WASHOUT MODELS

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List of symbols

F_{hto}	HTO deposition rate	$(A.L^{-2}.T^{-1})$
Z	height above the ground	(L)
C	HTO concentration in air	$(A.L^{-3})$
C_a	tritium ground level air concentration	$(A.L^{-3})$
C_{rain}	HTO concentration in rain water	$(A.L^{-3})$
X	downwind distance	(L)
H	effective release height	(L)
χ_0	atmospheric tritium concentration at ground level	$(A.L^{-3})$
Q	the tritium activity rate	$(A.T^{-1})$
σ_{ys}	the standard deviation of distribution of concentration in the y direction (L)	
U_s	the mean wind speed	$(L..T^{-1})$
$\chi_{0,cal}$	the mean concentration at the ground-level	$(A.L^{-3})$
D	tritium deposition	(A.L-2)
A	tritium activity emitted	(A),
N	number of sectors of wind direction	-
X	distance from emitter	(L)
q_{imt}	frequency of precipitation (with i: sector of wind direction, m:wind velocity level, t:precipitation intensity level)	-
U_m	wind velocity	$(L.T^{-1})$
Λ	washout rate	(T^{-1})
s	proportionality constant	(L)
δ	precipitation intensity	$(L.T^{-1})$
σ_y	horizontal dispersion parameter	(L)
σ_z	vertical dispersion parameter	
u	mean wind velocity	$(L.T^{-1})$
h	emission height	(L)
y	vertical position	(L)
z	crosswind position	(L)
$y_{AB\|Bkg}$	spatially invariant background	$(A.A^{-1})$
n_0	total number of raindrops in a volumetric space	$(.L^{-3})$
n(a)	associated probability density function for raindrops of size a	(L^{-1})
$Flux_s$	rain flux follows directly the wet-deposition flux of pollutant approaching the surface at *(A. $L^{-2}.T^{-1}$).*	

1 Introduction

Washout of HTO by the precipitations is the principal processes resulting in a wet deposition. During precipitations, HTO that exist in atmosphere dissolves into falling raindrop and is removed from the atmosphere. It can also be scavenged by all atmospheric hydrometeors such as cloud and fog drops, rain and snow. HTO is consequently deposited to the ground. For a wet removal, three steps are necessary. HTO must first be brought into the presence of condensed water. Then, HTO must be scavenged by the hydrometeors, and finally it needs to be delivered to the ground. Figure 1 shows conceptual framework of wet deposition processes for aerosols and gas by Seinfeld and Pandis (Seinfeld & Pandis 2006). Washout is a reversible process. Once HTO scavenged, raindrops can be evaporated before deposition to the ground.

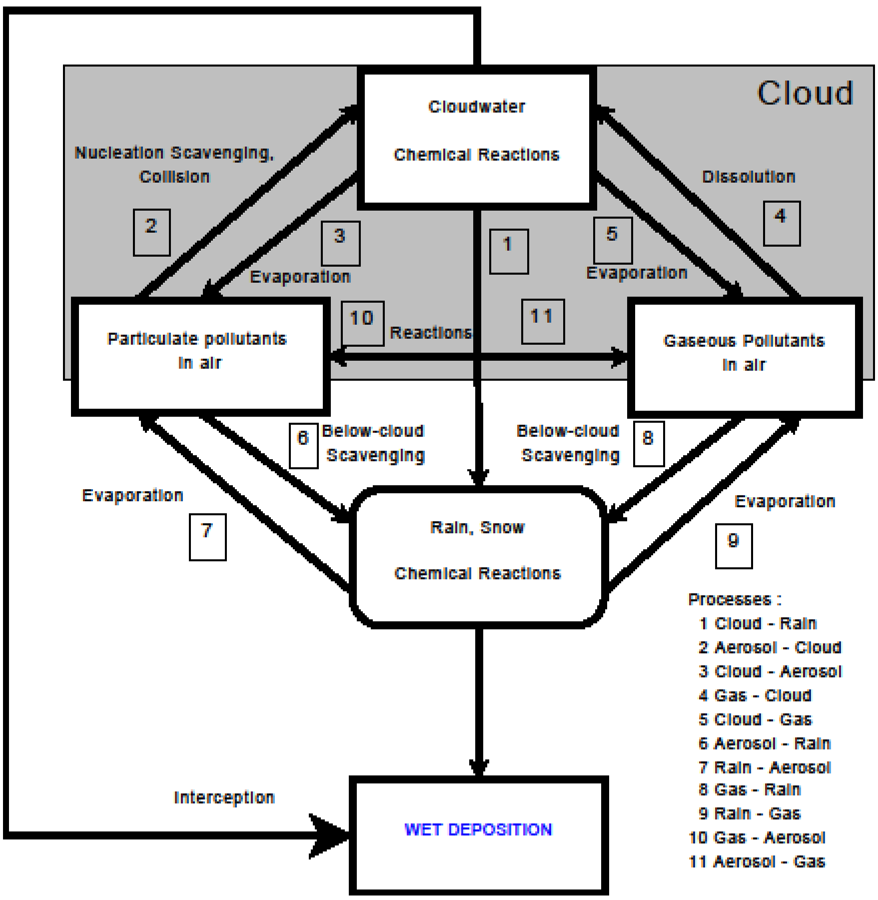

Figure 1 : Conceptual framework of wet deposition processes from Sienfeld & Pandis

2 Calculation of washout

Washout (Λ) is usually defined by the Engelmann (Engelmann 1968) equation:

$$F_{hto} = \Lambda \int C(z)dz$$

Where Λ is the washout rate (T^{-1}), F_{hto} is HTO deposition rate ($A.L^{-2}.T^{-1}$), C is the HTO concentration in air ($A.L^{-3}$), and z is the height above the ground (L). Tritium deposition rate can be expressed as

$$F_{hto} = C_p \times I_p$$

where is I_p the depth of falling precipitations collected over time ($L.T^{-1}$), and C_p is the tritium concentration in precipitation ($A.L^{-3}$).

$\int C(z)dz$ can be expressed by using a profile assumed to be Gaussian. Integral can be written:

$$\int C(z)dz = \left(\frac{\pi}{2}\right)^{0.5} C_a \sigma_z e^{\left(\frac{H_{eff}{}^2}{2\sigma_z^2}\right)}$$

where C_a is the ground level tritium air concentration ($A.L^{-3}$) at downwind distance x (L), σ_z is the dispersion parameter, and H the effective release height (L). It could also be expressed as

$$\int C(z)dz = \chi_0 \times H_{eff}$$

where χ_0 is the atmospheric tritium concentration at ground level ($A.L^{-3}$).

The effective height can be calculated by using the dispersion equation (Chamberlain & Eggleton 1964).

$$H_{eff} = \sum_s \frac{1}{\sqrt{2\pi}} \frac{Q}{\sigma_{ys} U_s} \frac{1}{\chi_{0,cal}}$$

where Q is the tritium activity rate ($A.T^{-1}$), σ_{ys} is the standard deviation of distribution of concentration in the y direction (L), U_s is the mean wind speed ($L.T^{-1}$), and $\chi_{0,cal}$ is the mean concentration ($A.L^{-3}$) calculated from the ground-level formula (IAEA (International Atomic Energy Agency) 1980). The subscript s refers to the atmospheric stability.

Therefore, washout can be expressed as

$$\Lambda = \frac{C_p \times I_p}{\chi_0 \times H_{eff}}$$

The washout rate can also be derived by field experiments measuring the depletion of the air concentration, χ as function of time.

$$\Lambda = \frac{1}{t} ln\left[\frac{\chi(t=0)}{\chi(t)}\right] = \frac{removal\ rate\ per\ unit\ volume\ and\ time}{HTO\ concentration\ per\ unit\ volume} \tag{2}$$

Λ	:	Washout rate	T^{-1}
t	:	Duration of the rainfall or the sampling period	T
χ	:	HTO conc. in the atmosphere	AL^{-3}

3 Washout rate from experimental data

Washout rate has been calculated from experimental data by several authors especially for rain but also for snow.

