity, scavenging coefficient, model validation*

INTRODUCTION

^{106}Ru has been detected in the environment on a nearly continental scale in Europe in the fall of 2017. The isotope of artificial origin was detectable in various environmental media, measurements of the environmental compartments as airborne particulates, deposition and terrestrial indicators were performed both on a national and international basis. The results of these environmental measurements provided input data for the atmospheric transport simulations (e.g. Jakab et al., 2019) and validation of the operational dry and wet deposition models.

This work is aimed to provide input data for validation of the operational deposition models and investigate the accuracy of distinct deposition velocities as well as the linear and exponential factors in the scavenging coefficient with the comparison of the measured and simulated deposition values. In this paper, principal considerations regarding the applicability of measured values for modelling of dry and wet deposition and the assumptions for comparing those results with monitoring data are summarized.

METHODS

Environmental measurements

Environmental measurement results (^{106}Ru activity concentration in aerosol air filters and deposition samples) were used as input data provided by the environmental radiological monitoring system of the

KFKI Campus in Budapest (located at latitude, 47°29'20.89"N and longitude, 18°57'13.44"E). Meteorological variables (e.g. precipitation amount over a pre-defined time period) observed at the environmental sampling location were also considered. As it was discussed by Jakab et al. (2018), ^{106}Ru activities bonded to airborne particulates were determined based on the continuous sampling of aerosol particulates on glass-fiber filters at a height of 2 m above ground for a constant air flow rate between the range of 4.2 and 6.3 $m^3 \cdot h^{-1}$. For the determination of the deposited ^{106}Ru activities, combined sampling of wet and dry deposition was performed at a height of about 1.2 m above ground, in permanent collectors with a surface area of 0.2 m^2. Precipitation quantities were measured at a height of 1 m above ground. The sampling frequencies of the environmental measurements determined the temporal availability of measurement results. Frequency of air sampling varied from daily to weekly periods, whereas the duration of the deposition sampling ranged from 1 week- to about 1 month-long intervals. Local meteorological measurements provided 10 minute-summation of precipitation quantities with a resolution of 0.1 mm.

Dry and wet deposition models

To compare the measurement results with simulated data, calculations with deposition models used in atmospheric dispersion modelling software were performed for determination of the deposited activities due to dry and wet deposition mechanisms.

In the dry deposition model (Sehmel, 1980) the dry deposition is considered to be proportional to the local air activity concentration determined by the following expression:

$$D(x,y) = C(x,y,z=0) \cdot v_d \quad (1)$$

where D(x,y): is the activity concentration deposited on the ground surface at point (x,y) due to dry deposition ($Bq \cdot m^{-2}$),
C(x,y,z=0): is the time integral of the activity concentration in air at ground level at point (x,y) ($Bq \cdot s \cdot m^{-3}$),
v_d: is the deposition velocity ($m \cdot s^{-1}$).

The value of the deposition velocity is affected by the physical-chemical attributes of the airborne material, the underlying surface (effect of ground roughness) as well as the atmospheric conditions (in particular the effect of wind speed and atmospheric stability category). Operational atmospheric transport models generally use distinct deposition velocity values for noble gases, aerosol particles, organic and inorganic iodine. In certain models, different aerodynamic diameter ranges of the particles are also taken into account.

In the wet deposition model (Brenk and Vogt, 1981) wet deposition is estimated from the following formula:

$$W(x,y) = \int_0^{PBL} C(x,y,z)dz \cdot (1-\varepsilon_w) \cdot \lambda_w \quad (2)$$

where W(x,y): is the activity concentration deposited on the ground surface at point (x,y) due to wet deposition ($Bq \cdot m^{-2}$),
ε_w: is the wash out fraction (-), subject to continuous rainfall at a constant rate, defined as $\varepsilon_w = 1\text{-}e^{-\lambda w \cdot \Delta t}$, where Δt is the duration of rainfall (s),
λ_w: is the scavenging coefficient (s^{-1}), determined as $\lambda_w = \alpha \cdot I^{\beta}$, where I is the precipitation intensity ($mm \cdot h^{-1}$) and α is the linear ($h \cdot mm^{-1} \cdot s^{-1}$) and β is the exponential coefficient (-) of the scavenging function.

