STUDIES OF HT AND HTO BEHAVIOR IN THE VICINITY OF LONG-TERM EMISSION SOURCE: MODEL – EXPERIMENT INTERCOMPARISON

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ABSTRACT

There are presented in the research results of HT and HTO deposition and the model of HT (HTO) atmosphere concentration in the vicinity of a long-term HT and HTO emission source. Scavenging of HTO by precipitations was studied in 6 field experiments. The site of the scavenging experiments was around a 30 m emission source. The sampling arcs were chosen at 150-300 m from the base of the source to minimize dry deposition on the precipitation collectors. Data of the scavenging experiments are presented.
Kinetics of HT deposition to soil through its oxidation has been studied in laboratory conditions. The activity of HTO converted in the soil sample during a certain period of time was used to determine the oxidation rate. This rate varies, depending on the catalytic and/or biological activity of the soil material. Theoretical considerations have shown that the deposition rate can be expressed by the effective rate of oxidation, which formally corresponds to the first-order HT oxidation. HT deposition rates are reported.
The model, used for assessments, takes into account atmospheric dispersion, deposition and re-emission. The model of HTO wet deposition is taken into account kinetics of HTO exchange between vapor and liquid phase with parameters such as rain drop spectra, rain intensity, condensation-evaporation on drop's interface. Gauss type formulae for continuous emission source is used to calculate HTO atmospheric concentration. Meteorological data are used as input parameters for modeling.
The data presented on HT deposition to soil and HTO washout by precipitation is required for assessment of consequences of HT (HTO) release into the atmosphere.

INTRODUCTION

Currently hundreds of nuclear power units are being operated and tens more power units are being built. Besides, researches in the area of fusion science and technology continue. Several tens research facilities utilizing essential amounts of tritium are available in the world. So, wide spread of nuclear and tritium technologies render efforts to study the environmental behavior of tritium and its compounds.
To study tritium behavior in the vicinity of long-term emission source the following efforts were taken under the ISTC Project 654-99

- Modelling of tritium transport in the environmental compartments in the vicinity of the continuous emission source
- Verification of the parameters of the environmental tritium transport, including kinetics of tritium oxidation in soil and kinetics of tritium oxide washout with precipitations from the atmosphere under field conditions.

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Brief description of the TRIEF model

TRIEF model is designed for assessment purposes, in particular to assess consequences of tritium long-term atmospheric emissions. The model is described in details in (1).
Atmospheric dispersion during the emission from the primary source is calculated by Gauss model. In case of HT emission from the primary source the concentration of HTO in the atmosphere is determined by reemission from the soil. The model assumes that deposition and evaporation from the surface of the soil is going on evenly all over the area. All HT oxidation takes place in the surface layer of the soil. The secondary source in the model (reemission) is a multitude of elementary sources, each of which is considered to be point. There are two approaches to model reemission in the TRIEF model, they are Gauss and Lagrange approaches. It is assumed that HTO content in soil moisture is formed due to rain precipitation, condensation of atmospheric moisture and spring snow melting. The content of HTO in plants' tissue free water is estimated as a combination of HTO in soil and atmospheric moisture. The content of tritium in organic matter of plants is calculated based on HTO content in plants' tissue free water.
Modeling of HTO transport in aquifer and contamination of water wells is conducted using real geological-hydrological area structure. Infiltration, rivers and water wells are taken into account. Filtration problem is solved as a stationary one.
To validate the TRIEF model two test data sets have been used. The first test data set was developed using Russian data of long-term monitoring the source of atmospheric tritium release. The second test data set was developed on French data for long-term emission source. Intercomparison of modelling results and observations was carried out in frame of the IAEA's Research Programme BIOMASS and demonstrated a good agreement for both data sets.

