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# 1-D MODELING OF THE DENEbulIZATION OF FOGS BY HYGROSCOPIC SEEDING

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## ABSTRACT

The purpose of the study is the modeling of the denebulization of fogs by micronic hygroscopic particles from the initial seeding to the fall of the rain droplets on the ground. The microparticles generated by flares are made of hygroscopic salts. Three stages occur: first, the condensation of water vapor on solid particles, this is the deliquescent stage which is very fast and can be ignored by the model, then the stage of condensation of water vapor on liquid salted particles / evaporation of fog droplets and finally the coalescence stage when the liquid salted particles diameter become large enough. Three different salts have been tested: NaCl, CaCl<sub>2</sub> and KCl. The 1-D modeling approach studied considers the conservation equations (salt mass, water mass, salted droplets number density, fog droplets number density), the hygroscopicity of the salts through the water activity in the aqueous solution and the coalescence induced by gravity.

The model reveals that NaCl is the most efficient salt because of its stronger hygroscopicity at high dilution rates although CaCl<sub>2</sub> is the more hygroscopic at low dilution rates. A cloud of NaCl particles of 9 μm diameter initially generated over a height of about 7 m with a number density of 12 cm<sup>-3</sup> allows to completely dissipates a typical fog layer of 40 m within less than 13 mn. This means that 10 kg of NaCl particles could be able to clear such a fog over 13.25 ha. The model shows that a layer of 100 m can be cleared within 29 mn. From the calculations, the contribution of the coalescence to the denebulization process is less than 1%, phenomena of

condensation / evaporation are preponderant for these operating conditions.

## NOMENCLATURE

$a_{w,\beta}$	Water activity in the solution (-)
$D_v$	Diffusion coefficient of water vapor in air (m <sup>2</sup> .s <sup>-1</sup> )
$F_{\alpha\beta}^g$	Coalescence frequency between droplets $\alpha$ / $\beta$ (m <sup>3</sup> .s <sup>-1</sup> )
$g$	Standard gravity acceleration (m.s <sup>-2</sup> )
$HV$	Horizontal visibility (m)
$IE$	Efficiency index (-)
$m$	Water refractive index (-)
$M$	Molar mass (kg.mol <sup>-1</sup> )
$N$	Number density (m <sup>-3</sup> )
$P_{vap}$	Water vapor partial pressure (Pa)
$P_s$	Saturation water vapor partial pressure at the salted droplet surface (Pa)
$P_{sat}$	Saturation water vapor partial pressure (Pa)
$Q_{ext}$	Extinction efficiency (-)
$R$	Gas constant (J.mol <sup>-1</sup> .K <sup>-1</sup> )
$R_0$	Initial radius of seed particles (m)
$r_a$	Fog droplet radius (m)
$R_\beta$	Seed particle / salted droplet radius (m)
$RH$	Relative humidity (%)
$Sh$	Sherwood number (-)
$t$	Time (s)
$T$	Temperature (K)
$v$	Terminal settling velocity (m.s <sup>-1</sup> )
$z$	Vertical coordinate (m)

Greek

$\beta_{ext}$	Extinction coefficient ( $m^{-1}$ )
$\varepsilon$	Volume fraction (-)
$\lambda$	Wavelength (m)
$\rho$	Density ( $kg.m^{-3}$ )
$\mu$	Dynamic viscosity (Pa.s)

Subscript

$a$	Air
$\alpha$	Fog droplet phase
$\beta$	Seed droplet phase
$w$	Liquid water

## INTRODUCTION

Artificial rain by hygroscopic seeding is an old technique, the first attempts were performed in South-Africa in the 90's [1] with significant results. The seeding was performed using hygroscopic flares made of hygroscopic salts and combustion agents. Flares were carried by planes. Nowadays, with the developments of the drones, a new opportunity arises to reduce the costs of this technique. A consortium led by Etienne LACROIX with several partners works on the project FIRST. The aim is to use a drone equipped with hygroscopic flares to perform the denebulization of fogs over sensitive area (e.g. airports). Therefore, in the present study, the efficiency of this technique is investigated regarding fog dissipation.