3.1 Washout for rain

In the state of Michigan (USA), tritium release from the Cook Nuclear Plant was studied and the tritium vapor was sampled in and analyzed from precipitation, air-conditioning condensate, surface and well water. The tritium deposition by precipitation scavenging as determined from the tritium activity collected in rain water samples. Samples of atmospheric water vapor were also collected to determine the ground-level tritium concentration required for the washout coefficient (Harris et al. 2008). Water vapor samples were collected far from the site to serve as a baseline for environmental tritium levels. **The washout rate varied from 2.4×10^{-5}-1.5×10^{-4} $s^{-1}$and the mean value of the 15 data is $(9.2\pm8.4)\times10^{-5}$ s^{-1}.**

In Fukui Prefecture (Japan), washout rate was computed from available data of tritium concentration in water vapor and rainwater for the years 1986-1992 in Tsuruga area (Hideki & Masaki 1997). Rain water was sampled and gathered monthly. The rainfall intensity observed is 2 $mm.h^{-1}$. Samples of water vapor at ground level were collected continuously and analyzed monthly. **The washout rate varied from 1.3×10^{-5}-1.6×10^{-4} s^{-1} and the mean value of the 29 data is $(7.3\pm4.1)\times10^{-5}$ s^{-1}.**

On the other part of Japan, at Tokai village (Ibaraki Prefecture), the yearly average tritium deposition was calculated and compared (Inoue et al. 1985) with observed data by using the following equation:

$$D = A\frac{N}{2\pi x}\sum q_{imt}\frac{\Lambda_t}{U_m}$$

with:

$$\Lambda = s\delta$$

Where D is the tritium deposition (A.L-2), A is the tritium activity emitted (A), N the number of sectors of wind direction, x the distance from emitter (L), q_{imt} is the frequency of precipitation (with i: sector of wind direction, m:wind velocity level, t:precipitation intensity level), U_m is the wind velocity ($L.T^{-1}$), Λ is the washout rate (T^{-1}), s is the proportionality constant (L^{-1}), and δ is the precipitation intensity ($L.T^{-1}$).

Comparison between observed and calculated tritium deposition leads to use a proportionality constant of 8.2×10^{-1} mm^{-1} thus $\Lambda=2.3\times10^{-4}$ s^{-1} for a rainfall of 1 $mm.h^{-1}$.

4.6×10^{-4} s^{-1} (Inoue et al. 1985)

In the same ways, rain water was collected and the tritium concentration determined at the Karlsruhe Nuclear Research Center (KFK) in the years 1982,1983 and 1984 (Papadopoulos et al. 1986). **The proportionality constant used was 9.4×10^{-2} mm^{-1} thus $\Lambda=2.6\times10^{-5}$ s^{-1} for a rainfall of 1 $mm.h^{-1}$.**

The weekly concentrations of tritium in air and in rain water were measured at the vicinity of the Savannah River Plant (Tadmor 1973) (South Carolina USA*)*. Therefore, experimental data and calculation of the height of the radioactive cloud for the meteorological conditions (81 m) lead to calculate the washout**. Authors concluded that a washout rate of 3.6×10^{-4} s^{-1} is a good approximation for a rainfall of 4 $mm.h^{-1}$.**

More recently, around the Paks nuclear power plant (Hungary), rainwater was collected and analyzed for tritium (Köllö et al. 2011). Based on emission and meteorological data, a reversible washout model (Hales 1972) was used to calculate the tritium concentrations, which were then compared with measured values. Washout rate were calculated only for samples above the background levels. **The washout rate varied from 8.2×10^{-6}-1.9×10^{-4} $s^{-1}$and the mean value is 5.5×10^{-5} s^{-1}.**

In order to study the kinetics and mechanisms of HTO exchange between vapor and drops, experiments using a unit for generating the drops of specific size, a flight gap with known HTO concentration and a drop collector were performed (Belovodski et al. 1997). **In laboratory conditions, for a radius of drops of 0.02 cm, the washout rate is is $1.4\times10^{-4}s^{-1}$.**

In order to study the HTO washout with rain from the atmosphere and thefollow-up development of the washout project, an experimental project (Project 654) was performed (Belovodski 2010). Tritium was released through a stack of 30 meters. Meteorological conditions as wind velocity, wind direction, air temperature, relative humidity were recorded by a weather station. During the experiments

, rain characteristics as rain intensity, drop size distribution and dependence of the falling rate of drops on their diameter were also recorded. The source parameters were measured and volumetric activity of HT and HTO are known. HTO activity was measured in rainwater of 10 samplers which were installed downwind in the ±45° sector. **The washout rate varied from 12.4×10^{-5}-18×10^{-4} $s^{-1}$and the mean value of the 4 data is $(14.5\pm2.1)\times10^{-5}$ s^{-1}.**

3.2 Washout for snow

Based on data collected following an accidental release of HTO to the atmosphere from a reactor at Chalk River Laboratories in January 1991, washout coefficient of HTO by falling snow was calculated (Davis 1997). Dispersion of the atmospheric plume was modeling by a simple Gaussian model in order to calculate the total amount of tritium deposited to the snowpark over the release period and compared them with observed values. **The washout rate for snow is $(2.1\pm1.0)\times10^{-5}$ s^{-1}.** This value is to compare with the scarce washout rate for snow. Semi-empirical value of **2.6×10^{-5} s^{-1}** is given for a snowfall rate of $1mm.g^{-1}$ (Konig et al. 1984).

3.3 Synthesis

The figure 1 shows the compilation of washout values from. These values are computed from experimental work or based on models taking account of data measurements. **The washout rate varied from 1.3×10^{-5}-3.6×10^{-4} $s^{-1}$and the mean value of 54 data is $(9.2\pm5.8)\times10^{-5}$ s^{-1}** according to tritium release parameters and meteorological conditions.

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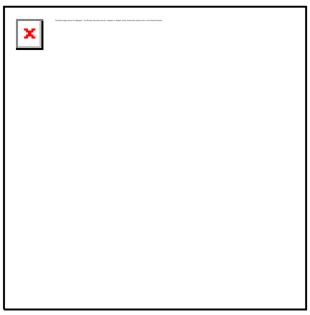


Figure 2: Bibliography review of washout rate based on experimental data

There are some type of liquid (rain, sleet), solid (Hail) or mixed precipitations leading to a wet deposition. Several classifications exist but considering little of existing experimental value, the simplest of classifications is proposed. According to American Meteorology Society the precipitation may be classified as following:

- **drizzle-fog**: drops are generally less than 0.5 mm in diameter, are very much more numerous;
- **light rain**: the rate of fall varying between a trace and 2.5 mm.h^{-1}, the maximum rate of fall being no more than 0.25 mm in six minutes;
- **moderate rain**: from 2.6 to 7.6 mm.h^{-1} , the maximum rate of fall being no more than 0.76 cm in six minutes;
- **heavy rain**: over 7.6 mm.h^{-1} or more than 0.76 mm in six minutes;
- **snow**: precipitation in the form of crystalline water ice of all size.

According to the experimental data, the Table 1 gives a average value of the washout rate. The database with the main washout rates given by the bibliograpy are shown in the table 2. Workgroup participants have to complete the database with their experimental data in order to improve it. Once this step done, average washout and other statistical data should be calculated for each type of precipitations.

Table 1: Proposed washout rate according to the type of precipitation for using in the simple and robust HTO models

Precipitation	Intensity (mm.h^{-1})	Washout (s^{-1})
drizzle-fog	all	no data > rain ?
light rain	$\leq$ 2.5 mm.h^{-1}	2.5×10^{-4}
moderate rain	2.6-7.6 mm.h^{-1}	3.6×10^{-4}
heavy rain	> 7.6 mm.h^{-1}	1.0×10^{-3}
snow	all	2.2×10^{-6}

4 HTO Models conception

4.1 Generalized equation calculation of washout rate

Generalization of washout rate according to rainfall intensity was proposed by several authors.

$$\Lambda = s \times (\delta)^b$$

Where s is the proportionality constant (L), δ precipitation intensity (L.T^{-1}).

