

Review on HTO washout

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Sommaire

- 1 Introduction
- 2 Scavenging of HTO
- 3 Washout rate and washout ratio
- 4 Rain characteristics
- 5 Models
- 6 Conclusions

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Context.

- To calculate HTO transfer from atmosphere to soil, dry and wet deposition should be calculated;
- Dry deposition
 - HT velocity to the soil surface is about $4 \cdot 10^{-5}$ - $4 \cdot 10^{-3} \text{ m.s}^{-1}$
 - HTO velocity to the soil surface is about 10^{-3} - 10^{-2} m.s^{-1}
 - depends of soil composition, soil humidity, landcover...
 - exchange velocity follow the Fick law;
 - [Foe88b, Foe88a, Gar80, OSB88, PCC⁺88, TWB88]
- Wet deposition during rain
 - HT solubility is very weak → deposition negligible
 - HTO solubility is important → HTO is exchanged with H_2O in the rain drop
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 - a washout rate or a washout coefficient



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Exchange of tritiated water vapour between a falling rain drop follow the steps :

- 1 HTO molecules migrate from the atmosphere to the surface of drop,
- 2 HTO molecule pass through the liquid-vapour interface,
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Main knowledge.

- **Total solubility is inadequate for HTO;**
- HTO exchange between atmosphere and drop is reversible;
- Under the plume, desorption can be possible;
- but never in equilibrium because the raindrop velocity is high.
- [CE64, DWW78, Hal72]



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Gas scavenging model :

$$\frac{\delta c(s; x, y, z)}{\delta z} = \frac{3v_d(s)}{v_t(s)s} [\chi(x, y, z) - H'c(s; x, y, z)]$$

- $c(s; x, y, z)$: concentration of HTO in raindrops (Bq.m^{-3});
- z : height of a drop above ground level (m);
- $v_d(s)$: HTO deposition velocity at the drop surface;
- $v_t(s)$: deposition velocity of the drop;
- s : radius (m);
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$$\Lambda = \frac{1}{t} \ln \left[\frac{\chi(t=0)}{\chi(t)} \right] = \frac{\text{removal rate per unit volume and time}}{\text{HTO concentration per unit volume}}$$

- is derived from relatively short measurement interval and single individual precipitation
- is functions of precipitation rate
- is apply preferably to individual scavenging conditions (drop-size distribution, different rain types, vertical variation)



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χ : HTO conc. in the atmosphere AL^{-3}

t : Time, T

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washout ratio.

$$\omega = \frac{L \int \int_0^{\infty} \pi R^2 E(a, R) N(R) u_t(R) \chi_{(x,y,z)} dR dz}{\int_0^{\infty} \chi_{(x,y,z)} dz}$$

- is multiple integral over a variety of different scavenging parameters
- most of the washout ratios has been derived from long measurement periods
- assure the influence of a large variety of scavenging mechanisms = relatively small range



ω : Washout ratio, dimensionless

E : Collection efficiency, dimensionless

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Washout rate.

- Used for a individual scavenging;
- Even if washout rate are determined as functions of drop size, drop size distribution, precipitation rate...;
- \Rightarrow Represents a space-averaged value and is derived from relatively short measurement interval;
- Could be considered as function of the downwind distance and source height;
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Washout ratio.

- Has a strongly averaging character because ω is a multiple integral;
- Vertical distribution of air concentration is considered as quasi-homogeneous;;
- multiple integral character leads to a significantly reduction of the variability.



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Washout ratio or rate

- ω or Λ can be used for either continuous or accidental releases;
- Nevertheless, Λ should be applied to short-period events (accidental release)
- ω should be used to long-term problems (routine releases)

Washout ratio or rate

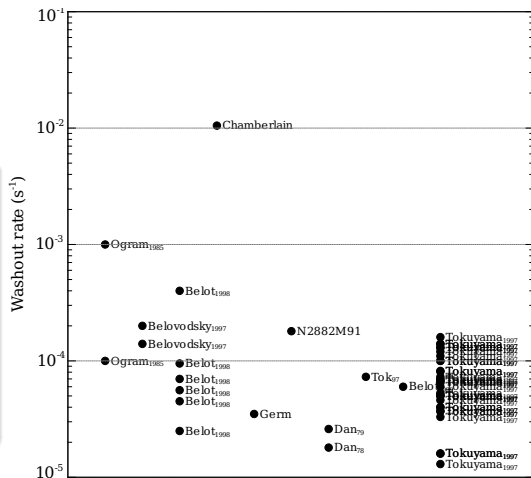
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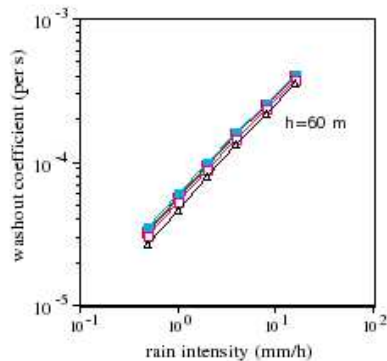
Washout rate in literature