A suspension of droplets in the air above the ground is considered as a fog when the horizontal visibility ( $HV$ ) is lower than 1000 m. Typical fogs liquid water content is in the range of 0.05 and 0.5  $g.m^{-3}$ , mean droplet diameter is between 5 and 15  $\mu m$  and the droplet number density can vary between 100 and 600  $cm^{-3}$ . Moreover, air is saturated by water vapor, i.e. the relative humidity ( $RH$ ) is of 100%. In fogs, wind and turbulence are quite low.

Modeling studies of clouds hygroscopic seeding are available in the literature, but only the team of Silverman studied the dissipation of fogs. In [2], a 1-D approach considers monodisperse NaCl particles seeded over a bidisperse fog. Evaporation / condensation phenomena and coalescence induced by gravity are taken into account. The results show that the fog dissipation can be efficient provided the initial salt particles is well chosen (around 10  $\mu m$ ) and that the amount of particles to seed is proportional to the initial water mass fraction in the fog. In [3], the same 1-D approach was used to test different salts. From this study, hydroxides are the most efficient, followed by chlorides, then bromides, nitrates, iodides and finally sulfates.

## ASSUMPTIONS AND GOVERNING EQUATIONS

The following assumptions are considered: *i*) isothermicity between gas and liquid phases, *ii*) the temperature in the fog layer remains constant over time, *iii*) the Kelvin effect, i.e. the vapor pressure change due to a curved liquid / vapor interface, is negligible (droplet diameters  $> 0.1 \mu m$ ), *iv*) the Brownian motion is negligible (droplet diameters  $> 1 \mu m$ ), *v*) the deliquescence step, i.e. the diffusion of water in the solid salt

particles, is very short ( $< 1s$ ) and is not considered in the model, initial salt particles are considered in a liquid state, *vii*) gravity induced coalescence only is considered, *viii*) axial diffusion and convection of air and water vapor are neglected, *ix*) fog droplets and salt particles are monodisperse, *ix*) for an initial salt particle of radius  $R_0$  and density  $\rho_0$ , we consider instead a radius  $R'_0 = R_0 (\rho_0 / \rho_w)^{1/3}$  and a constant density  $\rho_w$ .

Considering two liquid phases,  $\alpha$  for the pure water phase (fog droplets) and  $\beta$  for the salted droplets phase, we obtain the following governing equations:

$$\frac{dN_\alpha}{dt} = -\frac{\partial}{\partial z}(N_\alpha v_\alpha) - F_{\alpha\beta}^s \quad (1)$$

$$\frac{dN_\beta}{dt} = -\frac{\partial}{\partial z}(N_\beta v_\beta) \quad (2)$$

$$\frac{d\varepsilon_\alpha}{dt} = -\frac{\partial}{\partial z}(\varepsilon_\alpha v_\alpha) + \frac{3}{2}\varepsilon_\alpha Sh_\alpha D_v M_w (\rho_w R T r_\alpha^2)^{-1} (P_{vap} - P_{sat}) - (4/3)\pi r_\alpha^3 F_{\alpha\beta}^s \quad (3)$$

$$\frac{d\varepsilon_\beta}{dt} = -\frac{\partial}{\partial z}(\varepsilon_\beta v_\beta) + \frac{3}{2}\varepsilon_\beta Sh_\beta D_v M_w (\rho_w R T R_\beta^2)^{-1} (P_{vap} - P_s) + (4/3)\pi r_\alpha^3 F_{\alpha\beta}^s \quad (4)$$

$$\frac{d\varepsilon_w}{dt} = -\frac{\partial}{\partial z}(\varepsilon_w v_w) - \frac{\partial}{\partial z}(\varepsilon_{w,\beta} v_\beta) \quad (5)$$

$$P_{vap} = \rho_w R T M_w^{-1} (\varepsilon_w - N_\alpha \varepsilon_\alpha - N_\beta \varepsilon_{w,\beta}) \quad (6)$$