The wash out mechanism depends on the form as well as the intensity of precipitation and the physical-chemical attributes of the airborne material. The effects of the wet deposition removal processes through the washout (below cloud scavenging) and the rainout (in-cloud scavenging) may be considered with different scavenging coefficients. In this work no distinction was made and the scavenging coefficients for the two wet removal processes were taken with the same parameter value.

CALCULATIONS

As it was stated by Jakab et al. (2018), daily variation of activity levels indicated the presence of ^{106}Ru in ground level air from 30 September until the morning of 4 October 2017 in the Budapest region. In the examined situation the number of the environmental measurements and their temporal availability limited the time step considered in the simulation of deposition to one hour. For the calculation, total beta activity

concentrations in aerosol air filters (Table 1.) collected with a daily sampling frequency at Station 5 (of the radiological monitoring system of the KFKI Campus) were used, as these values were available with the most sufficient temporal resolution. The measured total beta activities, corresponding to the aggregate activity of the beta-emitter particles present in the samples, were corrected with the representative background total beta activity concentration at the given measurement station (1.5 ± 0.7 mBq·m^{-3}; due to the presence of natural beta-emitter components), thus estimating the additional activity due the presence of pure β-emitter ^{106}Ru and its short-lived descendant ^{106}Rh in ground level air. ^{106}Ru activity concentrations were taken to be constant in the hourly time steps over the given sampling period. Because of the high distance from the estimated release area (Jakab et al., 2019), time integrated activity concentrations were taken to be spatially constant as well, constant vertical distribution of the time integrated activity concentration was assumed below the planetary boundary layer (PBL). Default value of 500 m was used for the PBL height for D stability category defined by the Pasquill scheme (Jow et al., 1990).

Table 1. Measured activity concentrations in ground level air

Sampling start	Sampling end	Sampling duration (h)	Activity concentration (mBq·m^{-3})	Time integrated activity concentration (Bq·s·m^{-3})	Constant time integrated activity concentration in hourly time step (Bq·s·m^{-3})
30.09. 06:50	01.10. 06:44	23.9	7.8 ± 0.4	$6.7 \cdot 10^{2}$	$2.8 \cdot 10^{1}$
01.10. 06:44	02.10. 09:22	26.6	33.5 ± 1.9	$3.2 \cdot 10^{3}$	$1.2 \cdot 10^{2}$
02.10. 09:22	03.10. 09:25	24.0	15.5 ± 0.9	$1.3 \cdot 10^{3}$	$5.6 \cdot 10^{1}$
03.10. 09:25	04.10. 09:13	23.8	7.6 ± 0.4	$6.5 \cdot 10^{2}$	$2.7 \cdot 10^{1}$

Deposition of ^{106}Ru was calculated considering reference values of dry deposition velocities and factors of the scavenging coefficient for aerosol particles used in different operational models (Table 2.). The calculations were performed with the parameterizations used in the following models as examples:

- SINAC (Simulator Software for Interactive Modelling of Environmental Consequences of Nuclear Accidents) Gaussian puff model developed by the Hungarian Academy of Sciences Centre for Energy Research (Szántó et al., 2012),
- ldX Eulerian model developed by the IRSN, which is used for long-range transport modelling (the parameterization used in the model is described by Baklanov and Sørensen, 2001),
- NAME (Numerical Atmospheric-dispersion Modelling Environment) Lagrangian particle trajectory model developed by the UK Met Office (Maryon et al., 1991).