Study of the kinetics of HT oxidation in soil

It is known tritium oxidation takes place in the surface soil layer due to microbial activity (2). This process had been studied in field conditions elsewhere. Most of the results were received when tritium oxidation was governed by the HT diffusion in soil. The presented study is focused on evaluation of oxidation rates due to kinetics.
To minimize the diffusion a blowing of soil samples with air of constant tritium activity was used. The samples to be studied were sampled from the surface soil layer (0 - 5 cm), separated of plant roots and stored in a refrigerator prior to the run. The soil sample of bulk density was placed in experimental cell. Ambient temperature, airflow, tritium activity in the airflow and the volume of the air were measured. The effective rate of tritium accumulation in the soil sample under investigation was calculated using the following formula:

$$K_{eff} = \frac{\left(C_1^{soil} - C_0^{soil}\right) \cdot m_{s.} \cdot \varphi_s}{V \cdot C^{air.} \cdot \tau} \qquad \text{(Eq.1)},$$

where K_{eff} - relative rate of tritium deposition to soil, %/min; C_0^{soil}, C_1^{soil} - HTO concentration in soil water before and after the experiment, correspondingly Bq/g; m_s - sample mass; φ_s - relative content of water in soil, % weight; V - volume of the air passed through the air sample, l; C^{air} - tritium concentration in air flow at the cell input, Bq/l ; τ - run duration, min.
It was discovered, that activity accumulation follows to the linear law. HT oxidation rate in forestry soils is ~ 3 times greater than that of the cultivated soil (2,3). The average velocity of HT deposition is ≈ 0,08 cm/sec (4). This complies with the published values of deposition velocities 0,01 - 0,08 cm/sec (3). Decreasing of oxygen concentration down to 3,5% does not affect the activity of the soil. Thermal treatment of soil at +130°C reduces K_{eff} down to ≈ 10 times as compared with the initial one. Kinetics of HT oxidation is described formally by the first order reaction.

Washout of tritium oxide with rain from atmosphere

Washout of HTO with rain from atmosphere was not studied in details in field conditions in past. The only published field experiment studying the HTO exchange between the atmosphere and raindrops is presented in (5).
One of the main tasks under the ISTC Project 654 was to study the HTO washout with rain from the atmosphere and validation of the washout models. To this end, a number of field experiments were conducted to study the HTO washout. HTO was released in 6 filed experiments during the rain events during 1999-2001. The source of HTO release was at the height of ~30 m. Concurrently, rainwater was collected with samplers located at the distance of 150 m from the source. During the experiments, the air temperature, wind speed and direction and rain intensity were recorded (6).
The following two models were developed and validated (7): simple engineering model and complex exchange kinetics model. Fig. 1 presents observed and modeling results for engineering model and the vapor/raindrop exchange model. It can be seen that the "drop" model describes the experimental data "from the top" and simple model – those "from the bottom".