Equations (1) and (2) are the number density balance equations for phases  $\alpha$  and  $\beta$  respectively, equations (3-5) are the mass balance equations for phase  $\alpha$ ,  $\beta$  and water respectively, and relation (6) is the expression giving  $P_{vap}$ .  $P_{sat}$  is given by the following relation [4]:

$$P_{sat} = (10^5 / 760) \exp(18,3036 - 3816,44(T - 46,13)^{-1}) \quad (7)$$

The saturation water vapor partial pressure at the salted droplet surface  $P_s$  is given by:

$$P_s = a_{w,\beta} P_{sat} \quad (8)$$

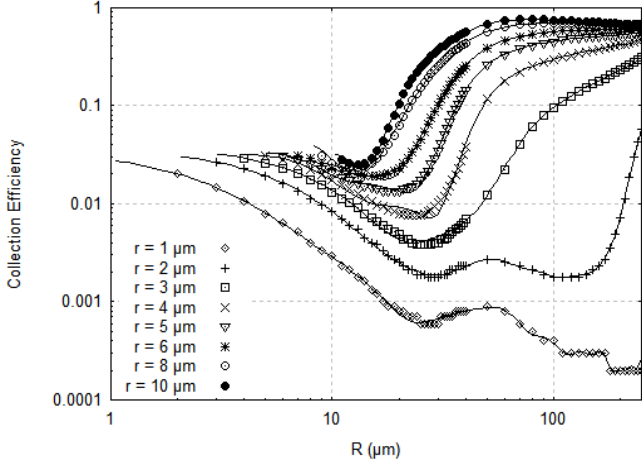
The water activity in the solution  $a_{w,\beta} (\leq 1)$  depends on the salt used and on its concentration in the solution (see next section). Note that, as the salts become more and more diluted in the growing salted droplets, the water activities tend toward 1 and therefore the condensation phenomenon vanishes.

Coalescence induced by gravity happens because droplets of different diameters (fog droplets / salted droplets) fall at different terminal settling velocities. At a given height, there are only two populations of droplets: fog droplets with a radius  $r_\alpha$  and salted droplets with a radius  $R_\beta$ . The coalescence frequency between these two populations is given by:

$$F_{\alpha\beta}^s = \pi (r_\alpha + R_\beta)^2 N_\alpha N_\beta (v_\beta - v_\alpha) E_{coll}(r_\alpha, R_\beta) E_{coal}(r_\alpha, R_\beta) \quad (9)$$

$E_{coll}$  is the collision efficiency, this is the fraction of droplets which, initially moving on a collision course with respect to other droplets, actually collide (make surface contact) with the other droplets, it strongly depending on  $r_\alpha$  and  $R_\beta$ , and is given by [5].  $E_{coal}$  is the coalescence efficiency given by [6]. Note that

these references are the most recent and supposed to be much most reliable than the ones used in older studies. The product of  $E_{coll}$  and  $E_{coat}$  is the collection efficiency which is shown by Figure 1. The data have been fitted by polynomials of degree 20.



**FIGURE 1. COLLECTION EFFICIENCY AS A FUNCTION OF DROPLETS RADII ( $R > r$ )**

Due to the appearance of some numerical oscillations during the calculations, equations (3) and (4) have been replaced by combinations of equations (1) / (3) and (2) / (4) in order to express straightly the time derivative of the droplets radii:

$$\begin{cases} \frac{dr_\alpha^3}{dt} = -v_\alpha \frac{\partial r_\alpha^3}{\partial z} + \frac{3}{2} r_\alpha Sh_\alpha D_v M_w (\rho_w RT)^{-1} (P_{vap} - P_{sat}) \\ \frac{dR_\beta^3}{dt} = -v_\beta \frac{\partial R_\beta^3}{\partial z} + \frac{3}{2} R_\beta Sh_\beta D_v M_w (\rho_w RT)^{-1} (P_{vap} - P_s) \\ + I_\alpha^3 N_\beta^{-1} F_{\alpha\beta}^g \end{cases} \quad (10)$$

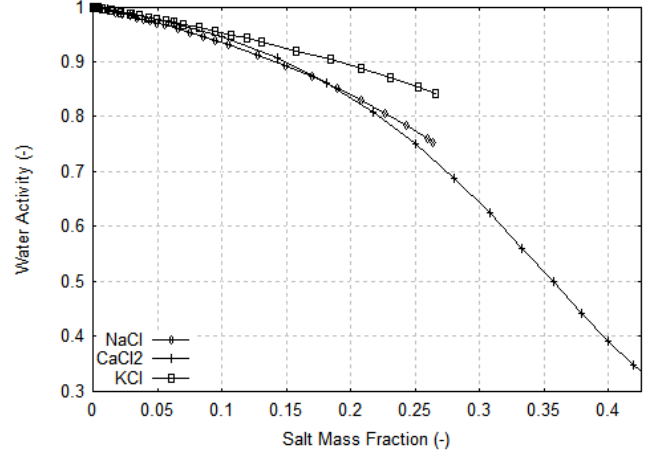
For the range of diameters studied here and densities ( $\rho_w/\rho_a < 1000$ ), the droplets terminal settling velocity is given by:

