# Desalination of seawater by spray freezing in a natural draft tower

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Author post-print (accepted) deposited by Coventry University's Repository

### Original citation & hyperlink:

Liu, Y, Ming, T, Wu, Y, Richter, R, Fang, Y & Zhou, N 2020, 'Desalination of seawater by spray freezing in a natural draft tower', Desalination, vol. 496, no. 1, 114700. <u>https://dx.doi.org/10.1016/j.desal.2020.114700</u>

DOI 10.1016/j.desal.2020.114700 ISSN 0011-9164

Publisher: Elsevier

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1	Clear Version
2	Desalination of seawater by spray freezing in a natural draft tower
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16	
17	Abstract
18	The freeze-melting process can be a viable method for the purposes of desalination
19	because of its low energy consumption, ignorable corrosion issues, and without huge
20	pressure or membrane replacement work. Large contact area for heat and mass transfer
21	per unit mass of water between the water and air and low heat resistance results in
22	higher energy efficiency during spray freezing desalination process compared to other
23	freezing desalination methods. A 200m high desalination tower was proposed in this
24	paper that could generate 27.7 $kg/s$ fresh water in the form of water droplets with
25	2mm diameter at an atmospheric temperature of -26°C. This research has founded that
26	the natural convective airflow induced by the heat released by the warm water in the
27	freezing process could generate through the wind turbine mounted in this system
28	approximately one-third of the energy consumed by the water pump of the system. This
29	free energy has never been studied in previous research. The power consumption
30	required to produce 1 $m^3$ fresh water in this system is approximately 1.07 kWh.
31	Compared to traditional desalination methods, the power consumption of our new spray
32	freezing desalination system is much lower than previous systems with the same mass
33	flow rate of fresh water. Only 375.4 kJ cold energy to produce one-kilogram fresh
34	water. Thus, this spray freezing desalination system could be employed in desalination
35	industry if free cold energy (e.g. from the cold atmosphere or the regasification process
36	of LNG) and seawater resources are available.
37	

# 38 Keywords:

39 Compressible airflow; Natural draft tower; Seawater desalination; Spray freezing;

40 Water droplet

#### 41

#### Nomenclature

Α	Heat (mass) transfer area per tower unit	$S_h$
	volume (m <sup>2</sup> /m <sup>3</sup> )	t
$A_d$	Surface area of a water droplet (m <sup>2</sup> )	Т
Bi	Biot number	v
$C_d$	Drag coefficient	$Y_{w}$
$C_{p,B}$	Specific heat capacity of dry air (J/kg·K)	$Y_{w}$
$C_{p,w}^{g}$	Specific heat capacity of water vapor (J/kg·K)	
$C_{p,w}^L$	Specific heat capacity of liquid water (J/kg·K)	Ζ
d	Diameter of a water droplet (m)	
D	Diameter of the tower (m)	Gre
$D_{w,a}$	Diffusivity of water vapor in air (m <sup>2</sup> /s)	α
fr <sub>ice</sub>	Ice mass fraction	γ
F	Force (N)	ε
$F_r$	Froude number	ζ
g	Gravitational acceleration, 9.81 (m/s <sup>2</sup> )	η
$G_B$	Mass flow rate of dry air per unit cross-	λ
	section area (kg/m <sup>2</sup> ·s)	ρ
h	Tower height (m)	μ
$h_{conv}$	Convective heat transfer coefficient (W/m <sup>2.</sup> K)	Δ
h <sub>mass</sub>	Mass transfer coefficient (kg/s·m <sup>2</sup> ·(kg/kg) <sup>-1</sup> )	
$H_d$	Specific enthalpy of a droplet (J/kg)	Sub
$H_G$	Specific enthalpy of moist air per unit mass of	а
	dry air (J/kg)	b
$\Delta H_s$	Specific latent heat of fusion (J/kg)	br
$H_w$	Specific enthalpy of the liquid phase (J/kg)	d
$\Delta H_v$	Specific heat of vaporization of water (J/kg)	da
k	Thermal diffusivity ( $W/m \cdot °C$ )	f
L	Mass flowrate of water per unit cross-section	g
	area (kg/m <sup>2</sup> ·s)	in
М	Mach number	ice
$M_a$	Molar mass of moist air (kg/kmol)	k
$m_d$	Mass of a water droplet (kg)	la
$m_w$	Mass flow rate of the feed water (kg/s)	los
Ν	Number of water drops falling through a unit	n
	volume per second	ои
Nu	Nusselt number	po
Р	Air static pressure (Pa)	tu
$P_r$	Prandtl number	w
$P_{w,sat}$	Vapor pressure of the seawater (Pa)	$\infty$
q	Heat-transfer flux (w/m <sup>2</sup> )	0
Q	Heat transferred per unit mass (w/kg)	
R	Specific gas constant for air, 287 (J/kg·K)	Sho
$R_e$	Reynolds number	MZ