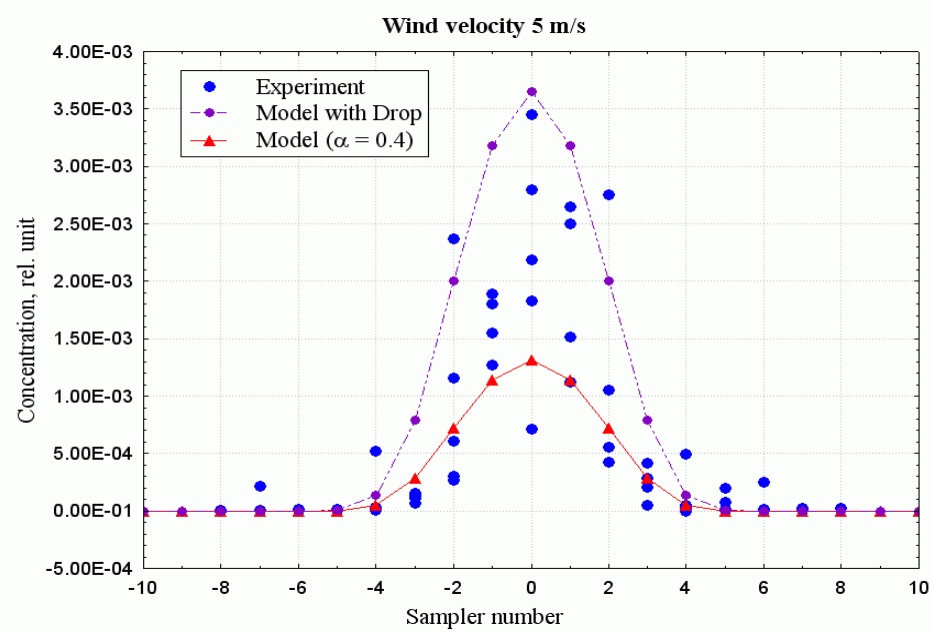

Fig.1 Comparison of observed and modeling results.

The α factor of the engineering model was estimated using minimization of experimental results in the linear and quadratic metrics (see Fig. 2). It is seen that the minimum in both the metrics corresponds to the α-factor value of 0.4.

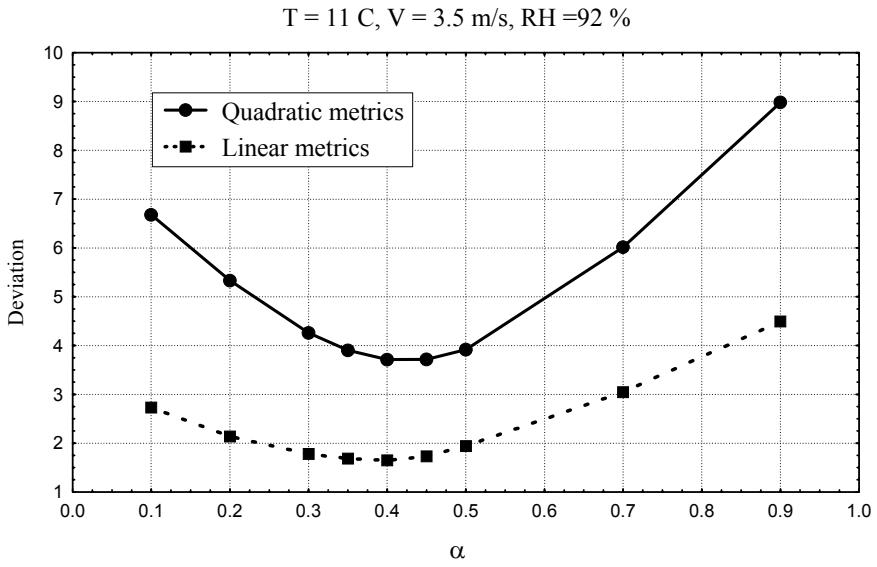

Fig. 2. The total deviation function of modelling results and observations in different metrics.

Washout rate Λ has been estimated using experimental results in a following way:

$$\Lambda = \frac{u}{2}\frac{J}{Q}\sum_{1}^{n-1}(C_i + C_{i+1})\cdot\Delta_{i,i+1} \qquad \text{(Eq.2)},$$

where
u - is the wind velocity, m/s;
J - is the precipitation intensity, m/s;
Q - is the HTO source intensity, Bq/s;
C_i - is the tritium concentration in rainwater in the i-th sampler, Bq/m^3;
$\Delta_{i,i+1}$ - is the arc length between the i-th and the i+1-th sampler, m.

The wind velocity for each of the experiments was taken as the average value. Rain intensity in all the experiments was assumed to be ~1 mm/hour. The release rate was estimated the average value during the entire experiment. The resulted average value of the washout coefficient is $\sim 14.5 \cdot 10^{-5}$.

CONCLUSION

The study of HT and HTO behavior in the vicinity of long-term emission source was carried out in frame of the ISTC #654 Project. The following results have been achieved.
The parameters of HT and HTO transport in the environment have been determined. They are:

- HT oxidation rate and HT oxidation kinetics by soil samples and
- HTO washout from atmosphere with rain.

The TRIEF model to assess long-term tritium releases was developed and validated in model-experiment exercise.

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