$$v(r) = \frac{g(2r)^2(\rho_w - \rho_a)}{18\mu_a} \quad (12)$$

### WATER ACTIVITY IN THE SALTED SOLUTIONS

Hygroscopicity of the salts is taken into account by the water activity in the solution ( $a_{w,\beta}$ ) of the salted droplets. Water activity is defined as the ratio of the vapor pressure of water in a material to the vapor pressure of pure water. The lower is the water activity, the higher is the hygroscopicity of the solution. Water activities of solutions containing NaCl, CaCl<sub>2</sub> and KCl in an ionic form are given by [7, 8] and are shown by Figure 2 as a function of the salt mass fraction  $w_s$ . At low dilution rates, CaCl<sub>2</sub> is the most hygroscopic (i.e. lowest water activity) but this is not the case at high dilution rates: for  $w_s$  lower than 0.18,

NaCl solution becomes the most hygroscopic and even KCl solution is slightly more hygroscopic than CaCl<sub>2</sub> solution for  $w_s$  lower than 0.025.



**FIGURE 2. WATER ACTIVITY IN THE SOLUTION WITH SALTS NaCl / CaCl<sub>2</sub> / KCl**

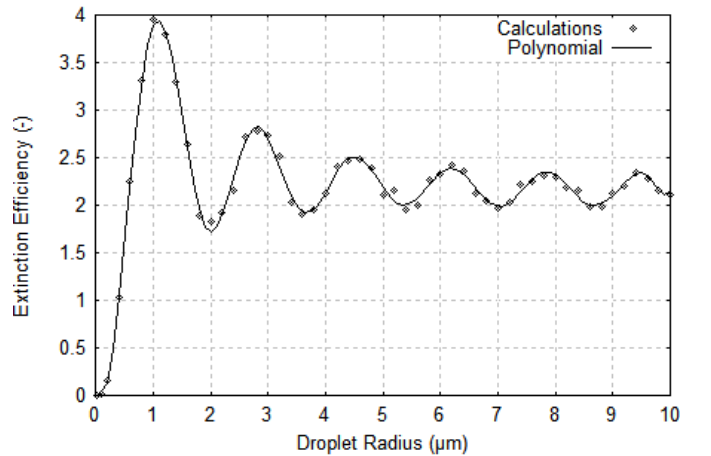
### HORIZONTAL VISIBILITY

Results have been analyzed in term of final horizontal visibility. By convention, the horizontal visibility is given by [9]:

$$HV = 3.91/\beta_{ext} (\lambda = 550 \text{ nm}) \quad (13)$$

$$\text{with: } \beta_{ext} = \pi N_\alpha r_\alpha^2 Q_{ext}(m, r_\alpha, \lambda) \quad (14)$$

where the liquid water refractive index  $m$  is equal to  $1.333 - 1.96 \times 10^{-9}i$  at a wavelength of 550 nm [10]. Figure 3 shows the extinction efficiency  $Q_{ext}$  calculated as a function of  $r_\alpha$  at a wavelength of 550 nm using a Mie scattering calculator [11].



**FIGURE 3. EXTINCTION EFFICIENCY  $Q_{ext}$  CALCULATED FOR A WAVELENGTH OF 550 nm VS. DROPLET RADIUS  $r_\alpha$**

Note that the calculated data have been fitted by a polynomial of degree 20.

### SIMULATIONS PARAMETERS

The 1-D model has been introduced into Matlab using the partial differential equation solver pdepe (finite elements). This solver uses a second order accurate spatial discretization and the time integration is done by ode15s.