The Figure 3 shows the washout rate according to the rainfall intensity. For a 1 mm.h^{-1}, the washout rates given by the equation above rainfall vary from 2.6×10^{-5}-2.27×10^{-4} s^{-1} that is a good agreement with washout rate calculated from experimental data. The recommended values in 2002 were $a=6.10^{5}$ and $b=0.73$ (Melintescu 2002) ??.

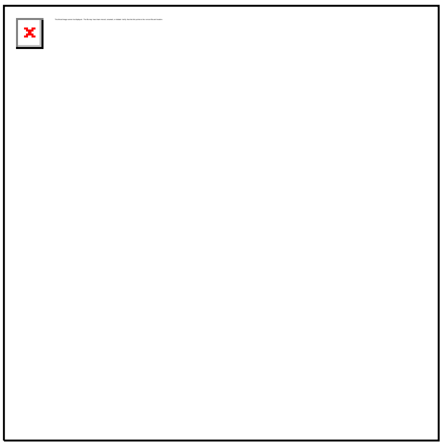


Figure 3: Generalized equation to calculate the washout rate according to several authors with δ in mm.h^{-1}

4.2 Modelling of the HTO concentration in rain water

Several model are used to computed wet deposion of tritiun. The simplest model like $C_{rain=}\alpha C_a$ to the sophistical models, like the Eulerian Stationary model which was developed by Atanassov (Atanassov & Galeriu 2007).

Comparison of the simplest model results with experimental data have shown that the value α =0.4 allows description of averaged experimental data in the best way. At the same time, for the wind velocities of 3 ms^{-1} the values of this coefficient are 0.35, for velocities of 6 m.s^{-1} it is 0.45 . REF

4.2.1 Hales model

In order to calculate wet removal of pollutants from Gaussian plumes, basic linear equations and computational approaches were proposed by Hales (Hales 2002). The approach takes the form of a set of analytical equations that correspond to five kinds of Gaussian plume formulations: standard bivariate-normal point-source plumes, line-source plumes, unrestricted instantaneous puffs, and point-source plumes and puffs that experience reflection from inversion layers aloft. These equations represent the concentration of scavenged pollutants in falling raindrops and are similar in complexity to their associated gas-phase plume equations. They are strictly linear, thus allowing superposition of wet-deposition contributions by multiple plumes. Numerical solution and analytical approximation are given but up to now, there is no direct application for tritium (HTO). Equation, for gaseous pollutant scavenging from point-source plumes, is based on the concept of gas scavenging model and was developed by Chamberlain and Eggleton (Chamberlain & Eggleton 1964).

$$\frac{dc(a,z)}{dz} = \frac{3K_y(a)}{v_z(a)a}\left[y_{AB} - H'c(a,z)\right]$$

Where C pollutant concentration with respect to height in a raindrop falling through a plume ($A.L^{-3}$), a is the raindrop's radius (L), H' is a solubility parameter ($L^{-3}.A^{-1}$), Y_{AB} is the mixing ratio of pollutant in air ($A.L^{-3}$), Vz is the raindrop's vertical velocity ($L.T^{-1}$), and k_y is an overall mass-transfer coefficient and can be estimated on the basis of physical properties.

The transfer of the pollutant to the drop from the gas phase is driven by the difference between the bulk gas concentration and the concentration at the drop surface.

$$c_{rain}(z) = \frac{4\pi n_0}{3\delta}\int_0^{\infty} a^3 n(a) v_z(a) c(a,z)\,da$$

where n_0 is the total number of raindrops in a volumetric space ($.L^{-3}$), n(a) is the associated probability density function for raindrops of size a (L^{-1}), and δ is the rain flux. The rain flux follows directly the wet-deposition flux of pollutant approaching the surface at z=s : $Flux_s = \delta C_{rain}(s)$ $(A.L^{-2}.T^{1})$.

For a plume with bivariate-normal distribution, the vertical distribution of gas-phase pollutant can be integrated in a straightforward (Hales 2002) and the distribution is:

$$y_{AB} = \frac{QF}{2\pi\sigma_y\sigma_z u}\exp\left(-\frac{y^2}{2\sigma_y^2}\right)\left\{\exp\left[-\frac{(z-h)^2}{2\sigma_z^2}\right] + \exp\left[\frac{(z+h)^2}{2\sigma_z^2}\right]\right\} + y_{AB|Bkg}$$

Where $\sigma_y\sigma_z$ are, respectively, the horizontal and vertical dispersion parameters (L), u is the mean wind velocity ($L.T^{-1}$), h is the emission height (L), y an z denote vertical and crosswind position (L), and $y_{AB|Bkg}$ is a spatially invariant background mixing ratio ($A.A^{-1}$).

Combining the both equations given the pollutant concentration in a raindrop and the equation given the mixing ratio of pollutant in air then integrating with respect to z:

$$c(a,z) = -\frac{\alpha}{2\sqrt{2\pi}H'}\exp\left(-\frac{y^2}{2\sigma_y^2}\right)\left\langle \exp\left(\frac{\sigma_z^2\zeta^2}{2}\right)\left\{\exp\left[-\zeta(z-h)\right]erfc(\beta_1) + \exp\left[-\zeta + h\right]erfc(\beta_2)\right\}\right\rangle + \frac{y_{AB|Bkg}}{H'}$$

Where:

$\alpha = (QF\zeta)/(\sigma_y u)$ *(dimensionless),*

$\beta_1 = (-\zeta\sigma_z^2 + z - h)/\sqrt{2}\sigma_z$ *(dimensionless),*

$\beta_2 = (-\zeta\sigma_z^2 + z + h) / \sqrt{2}\sigma_z$ *(dimensionless),*
$\zeta = \left[3K_y(a)H'\right] / \left[\mathrm{v}_\mathrm{z}(a)a\right]$ (L^{-1})

Others equations are proposed by Hales for computer using single-precision arithmetic and for large plume spreads with rapid mass-transfer rates. **To compute concentrations in bulk deposited rain water, integration must be made over the total drop size spectrum or on suitable approximation.**

4.2.2 Project #654 model

The study performed at the Russian Federal Nuclear Center by Alexey Golubev under the Project #654 of International Science and Technology Center funded by the U.S.Government and European Union proposed a model of HTO with rain. It based on the molecular flow of condensation which passes on to the liquid phase (J_+) and on molecular flow of evaporation which passes on to the gas phase (J_-). Variation of HTO concentration in drop is described by:

$$V_{drop}\frac{dC_{drop}(t)}{dt}=f\cdot S_{drop},$$

where

$$f=\alpha\cdot\sqrt{RT/2\pi\mu}\cdot(C_{air}-\gamma\cdot m\cdot C_{drop})$$

Vdrop is the drop volume, S_{drop} is the area of the drop surface, α is the condensation factor depending on the environmental conditions as well as on the water's aggregative state, Cair is HTO concentration in the surrounding air, γ is the H2O/HTO isotope separation factor at 20°C (γ=0.9), Cdrop is the HTO concentration in liquid (drop), being in the equilibrium with the vapor; m is the percentage of the saturated moisture in the air, T is the temperature, K; μ is the molecular mass.