- from 10^{-5} to 10^{-2} s^{-1}
- depends of rainfall intensity (Ogram, Belot,...)
- depends of the stack height (Belot)
- depends of raindrops radius (Chamberlain)
- monthly mean (Tokuyama)



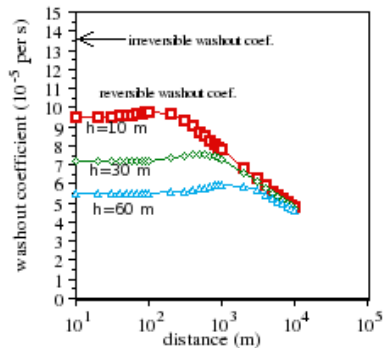
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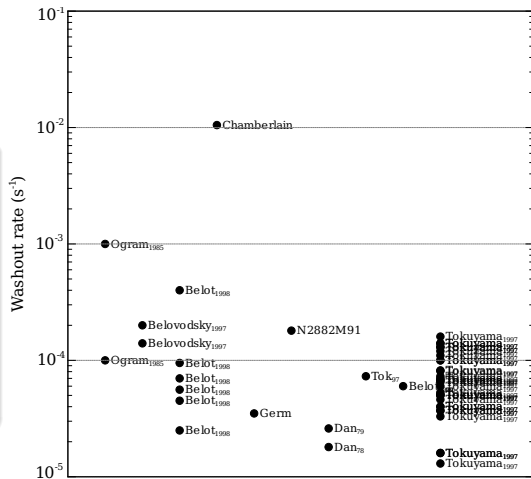
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Samples of equation

$$\Lambda = a \cdot (J)^b$$

- a and b are two empirical coefficient which depends on local conditions;
- According to Melintescu the recommended value are $a=6 \cdot 10^5$ and $b = 0.77$;
- In literature b ranges between 0.7 and 1.

Sommaire

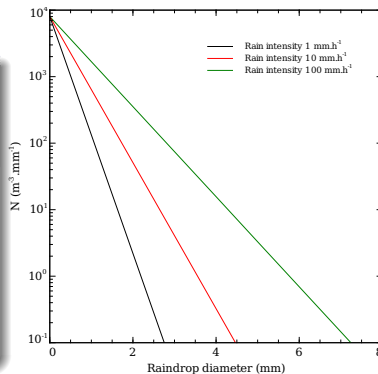
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General meaning

- Washout rate strongly depends of drop characteristics;
- HTO rain activity depends of duration of raindrops throughout the plume;
- thus raindrop velocity depends
 - Rainfall intensity
 - Raindrop size
 - Raindrop size distribution

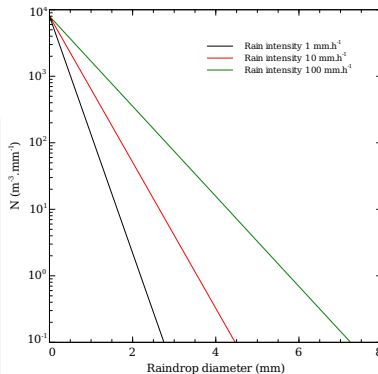
Raindrop distribution - Drop Size Distribution (DSD)

- can be described by several density functions;
- Marshall-Palmer;
- Gamma;
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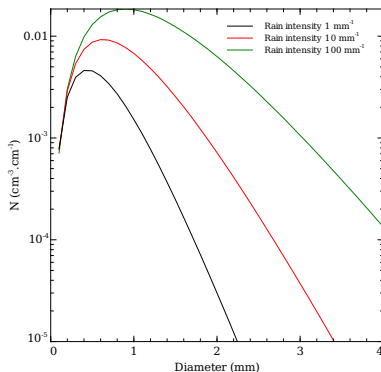
$$N_D = N_0 e^{-\Lambda D}$$

$$N_0 = 0.08 \text{ cm}^{-4}$$

$$\Lambda = 41 \cdot R^{-0.21} \text{ cm}^{-1}$$

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$$N(D_p) = \frac{N_D}{\sqrt{2\pi} \cdot D_p \cdot \log \sigma_D} e^{-\frac{(\log D_p - \log \bar{D}_p)^2}{2 \log^2 \sigma_D}}$$