A typical monodisperse fog has been considered with an initial fog droplets number density  $N_{\alpha,max}$  of  $400 \text{ cm}^{-3}$  and an initial droplet diameter of  $10 \text{ }\mu\text{m}$ . The initial fog layer is about 40 m. The temperature has been fixed to  $10^\circ\text{C}$  and the  $RH$  to 100%. The salt particles have also been considered monodisperse with an initial diameter of  $9 \text{ }\mu\text{m}$  and initially seeded at the top of the fog layer with an initial maximum number density  $N_{\beta,max}$ . Initial profiles of  $N_{\alpha}/N_{\alpha,max}$  and  $N_{\beta}/N_{\beta,max}$  are shown by Figure 4. Note that the initial shape of the number density of the seed particles is conjectural. Dirichlet boundary conditions have been considered at the top of the layer.

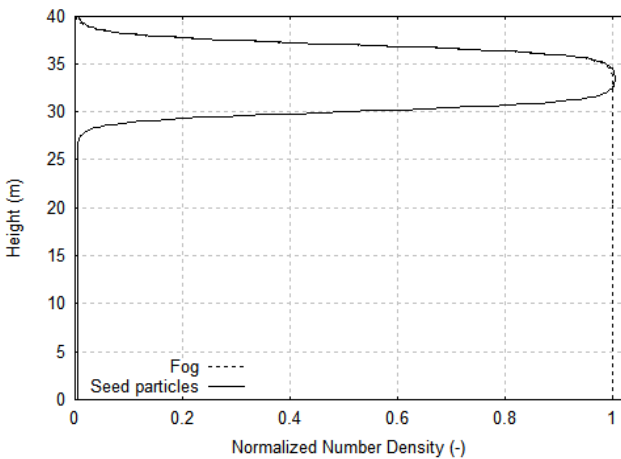


FIGURE 4. INITIAL NORMALIZED NUMBER DENSITIES OF FOG AND SEED PARTICLES

### RESULTS

For the first calculations, a value of  $12 \text{ cm}^{-3}$  was considered for  $N_{\beta,max}$ . The Figure 5 shows the initial and final horizontal visibilities vs. height. As a result, it appears that the fog layer is almost entirely cleared using the salt NaCl. Only about 80% of the layer is cleared using KCl or  $\text{CaCl}_2$ . KCl seems slightly more efficient than  $\text{CaCl}_2$ . These results are very interesting because they show that the hygroscopicity of the salt at high dilution rates is determinant. The denebulization processes are completed after 12 mn 30 s, which is rather long. Note that the final time is considered when all the salted droplets have fallen to the ground ( $z = 0$ ), thus they can not impact the final visibility. Moreover, coalescence induced by gravity is very low: only 0.3% of the fog droplets are captured by the salted droplets, evaporation (of the fog droplets) /

condensation (on salted droplets) are the preponderant phenomena.

Let us define an efficiency index ( $EI$ ) as follows:

$$EI = \frac{\text{Total of absorbed water/Mass of salt seeded}|_{\text{run } x}}{\text{Total of absorbed water/Mass of salt seeded}|_{\text{reference run}}} \quad (13)$$

where the reference run is the previous one with the salt NaCl. For the three previous runs, we get:  $IE(\text{NaCl}) = 1$ ,  $IE(\text{KCl}) = 0.91$  and  $IE(\text{CaCl}_2) = 0.89$ .

Figure 6 shows the final relative humidity in the cleared layer. A depletion has appeared: toward the middle of the layer,  $RH$  is of 99.3%. It means that the water absorbed by the salted droplets do not come only from the fog droplets but also from the ambient humidity. This is a very interesting result because it means that a new fog cannot reappear immediately after the end of the process of denebulization in the cleared zone, fog cannot reappear as long as the air is not saturated with water vapor