The velocity of concentration variation depends on the difference of flow densities of HTO molecules directed into the drop and out of the drop. **The variation in HTO concentrations due to the change in the total number of molecules in the drop (condensation or evaporation) is taken into account by the third equation of the system.**

So, the full system of the equations describing the variations in the HTO concentration in the drop has the view:

$$\begin{cases}\dfrac{\rho_w}{6}\dfrac{dD^3}{dt}=D^2(J_+-J_-)\\ C_w\rho_W\dfrac{1}{6D^2}\dfrac{d(D^3T_{drop})}{dt}=\beta\left[T_{air}(L)-T_{drop}\right]+\lambda(J_+-J_-)\\ \dfrac{d\hat{n}^{HTO}}{dt}=\dfrac{6}{D(t)}\left(V_+^{HTO}\cdot n^{HTO}(L)-V_-^{HTO}\cdot\gamma\cdot\left(\dfrac{\tilde{n}^{H_2O}}{\hat{n}^{H_2O}}\right)\cdot\hat{n}^{HTO}\right)-\dfrac{n^*}{3D(t)}\cdot\dfrac{dD}{dt}\end{cases}\qquad(16)$$

where

$$J_+=\alpha\cdot\mu\cdot p(T_{air}(L))\cdot\psi(L)\Big/\sqrt{2\pi\cdot\mu\cdot RT_{air}(L)}$$

$$J_-=\alpha\cdot\mu\cdot p(T_{drop})\Big/\sqrt{2\pi\cdot\mu\cdot RT_{drop}}$$

$$C_{air}\equiv n^{HTO}$$

$$C_{drop}\equiv\hat{n}^{HTO}$$

$$n^* = (\hat{n}^{HTO} - \hat{n}^{H_2O}) = const$$
$$V_+^{HTO} = 4\alpha\sqrt{RT_{air}(L)/2\pi\cdot\mu^{HTO}}$$
$$V_-^{HTO} = 4\alpha\sqrt{RT_{drop}(L)/2\pi\cdot\mu^{HTO}}$$

where

D_{drop} is the drop diameter, α is the fraction of molecules, which pass on to the liquid phase (the probability of passing from the gas phase into the liquid phase) at encountering the drop surface and (1-α) is the share of reflected molecules, $p(T_{air}), p(T_{drop})$ are the pressure of H_2O saturated vapors at the temperatures of the atmosphere and the drop T_{air}, T_{drop} (taking into account the curvature of its surface), ψ is the relative humidity of the atmosphere, C_w, is the heat capcity of the liquid phase, ρ_w is the density of the liquid phase, λ is the condensation heat (evaporation), β is the heat transfer factor, $\tilde{n}$ relates to saturated vapors "produced" by the liquid phase and $\hat{n}$ relates to the liquid phase proper

For the full completion, this system is to be supplemented with the dependences $\psi = \psi(L)$ and $T_{air} = T_{air}(L)$ or some model allowing determining these dependences.

The specific activity of rainwater falling down onto the soil surface is described with the equation [Golubev A.V., Aleinicov A.Y., Golubeva V.N. et al. (2002)]:

$$C = \frac{\int_0^\infty C_{drop}\left(D_{drop}\right)\cdot\frac{\pi}{6}\cdot D_{drop}^3\cdot F'\left(D_{drop}\right)dD_{drop}}{\int_0^\infty \frac{\pi}{6}\cdot D_{drop}^3\cdot F'\left(D_{drop}\right)dD_{drop}}, \quad (17)$$

where C is the specific activity of rainwater; $C_{drop}\left(D_{drop}\right)$ is the specific activity of the rain drop of the diameter D_{drop}, falling onto the soil surface, $F'\left(D_{drop}\right)$ is the fraction of drops ranging in size from D_{drop} up to $(D_{drop}+dD_{drop})$. The function $F'\left(D_{drop}\right)$ represents the derivative of the empirical Best formula

$$F'\left(D_{drop}\right) = \left(\frac{D_{drop}}{A}\right)^n \cdot \frac{n}{D_{drop}} \cdot exp\left(-\left(\frac{D_{drop}}{A}\right)^n\right) \quad (18)$$

where n=2.25; A – is the parameter depending on the rain intensity [Belot Y. (2002)].

The vertical velocity of the rain falling on their sizes $V_\perp(D_{drop})$ is described by the following empirical dependence:

$$V_{\perp}\left(D_{drop}\right) = 4.874 \cdot D_{drop} \cdot \exp\left(-0.195 \cdot D_{drop}\right) \qquad (19)$$

Adding the horizontal component of the velocity $\vec{V}_{=} = \vec{V}_{=}(t)$, whose direction and value coincide with the instantaneous values of the wind velocity, we will obtain the equations for drop motion of D_{drop} size in the atmosphere. Integration of these equations determines the trajectory of the drop flight.

So, the presented model is the further development of the model (Golubev et al. 2002) and allows describing the exchange with HTO molecules between the stationary (in volume) drop and the atmospheric moisture.

This model was compared with experimental data. Gaussian model is used to calculate the atmospheric dispersion.

The washout model developed by VNIEF (Sarov) relates to the average-level-sophistication model. This model accurately describes experimental results. As against the simplest model (α=0.4=const), the developed model yields higher values of the washout coefficient in the vicinity of the source and at long distances away from it. The explicit introduction of wind velocity and a better choice of drop velocity formula explain the difference visa Belot model.

4.2.3 Eulerian model

To be improved : An attempt to generalize the washout modeling was done recently (ref????), after a collaboration between IFIN-HH and Bulgarian meteorology researchers. A numerical Eulerian model that describes washout independently of dispersion is developed.

$$\frac{dC}{dt} = \frac{6K}{\alpha \mathrm{d}}\left(\alpha\, C_g - C\right)$$

Here, following (Ogram 1985), the mass of gaseous and liquid HTO is expressed in term of concentration instead of mole-fraction t(T) is the time, C is the concentration of liquid phase

(M.L^{-3}), Cg is the concentration of the gas phase HTO in the drop's environment (M.L^{-3}), d is the drop diameter (L). (M.L-3). K is the Overall Mass-Transfer Coefficient and it is calculated by using semi-empirical expression, referred to as the Froessling.

$$K = k_y = \frac{D_g}{\mathrm{d}}\left(2 + 0.6\,\mathrm{Re}^{1/2}\,Sc^{1/3}\right) = \frac{D_g}{\mathrm{d}}\left[2 + 0.6\left(\frac{\mathrm{d}\,\mathrm{v}}{\nu}\right)^{1/2}\left(\frac{\nu}{D_g}\right)^{1/3}\right]$$

α is a dimensionless coefficient, constant with respect to C depending on temperature, Henry's low constant, universal gas constant and on density and molecular weight of water.

$$\alpha = \frac{T}{H}R\frac{\rho}{M}$$

The domain of the model is between the soil surface and the level H_{rain} from which the drops start their downfall. The uniform vertical grid is defined. At the top level H_{rain}=z(N) it assumes the liquid HTO into the raindrops is in equilibrium with the surrounding gaseous HTO CN=Cequil. The basic equation (1) is applied layer by layer downward the level H_{rain}, separately for all drop size intervals. If all parameters in equation are assumed constant within a grid layer, the analytic solution can be applied to a drop for the time it is passing through the grid layer.

Time t is determined for each drop diameter **d** in layer **I** by using an accepted formula for drops' downfall velocity.The concentration **C(d,i)**, calculated for this drop, after it has spent time **t** in the **i**-th layer, is used as initial condition for the next **i-1**-st layer. The calculations for the last **1**-st layer, the layer above the ground, give the spectral mass concentration of liquid HTO in raindrops at the surface **C(d,1)**.

$$C(t) = C_0\, e^{-\frac{6K}{\alpha \mathrm{d}}} + \alpha\, C_g\left[1 - e^{-\frac{6K}{\alpha \mathrm{d}}(t-t_0)}\right]$$

Sensitive analysis of eulerian model shows that the washout process is influenced most significantly by rainfall parameters and air temperature. Washout could varie from less of 1 % to 70 %. Atanassov conclude that the influence of the rain parameters and the temperature on the washout process is significant.

5 Sensitivity of models

In model, duration of raindrops throughout the plume has to be calculated and depend of the size of raindrops. Therefore, raindrop size distribution is most important parameter which can lead to high discrepancy in HTO drop concentration for same rainfall intensity.