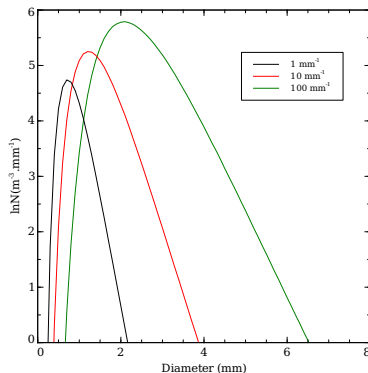
$$N = 172 R^{0.22} \text{ m}^{-3}$$

$$D_r = 0.72 R^{0.23} \text{ mm}$$

$$\sigma = 1.43 - 3.0 \times 10^{-4} R$$

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$$N_D = N_G D^{2.50} e^{-\Lambda D} \text{ cm}^{-4}$$

$$N_G = \frac{6.36 \times 10^{-4} M}{D_0^4} \left(\frac{1}{D_0} \right)^{2.50}$$

$$\Lambda = \frac{5.57}{D_0} \text{ cm}^{-1}$$

$$D_0 = 0.157 M^{0.168} \text{ cm}$$

$$M = 0.062 R^{0.913} \text{ g} \cdot \text{m}^{-3}$$

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$$D_r = \alpha \cdot J^\beta$$

α	: empirical coefficient	undimensionless
β	: empirical coefficient	undimensionless
J	: Rainfall intensity	$L.T^{-1}$

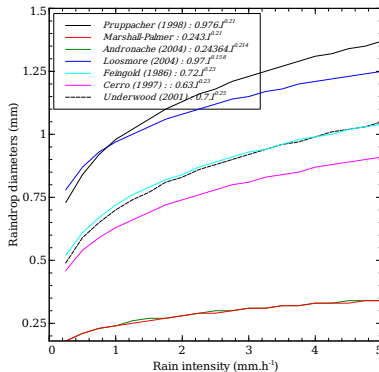
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Pruppacher and Klett	$0.96 \times J^{0.21}$	[PK98]
Marshall-Palmer	$0.243 \times J^{0.21}$	[MP48]
Andronache	$0.24364 \times J^{0.214}$	[And04]
Loosmore and Cederwall	$0.97 \times J^{0.158}$	[LC04]
Feingold and Levin	$0.72 \times J^{0.23}$	[FL86]
Cerro et al.	$0.630 \times J^{0.23}$	[CCBL97]
Underwood	$0.7 \times J^{0.25}$	[Und01]

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Raindrop velocity

- Stokes cannot used (diameters $> 20\mu m$)
- Several equations to computed raindrop velocity as function of diameter;
- Evolution of the raindrop velocity as function of diameter and rainfall intensity

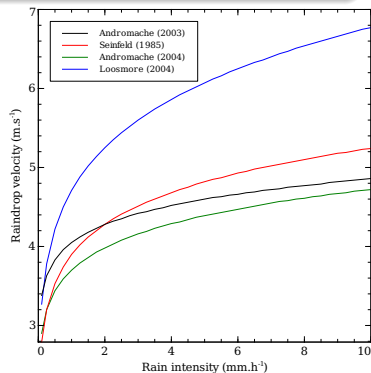
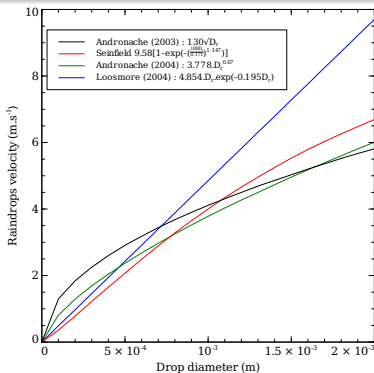
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- Several equations to computed raindrop velocity as function of diameter;
- Evolution of the raindrop velocity as function of diameter and rainfall intensity