Table 2. Reference values of dry deposition velocity and factors of the scavenging coefficient for aerosol particles in the examined operational models

Parameter	SINAC	ldX	NAME
$v_{d,\,aerosol}$ (m·s^{-1})	$1.0 \cdot 10^{-3}$	$2.0 \cdot 10^{-3}$	$1.0 \cdot 10^{-3}$
α (h·mm^{-1}·s^{-1})	$1.0 \cdot 10^{-4}$	$5.0 \cdot 10^{-5}$	$8.4 \cdot 10^{-5}$
β (-)	$8.0 \cdot 10^{-1}$	$1.0 \cdot 10^{0}$	$7.9 \cdot 10^{-1}$

The dry deposition velocity was taken to be constant over the given sampling intervals. The scavenging coefficient and the wash out fraction was calculated for each rainfall event, and the precipitation intensities were determined based on the hourly summation of 10 minute precipitation amounts. Stand-alone records with precipitation intensities within the range of the measurement resolution of 0.1 mm were omitted, because of their low intensity and short duration, their removal effect considered to be implausible throughout the whole of the plume volume, thus their contribution to the removal assumed to be negligible. In this calculation, the source depletion of the radioactive plume due to wet deposition was considered for every hour when rainfall was measured. In these instances, the dry deposition was calculated from the residual air activity concentration depleted at ground level based on the wash out fraction (i.e. multiplication of the time integrated activity concentration with the weighting factor of $(1- \varepsilon_w)$). Source depletion due to dry deposition was not taken into account, as it was two orders of magnitude smaller than the wet deposition (see in section Results and Discussion) and its removal effect prevails in the ground level of the plume.

RESULTS AND DISCUSSION

Deposited activities (distinct values for dry and wet deposition) were calculated from the time integrated activity concentrations at ground level, derived from the total beta activity concentration measurements with the above described models and parameters. The results of these calculations are summarized in Table 3. The comparison of the calculated values and the data from the deposition measurements (from those two, one week-long sampling periods which cover the plume residence) can be seen in Table 4.

Table 3. Calculated values for dry deposition (D) over the sampling intervals and wet deposition (W) values for each rainfall event. Summation of precipitation intensities (I) and deposited activities for the time period of 30.09-02.10. and 02.10-04.10. is highlighted in bold font. Dashed lines show the filter change of the daily air sampling.

Time period	I	SINAC		ldX		NAME	
	($mm \cdot h^{-1}$)	D ($Bq \cdot m^{-2}$)	W ($Bq \cdot m^{-2}$)	D ($Bq \cdot m^{-2}$)	W ($Bq \cdot m^{-2}$)	D ($Bq \cdot m^{-2}$)	W ($Bq \cdot m^{-2}$)
30.09. 06:50-01.10. 06:44		0.67		1.3		0.67	
01.10. 06:44-20:00		1.6		3.2		1.6	
01.10. 20:00-21:00	0.2[a]	0.10	1.5	0.22	0.58	0.10	1.3
01.10. 21:00-22:00		0.12		0.24		0.12	
01.10. 22:00-23:00	0.3[a]	0.092	2.0	0.22	0.86	0.096	1.7
01.10. 23:00-02.10. 09:22		1.2		2.5		1.2	
30.09. 06:50-02.10. 09:22	**0.5**	**3.8**	**3.5**	**7.7**	**1.4**	**3.8**	**3.0**
02.10. 09:22-13:00		0.20		0.41		0.20	
02.10. 13:00-14:00	0.3[a]	0.043	0.93	0.10	0.40	0.044	0.81
02.10. 14:00-03.10. 09:25		1.1		2.2		1.1	
03.10. 09:25-15:00		0.15		0.31		0.15	
03.10. 15:00-16:00	0.6	0.017	0.72	0.044	0.37	0.018	0.63
03.10. 16:00-17:00	2.2	0.0071	1.3	0.025	1.0	0.0089	1.2
03.10. 17:00-18:00	0.8	0.015	0.85	0.041	0.47	0.016	0.75
03.10. 18:00-19:00	0.5	0.018	0.64	0.046	0.31	0.019	0.56
03.10. 19:00-20:00	0.8	0.015	0.85	0.041	0.47	0.016	0.7
03.10. 20:00-04.10. 08:00		0.33		0.65		0.33	
04.10. 08:00-09:00	2.5[a]	0.0061	1.3	0.022	1.1	0.0079	1.3
04.10. 09:00-10:00	2.9[a,b]	0.0051	1.4	0.019	1.2	0.0067	1.3
02.10. 09:22-04.10. 10:00	**10.6**	**1.9**	**8.0**	**3.9**	**5.3**	**1.9**	**7.3**

[a] Precipitation intensities, which deviate significantly from the daily summations of rainfall published in the daily reports of the Hungarian Meteorological Service (OMSZ) (OMSZ webpage, 2019).
[b] On 4 October the air sampling lasted until 9:13 but the rainfall happened prior to the filter change.