5.1 Raindrop distribution

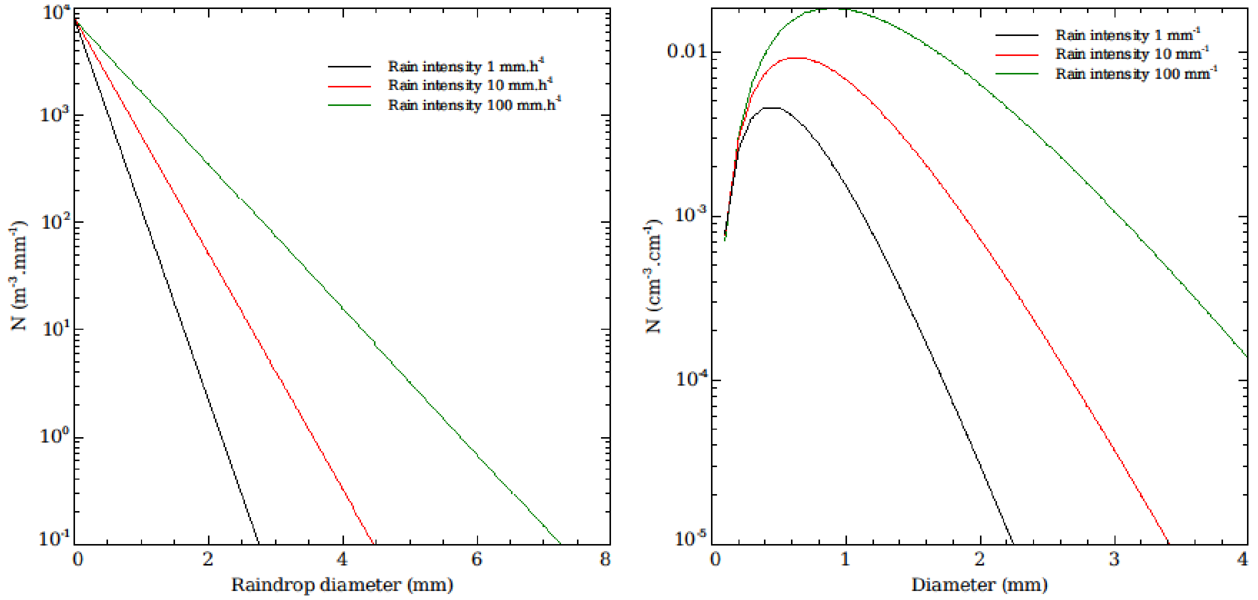
Add Best distribution used in the Golubev model

Raindrop size distributions are the end product of all of the cloud physical processes, cloud dynamical processes and interactions that affect the formation and growth of liquid precipitation. In addition, the raindrop distributions, once formed, can interact with the essential dynamics of the clouds through.

The number and size of raindrops within a unit volume is described by the number concentration,N(D) [number $m^{-3}.mm^{-1}$] also called the rainDrop Size Ditribution (DSD).

The raindrop distribution is usually described by a Gamma function (Ulbrich 1983) as the classical distribution of the Marshall-Palmer (Marshall & Palmer 1948). The Figure 4 hows the distribution of raindrops for three rainfall intensity: 1, 10 and 100 $mm.h^{-1}$. This distribution was developed using mid-latitude stratiform rain but leads to an overestimation of finest drops.

Paul T. Willis proposed some modifications to parameterizations of Marshall-Palmer distribution (Willis 1984) as shows the Figure 4. Feingold (Feingold & Levin 1986) described Log-normal distribution as raindrop distribution. Figure 5 shows log-normal distribution for three rainfall intensity. Table 3 shows equation for Marshall-Palmer, gamma and log-normal distributions.



Marshall-Palmer

Paul T. Willis

Figure 4: Marshall Palmer and gamma distribution function for modeling the drop size distribution

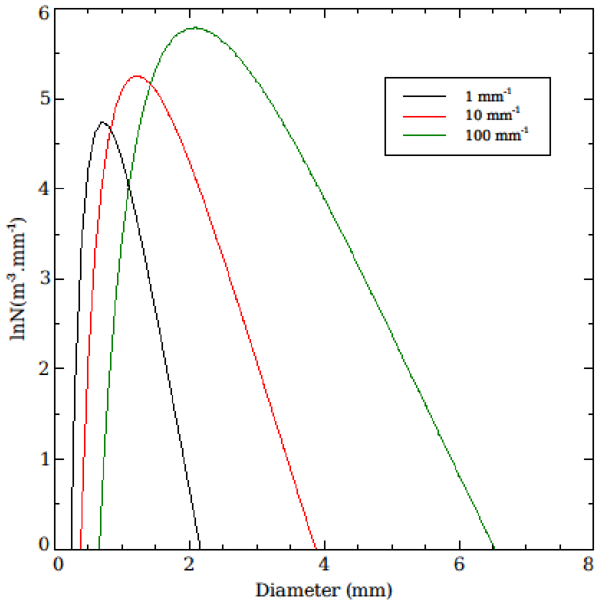


Figure 5: Lognormal distribution function for modelling the drop size distribution

Statistical raindrop distributions are well known and there are a lot studies but for HTO studies, we don't usualy have the description of rainfall and only intensity of rain is available.

Marshall-Palmer	Log-normal	Gamma

(Marshall & Palmer 1948)	(Feingold & Levin 1986)	(Willis 1984)
$N_D = N_0 e^{-\Lambda D}$	$N_{(D_p)} = \dfrac{N_D}{\sqrt{2\pi} \cdot D_p \cdot log\ \sigma_D} e^{-\frac{(logD_p - log\overline{D}p)^2}{2log^2\sigma_D}}$	$N_D = N_G D^{2.50} e^{-\Lambda D}$ cm $^{-4}$
$N_0 = 0.08$ cm $^{-4}$	$N = 172R^{0.22}$ m $^{-3}$	$N_G = \dfrac{6.36\times10^{-4} M}{D_o^4}\left(\dfrac{1}{D_0}\right)^{2.50}$
$\Lambda = 41 \cdot R^{-0.21}$ cm $^{-1}$	$D_r = 0.72R^{0.23}$ mm	$\Lambda = \dfrac{5.57}{D_0}$ cm $^{-1}$
	$\sigma = 1.43 - 3.0\times^{-4} R$	$D_0 = 0.157M^{0.168}$ cm
		$M = 0.062R^{0.913}$ g.m $^{-3}$

Table 2: Parameters for the Marshall-Palmer, Log-normal and Gamma raindrop size distribution

5.2 Raindrop diameter

Diameter of drop is often described as a function of the rain intensity. There exist several equations to calculate the representative diameter of raindrops. The most current form is $D_r = \alpha j^{\beta}$. From bibliographie α is range from 0.243 to 0.97 and β is range from 0.15 to 0.25. Figure 3 shows some parameterizations for the representative diameters of raindrops Dr as a fonction of rain intensity according to Andronache (Andronache 2004), Cerro (Cerro et al. 1997), Feingold (Feingold & Levin 1986), Loosmore (Loosmore & Cederwall 2004), Marshall-Palmer (Marshall & Palmer 1948), Pruppacher (Pruppacher & Klett 1998), and Underwood (Underwood 2001). The Figure 6 shows there is a range of factor 4 between diameters computed according to authors.

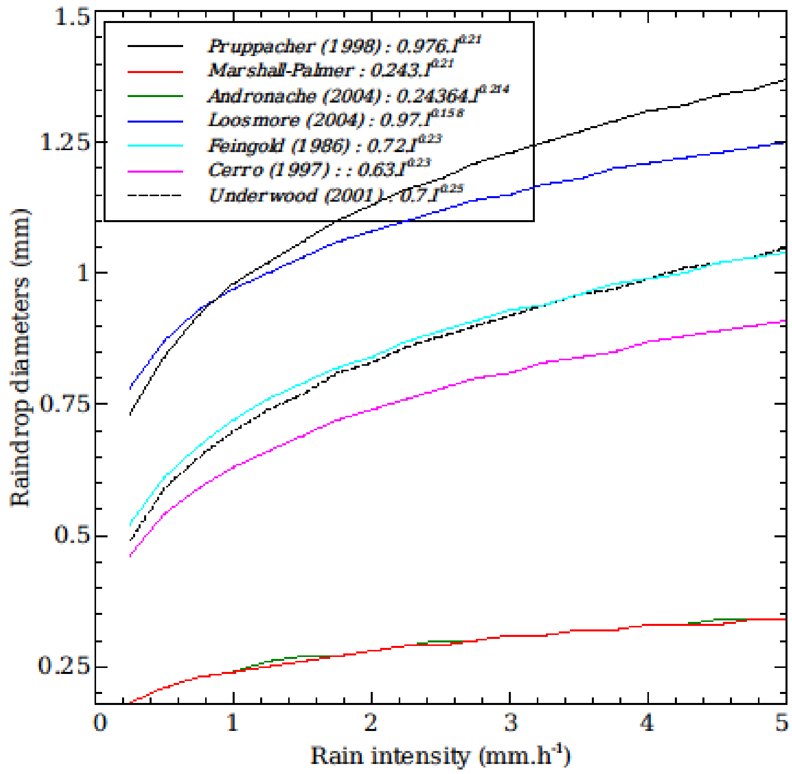


Figure 6: Evolution of the representative diameter as a function of the rain intensity according to different authors