Andronache	$130 \sqrt{D_r}$	[And03]
Seinfeld	$9.58 \left[1 - \exp \left(- \left(\frac{D_r}{0.171 \times 10^{-2}} \right)^{1.147} \right) \right]$	[Sei85]
Andronache	$3.778 \cdot D_r^{0.67}$	[And04]
Loosmore and Cederwall	$4.854 \cdot D_r \exp (-195 \times 10^{-3})$	[LC04]
Best	$V_t(s) = a \left\{ 1 - \exp \left[- \left(\frac{s}{b} \right)^n \right] \right\}$	
Chamberlain and Eggleton	$V_t(s) = 7000 \cdot s + 12000 \cdot s^{1.97}$ with $s = 0.037 \text{Log}(J) + 0.0661$	[Cha53]

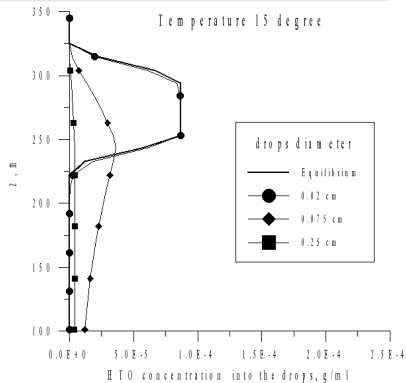
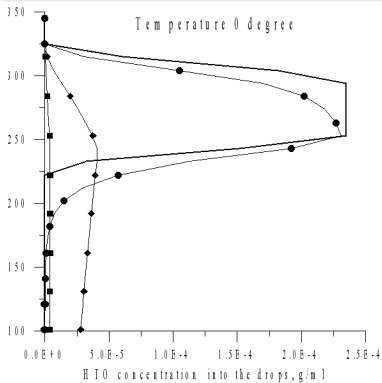
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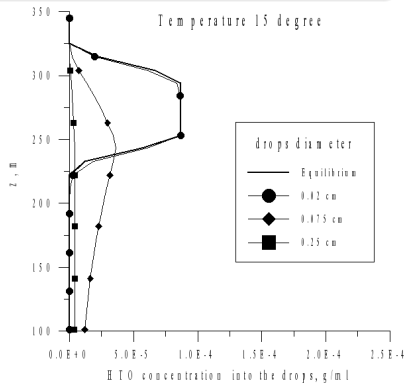
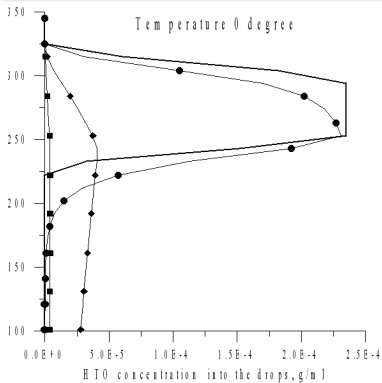
Influence of the temperature

- Washout process is influenced by air temperature
- Figure shows HTO drop concentration according to height
- Figure shows the temperature influence on the washout rate



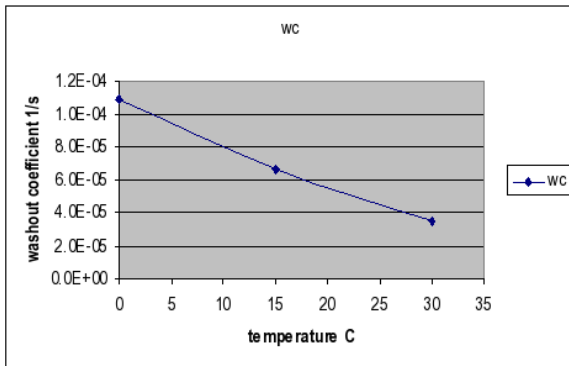
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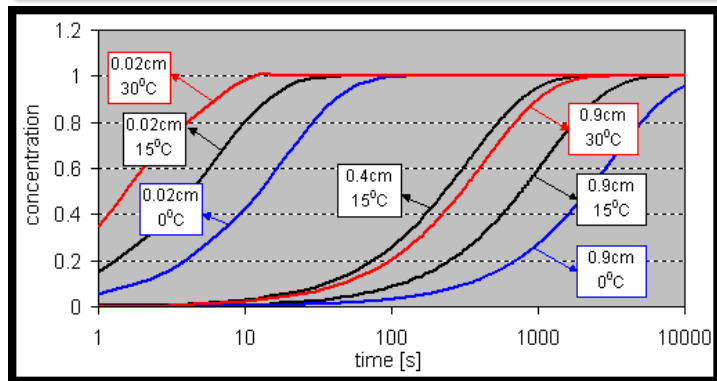


Sensitivity study from Atanassov

- Sensitive analysis of eulerian model (Atanossov) show the influence of rain parameters (70%) and the temperature (50%) on the washout process;
- Sensitivity of temperature, raindrop diameter;
- Sensitivity of atmospheric pressure.