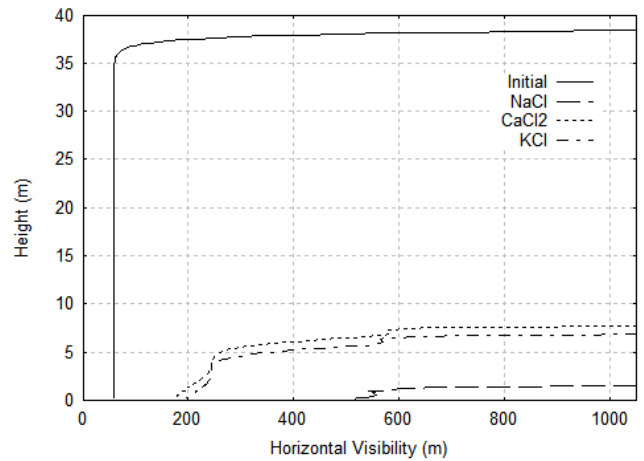


FIGURE 5. INITIAL AND FINAL HORIZONTAL VISIBILITY VS. HEIGHT – SALTS NaCl /  $\text{CaCl}_2$  / KCl

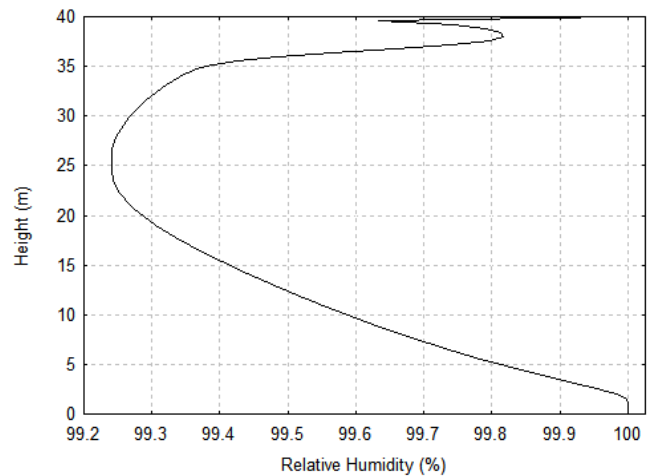


FIGURE 6. FINAL RELATIVE HUMIDITY VS. HEIGHT

again. Then, a characteristic time of water vapor diffusion in air can be estimated (by  $L^2/D_v$  with  $L = 20$  m), giving about 200 days. Therefore, the resaturation of the layer by water vapor will not be due to ordinary diffusion, but rather to convection, turbulent diffusion or temperature changes.

For the next calculations, we have retained NaCl because this is the more efficient salt. Three additional calculations have been performed considering thicker fog layers (75 m, 100 m and 150 m).  $N_{\beta,max}$  values have been adjusted to allow a total clearing of the fog layer. Results are reported in table 1. As expected,  $N_{\beta,max}$  must be increased as a function of the fog layer thickness, but not linearly: the thicker is the fog layer, the more time has the evaporation / condensation phenomena to occur. This is why  $IE$  increases with the layer height (Table 1). A value of  $N_{\beta,max}$  of  $18 \text{ cm}^{-3}$  is necessary to clear the 100 m fog layer and the process last for 29 mn.

**TABLE 1:** CALCULATIONS RESULTS WITH SALT NaCl

Fog height (m)	$N_{\beta,max}$ ( $\text{cm}^{-3}$ )	Salt mass ( $\text{mg}\cdot\text{m}^{-2}$ )	$D_{\beta,max}$ at ground ( $\mu\text{m}$ )	Total time of denebulization	$IE$ (-)
40	12	75.4	62.7	12 mn 30 s	1
75	15.8	99.3	70.3	22 mn 30 s	1.4
100	18	113.1	73.2	29 mn	1.6
150	20.7	130.1	79.8	45 mn	2.1

Finally, a last calculation has been performed still considering NaCl as salt and a layer of 40 m with a layer temperature now of  $0^\circ\text{C}$ . With these operating conditions,  $N_{\beta,max}$  must be increased to  $15.5 \text{ cm}^{-3}$  for a full denebulization of the fog and the process last longer: 16 mn 40 s. At this lower temperature,  $P_{vap}$  and  $P_{sat}$  are lower and therefore evaporation / condensation phenomena are slower.  $IE$  is of 0.74 only.

## CONCLUSION

Among the three salts considered in these simulations, NaCl is the most efficient to clear the fog in the operating conditions tested because of its highest hygroscopicity at high dilution rates. Evaporation / condensation phenomena are preponderant while coalescence induced by gravity is negligible.

From the calculations, 10 kg of NaCl seed particles can clear a fog layer of 40 m height over 13.25 ha in less than 13 mn. The technique seems really efficient but necessitates an appropriate anticipation in the treatment in order to provide the results in an appropriate timing.

Different operating conditions (particles sizes, fog density) and coalescence induced by turbulence will be considered in further investigations.

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