5.3 Raindrops velocity

The raindrop velocity can be computed as a function of diameters. For the terminal velocity, the Stokes formula cannot be used dues to the size of falling raindrops which have a diameter bigger than 20 μm. A non-linear system has to be solved [?] (figure 5).

$$v = \sqrt{\frac{4gd_pC_c\rho_p}{3C_D\rho_{air}}} \tag{5}$$

g	:	Gravity constant	$m.s^{-2}$
d_p	:	Diameter	$mm.h^{-1}$
C_c	:	Cunningham correction factor	undimensionless
ρ	:	Drop or air density	$kg.m^{-3}$
d_p	:	Rain Intensity	$mm.h^{-1}$

The raindrops velocity can be estimated by using some parametrizations as shows it the Figure 7 according to Andronache (() (Andronache 2004), Loosmore (Loosmore & Cederwall 2004) and Seinfield (Seinfeld & Pandis 2006).

The Figure 8 shows the velocity of raindrop as function of rainfall intensity. The drop diameter was calculted with the formula given by loosmore (Loosmore & Cederwall 2004) ($D_r = 0.97 p^{0.158}$ where p is the rainfall intensity (mm.h^{-1}))

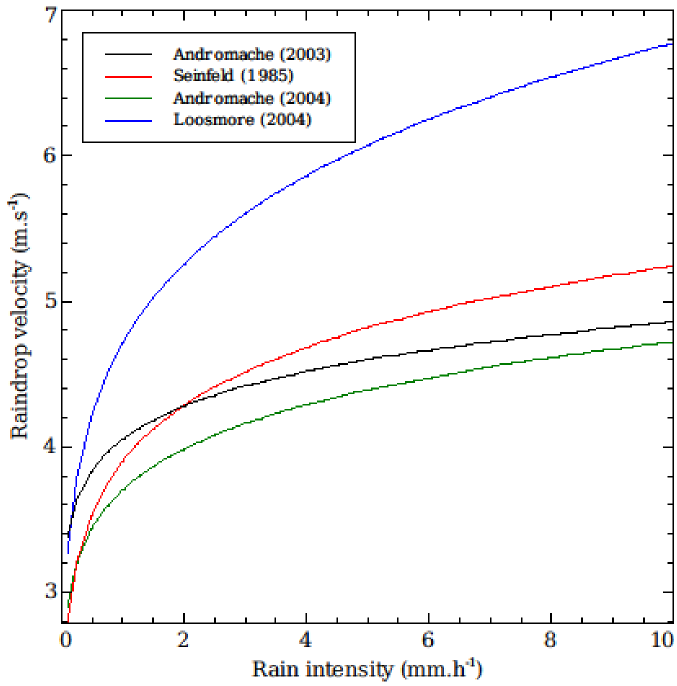


Figure 7: Evolution of the raindrop velocity as a function of the diameter according to different authors.

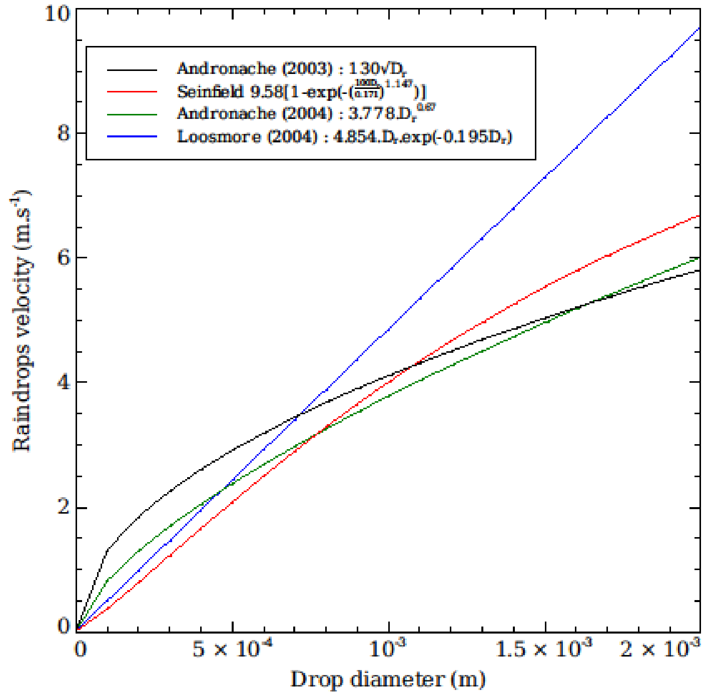


Figure 8:Evolution of the velocity as a function of the diameter

5.4 Sensitivity of models

Using the kinetic model from Alexey Golubev, let us consider the effect of the function of size distribution of drops on the exchange coefficient. The Figure 9 presents the computational dependences of the relative HTO content in precipitations for various functions of size distribution of drops.

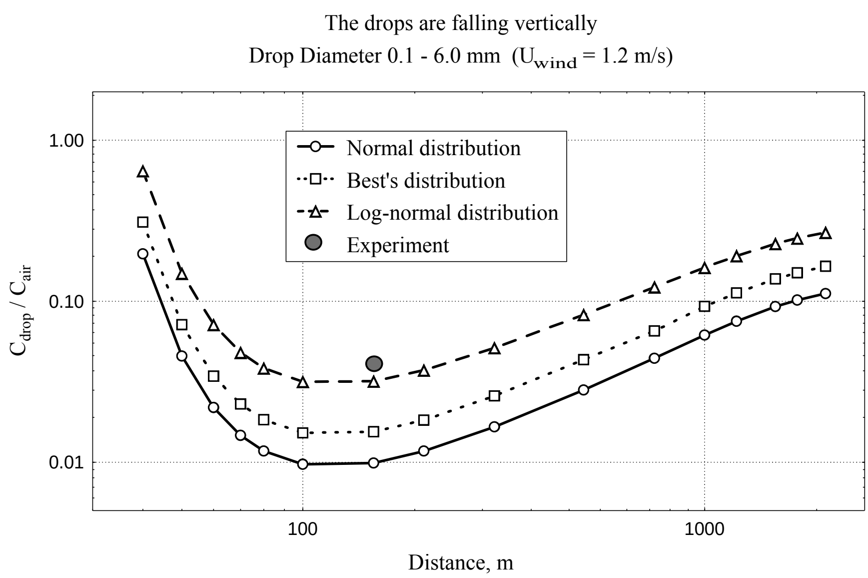


Figure 9:The computational dependences of the relative HTO content in precipitations for various functions of size distribution of drops

It is seen from the figure that the Best distribution and the uniform distribution yield close results; and the logarithmically-normal distribution of drops in size (the major mass of drops have the radius lower than 0.5mm) leads to higher values of the HTO concentration in precipitations. Especially great discrepancy of the curves is observed in the region of touching the ground with the emission tail, i.e., at our case this is 150m.

The studies with the model developed have shown that at the same precipitation intensity, the HTO content in rainwater essentially depends on the function of size distribution of raindrops. At that, the special attention should be drawn to the accuracy of describing the "tail" distribution corresponding to the largest drops since they make an essential contribution (from 20% up to 50%) into the precipitation intensity.