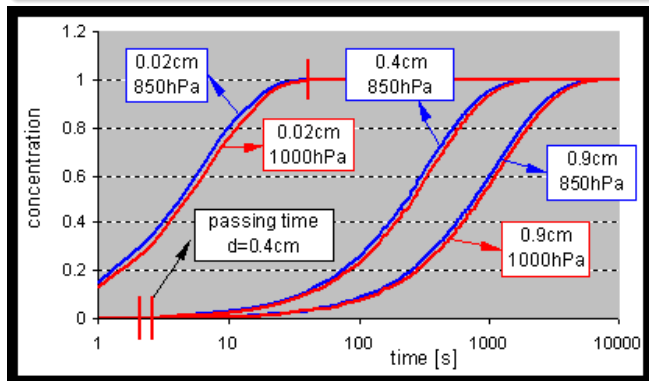
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Other parameters

- The wind influence the raindrop trajectory (taking account by CEA model)
- **Could the Wind influence the DSD ?**
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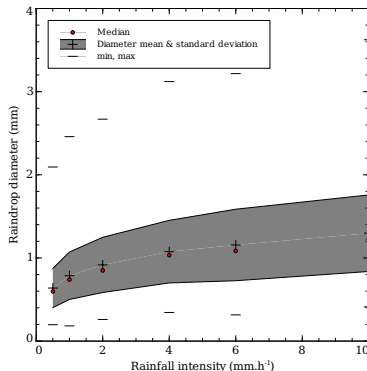
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Models incertainty

- Average rainfall intensity, distribution, diameter of raindrop, velocity are often used but which uncertainty do we do ?;

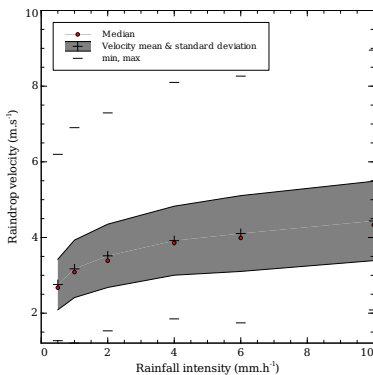
Models uncertainty

- Figure shows mean and uncertainty of diameter for the lognormal distribution by using the parameters given by Feingold (1000 simulations that represents raindrops) and the Andronache formula to compute diameter;



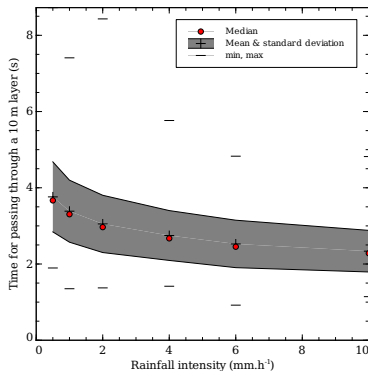
Models uncertainty

- Figure shows the raindrop average velocity according to Andronache formula;



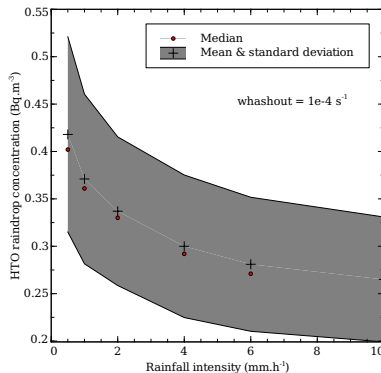
Models uncertainty

- Figure shows the time needs by drops to cross a 10 m layer according to the rain drop velocity calculate before;



Models uncertainty

- Figure shows the HTO average activity of raindrop computed by Chamberlain equation, for a specific activity of water vapor in air of 1000 Bq.m^{-3} and a washout rate of 10^{-4} s^{-1} .



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- 2 Scavenging of HTO
- 3 Washout rate and washout ratio
- 4 Rain characteristics
- 5 Models**
- 6 Conclusions

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 - Complex model (Golubev, VNIIEF) can take into account kinetics of HTO exchange between vapor and liquid phase with parameters (rain drop spectra, rain intensity, condensation-evaporation on drop's interface);
 - \Rightarrow wind velocity and better choice of drop velocity explain the difference with the Belot model;
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