Table 4. Comparison of measured (combined sampling of dry and wet deposition with a one-week long sampling frequency) and computed deposition values (summation of calculated dry and wet deposition activity concentrations)

Time period	SINAC	ldX	NAME	Measured
		D+W ($Bq \cdot m^{-2}$)		
30.09-02.10.	7.3	9.1	6.8	<2.1
02.10-04.10.	9.9	9.2	9.2	11.3 ± 2.2

The average ratio of the deposited activities due to dry deposition to wet deposition mechanism (when dry and wet removal occurred simultaneously) for SINAC, IdX and NAME models was 2.6%, 13.9% and 3.1%, respectively. The IdX model considers double value for dry deposition velocity compared to the other models, this resulted in a smaller difference between the dry and wet deposition values. The choice of the dry deposition velocity was especially critical in the first period (30.09-02.10.) in which the dry deposition mechanism was more dominant than the wet deposition. The dry deposition is a linear function of the dry deposition velocity, thus the significant overestimation of the dry deposition during the first sampling period (30.09-02.10.) indicates that the true value of dry deposition velocity in the examined situation was even smaller than the default values used by SINAC and NAME (i.e. $<1.0 \cdot 10^{-3}$ $m \cdot s^{-1}$). Because of the functional particle size dependency of the dry deposition velocity, it thereby implies the deposition of aerosol particles with small aerodynamic diameter (≤ 1 μm). Consideration of presumably locally prevailing (as they significantly deviate from the national observations; OMSZ webpage, 2019) precipitation intensities in the modelling also resulted in an overestimation of the contribution of wet removal compared

to the measured deposition activities. Such overestimation occurs predominantly in the case of intermittent records of rainfall on the 1st and 2nd of October, when the rainfall events were associated with small precipitation intensities but they occurred when the maximum value of activity air concentration was measured (see in Table 1.).

The deposition activity concentrations calculated for the second sampling period (02.10-04.10.), when wet deposition dominated, agree within the margin of error (± 19.5%) with the measured values. However, it must be noted that the dependence of the raindrop-size distribution and the form of the precipitation on the scavenging coefficient were disregarded, thus for each precipitation events the same linear and exponential factors of the given models were used to calculate the scavenging coefficient. The variation of the linear factor of the scavenging coefficient results in a proportional alteration in the value of the scavenging coefficient itself. On the other hand, because of the exponential function, the scavenging coefficient and thereby the deposition output varies inversely with the alteration of the exponential factor in the case of light rainfalls with intensities lower than 1.0 $mm \cdot h^{-1}$, whereas in the case of heavier rainfalls the direction of the alterations is the same. Perturbation of the exponential variable to determine its optimal value showed that the usage of distinct parameter values would have been accurate for light rainfalls in the first sampling interval and heavier rain events in the second sampling period.

CONCLUSIONS

Model-to-data comparisons were performed to provide input data for validation of dry and wet deposition models based on the environmental measurements after the ^{106}Ru release in the fall of 2017. Example model validation calculations were performed based on the examined case, through which the various limitations appearing in field measurements, such as the crude temporal resolution of environmental measurements and the reliability and spatial variability of meteorological observations were also showed. Parameterization for dry deposition and wet scavenging of a set of model configurations was evaluated. The results emphasized the sensitivity of the wet deposition model to the selection of the coefficients of the scavenging function as the deposition output can vary significantly depending on the parameter values chosen for different precipitation intensities.

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