In the same way, a sensitivity analysis of rain characteristics on HTO concentrations in drops was presented in Tritium 2010 conference [Patryl et al, 2010]. CEA CERES code, which is the CEA reference computational tool for impact assessment, calculates the coefficient of transfer of HTO to drop then the specific activity of HTO in the drop leaving the plume. The average diameter in cm and the velocity of the drop in cm/s are given by extrapolation of experimental data of Chamberlain (Chamberlain & Eggleton 1964): $\tilde{r} = 0.037 LOG(I) + 0.0661$ and $\tilde{V} = 7000\,\tilde{r} - 12000\,\tilde{r}^{1.97}$ where I is the rain intensity in mm.h^{-1}.

$$\lambda_r = \frac{3DfC}{\beta r^2 \rho}$$

$$C_r = \frac{\beta}{C} \mathrm{X}(1 - e^{-\lambda_r t})$$

λ_r	:	Rate constant for uptake of HTO by drop	s^{-1}
C_r	:	HTO drop activity	Bq.kg^{-1}
D	:	Diffusivity of HTO in air	m^2.s^{-1}
f	:	Ventilation factor	-
C	:	Concentration of H_2O in air	kg.m^{-3}
X	:	Specific activity of water vapor in air	Bq.kg^{-1}
r	:	Radius of drop	mm
ρ	:	Density of drop	kg.m^{-3}
β	:	Ratio of vapour pressure of H_2O/HTO	-
t	:	Transit time in the plume by the raindrop	s

To estimate the variability of the drop size distribution on the rain concentration and washout rate, water activities for each drop are computed for every distribution by using the CERES code. Marshall-Palmer, Willis and Log-Normal distributions are computed for several rain intensities (from 1 to 20mm.h 1). The drop velocities are calculated for each diameter by using several formula:

Kessler (Ref. 6)	$v = 130\sqrt{D}$
Andronache (Ref. 7)	$v = 3.778\left(1000 \cdot D\right)^{0.67}$
Loosmore (Ref. 8)	$v = 4854 \cdot D \cdot \exp\left(-0.195 \cdot D\right)$
Seinfield (Ref. 9)	$v = 9.58\left[1 - \exp\left(-\left(\frac{100 \cdot D}{0.171}\right)^{1.147}\right)\right]$

where v is the falling velocity of raindrops ($L.T^{-1}$), D raindrops diameter (L) according to Kessler, Seinfield, Andronache and Loosmore.

The constant rate for uptake of HTO by each drop diameter is computed by using Chamberlain equation. Considering the rain drops spherical, calculation of the total water volume corresponding to each diameter allows estimating the concentration of rain water. The HTO air concentration considered is 1000 Bq.m-3 and it is assumed constant in the time. The plume layer crossing by drops is 200m. In this study, the plume is considered near the ground, and the loss of HTO by exchange between the drops and the air is not considered.

The highest HTO concentrations in rain are calculated with the Marshall-Palmer distribution by using Andronache formula to estimate the drop velocity (Fig.3) and the lower with the log-normal distribution by using the Loosmore formula velocity. This can be explaining by the number of fine raindrop overestimated by the Marshall-Palmer distribution and the HTO rate Λ_r which increases with the finest drop. The λ_r calculated for air temperature of 9°C is of 3.1×10^{-1}, 1.2×10^{-2} and 3.4×10^{-3} s^{-1} respectively for a 0.1, 1 and 3 mm of drop diameters. For a rain intensity of 1 mm.h-1, rain concentration ranges from 318 to 592 Bq.l-1 according to the DSD and velocity of drops, thus a factor 2 between the concentration minimum and maximum computed. The total surface of exchange between drop and air increases with the rain intensity. So, the quantity of HTO removed from air is highest but leads to the lower concentration in rain water by dilution. The HTO concentrations in rain computed with the CERES, which doesn’t take into account the drop distribution, are very close to those given by the Log-normal distribution.

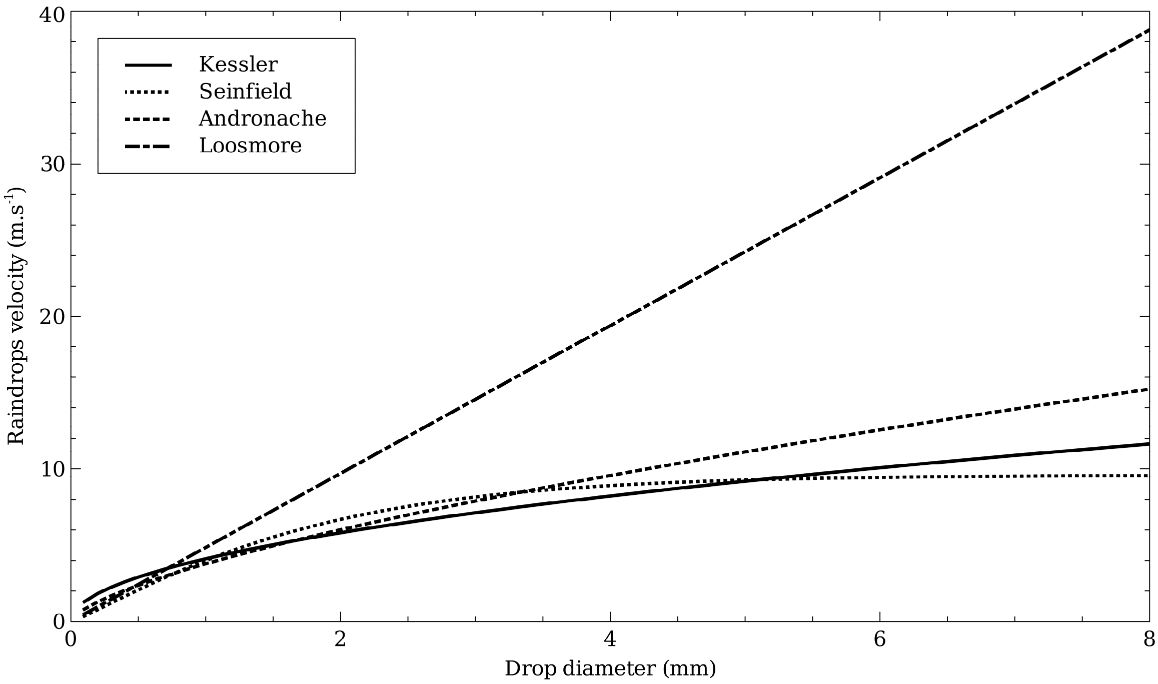
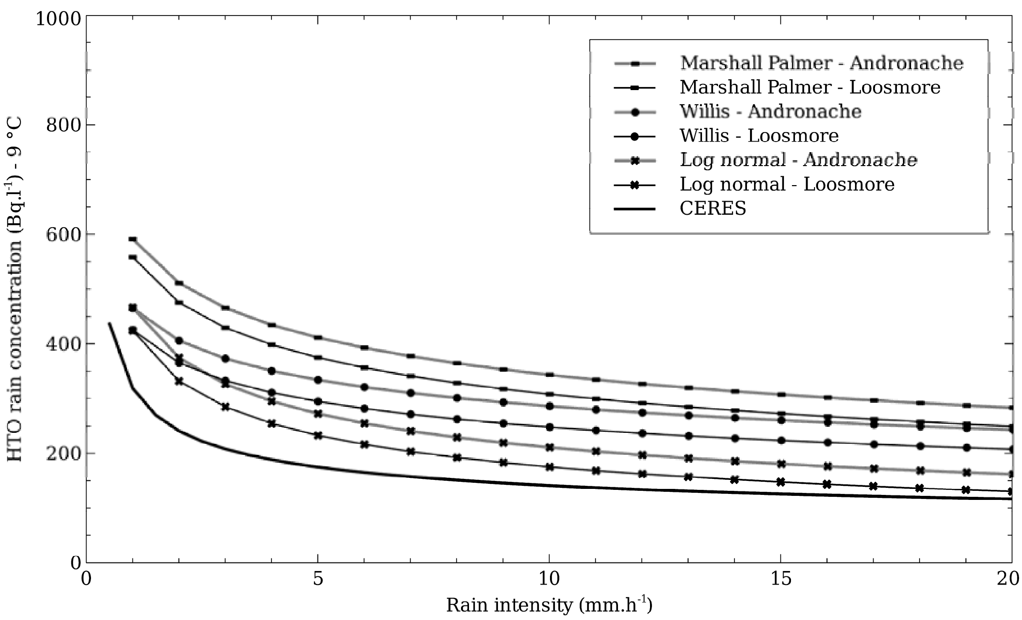


Figure 10: Comparison of HTO concentrations in the rain according to rain intensity between Marshall-Palmer, Willis and Log-normal drop distributions and the CEA code CERES – The calculations have been made for two air temperature: 9 and 20°C.

The rain concentration sensitivity to the calculations of the drop velocity is shown in the figure 4 for the Log-normal distribution. Rain concentrations have been computed also by using CERES to calculate the drop velocities. Average concentrations and standard deviations ranges from 673±30 to 241±23 Bq.l-1 respectively for rain intensity 1 and 20mm.h-1. The Loosmore formula which leads to have the highest drops velocities gives the lowest concentration.



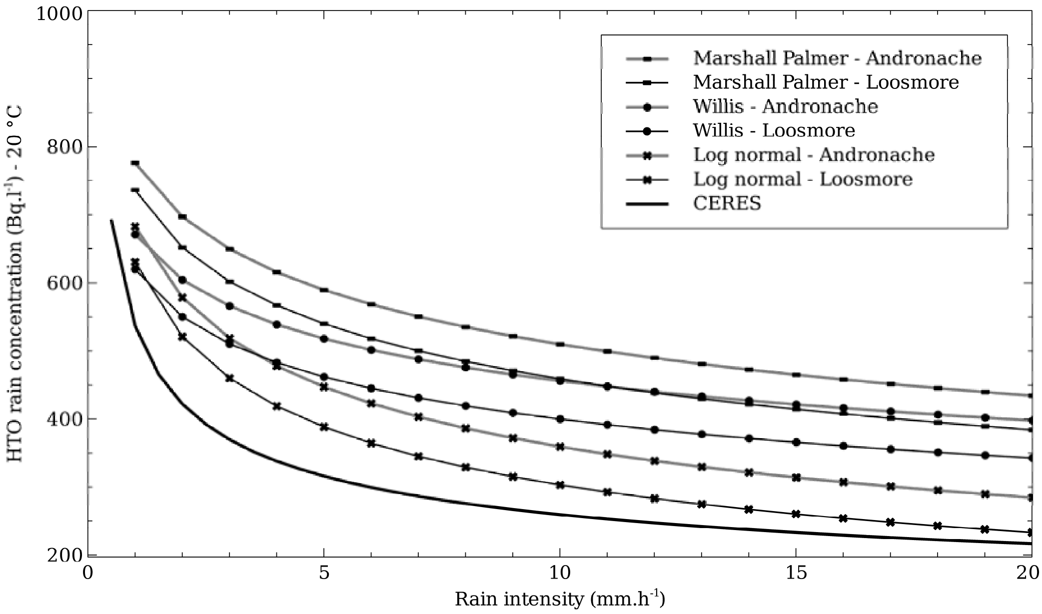


Figure 11: Sensibility of the raindrop velocity for the Log-normal distribution

The washout rate can be estimated by computing the amount of HTO removed per volume of air and with the equation 1. is the total volume of drop per volume of air, Nr the number of drop per radius r and Dr the drop diameter. At time t, the air concentration is equal to $C_{air}^{HTO}(t) = C_{air}^{HTO}(t=0) - C_{removed}^{HTO}$.

$$C_{removed}^{HTO} = \frac{\pi\beta X(t)\rho I}{6V_{tot}} \int_{r=0}^{\infty} \frac{N_r}{\nu_r} D_r^3 \cdot e^{-(\lambda_r t)}.$$

The Figure 12 shows the average washout computed with the equation 1 and by using the Kessler, Seinfield Andronache, Loosmore and Chamberlain velocity equations. The standard function with b=073 and b=1 and the washout rate calculated by CERES which doesn't take into account the rain drop distributions have been plotted. Standard deviations on each rain drop size distributions are plotted and represent the uncertainties due to the drop velocity. They can be estimated unless 20% with the Marshall-Palmer distribution and unless 15% for the two others.

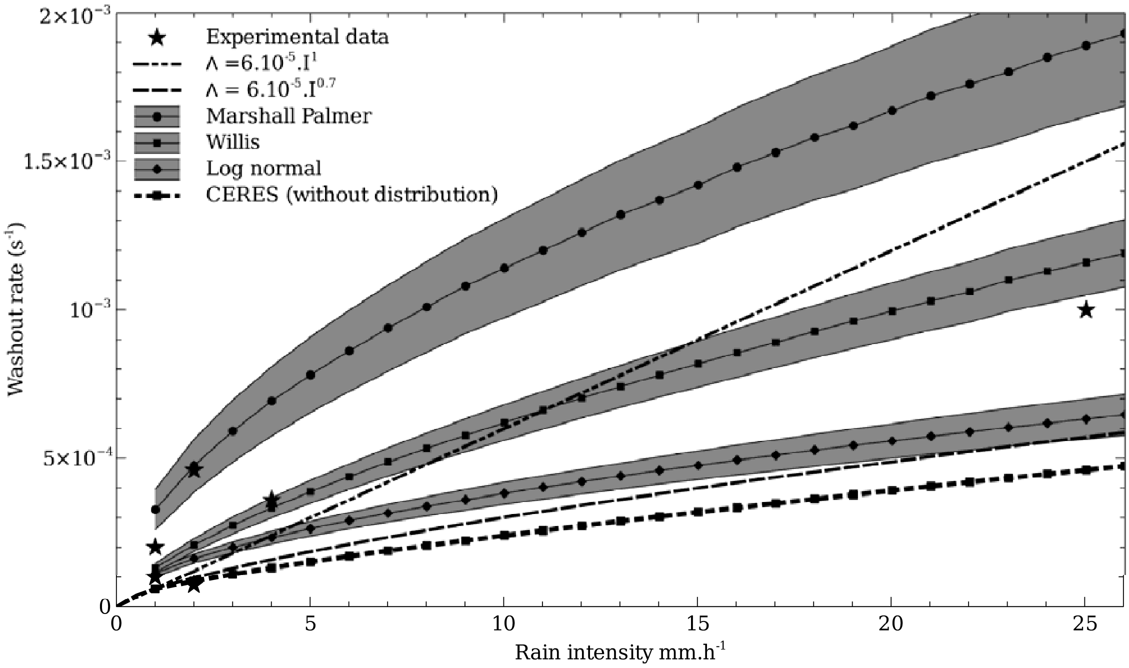


Figure 12: Average and standard deviation washout rate for the Marshall-Palmer, Willis and Log normal distributions computed by using Kessler, Seinfield, Andronache, Loosmore and Chamberlain drop velocity equations.

The washout rates calculated with the Marshall Palmer distribution are highest than Willis and Log normal distribution. A factor about 3 is observed between the Marshall Palmer and the Log normal distributions. For a rain of 1 mm.h^{-1}, the washout rate calculated ranges between 1×10^{-4} and 3×10^{-4} respectively for Willis and Marshall-Palmer distributions. Experimental data gives an average washout of 2.5×10-4 s^{-1}(Hideki & Masaki 1997) (Inoue et al. 1985) (Ogram 1985)for a light rain (<2.6 mm.h^{-1}), 3.6×10-4 s^{-1} (Ref. 14) for a moderate rain (2.6-7.6 mm.h^{-1}) and 1×10^{-3} s^{-1} (Ogram 1985)for a heavy rain (>7.6 mm.h^{-1}).

For light and moderate rain, the washout from experimental data is between those calculated with the Marshall-Palmer and Willis or Log normal distributions. Even if the Willis distribution seems to be the best distribution to estimate the washout rate, there is a good agreement between experiments and calculations with all formula used here. The evolution of the distribution during the rain is not taken into account but it certainly has an influence on the washout rate then on the HTO rain concentration. The number of small drops of rain induced an increase of washout and thus the concentration of rain. Considering the washout variability according to the rain drop distributions and methods to calculate it, the same uncertainties of about a factor 2 to 3 can be estimated on the wet deposition. Empirical equations that like used in CERES code seems to underestimate slightly the washout rate then the wet deposition. Conversely, they overestimate air concentrations in the air and thus inhaled and transcutaneous doses.

6 Conclusion

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