#### Sherwood number $S_h$ Time (s) t Т Temperature (K) Velocity (m/s) v Humidity mass ratio of moist air (kg/kg) ľw Humidity mass ratio at the gas-phase side of w,i interface (kg/kg) Height from the bottom of the tower (m) Ζ reek symbols Thermal diffusivity (m<sup>2</sup>/s) α Specific heat ratio of air γ Pressure loss coefficient ε Turbine pressure drop factor ζ Mechanical efficiency of turbine generator η λ Friction coefficient on the wall in the tower Air density (kg/m<sup>3</sup>) ρ μ Dynamic viscosity $(P_a \cdot s)$ Difference Δ ubscripts а air b buoyancy internal bracing br d droplet da dry air freezing point f gravitational g inlet in ice се k kinetic energy lapse rate la loss oss nucleation п outlet out potential oot turbine ur w water atmosphere $\infty$

sea level

#### horthand with no physical meaning

# ЛΧ

S	Cross-section area of the tower (m <sup>2</sup> )	EX	
S <sub>c</sub>	Schmidt number	EXw	

### 42 1. Introduction

Water resources are unevenly distributed on the global. Shortage of affordable clear water is one of the most severe problems in many parts of the world with high population density or rapid industrialization. However, many of these areas located near to the huge water resources - seawater, for instance, Singapore, Alaska, and North China [1-3]. Low-cost and high-quality water can be exploited by desalination from the seawater to meet the urgent demand of daily consumption of a huge population and for future industrial development.

50 When seawater is freezing, ice tends to exclude the impurities during the 51 crystallization process. For instance, the natural-growth sea-ice has a much lower salt 52 content than the original seawater and is drinkable for human after it thaws. This 53 phenomenon was found by sailors and the inhabitants living in polar regions [4]. Freeze 54 desalination consumes less energy input by 6-7 times compared to the evaporative 55 crystallization process due to the larger latent heat of vaporization compared to that of 56 solidification [5]. Also, freeze desalination with no need for ancillary chemicals [4] is 57 more environmental-friendly compared to the evaporative crystallization process. In 58 addition, compared to the Reverse Osmosis method, the freezing melting process does 59 not need huge pressure or membrane replacement [4, 6]. Furthermore, the freeze 60 desalination is insensitive to corrosion problems because of its low operating 61 temperature [7]. Following the freeze desalination process, the treatments such as 62 gravity drainage, crushing, centrifugation, filtering, washing by fresh water or even 63 microwave treatment can further improve the quality of ice by breaking the "ice-salt 64 pocket" in the ice and releasing the concentrated brine out of the ice [8-10]. In addition 65 to the above advantages, the freeze-melting process will be more economic and 66 attractive for freezing seawater if waste or renewable energy is used to freeze the 67 seawater.

68 Cao, et al.[11] utilized the cold (waste) energy released from the regasification 69 process of the LNG (liquefied natural gas) by a flake ice maker to desalinate seawater, 70 which generated 2kg ice with 1kg LNG re-gasification. Chang, et al. [3] desalinated the 71 seawater using the cold energy of LNG by the freezing process in Singapore. The total 72 dissolved solids at around 300 ppm were achieved by washing in the final product, 73 which meets the salinity of 500 ppm of WHO potable water standard. Wang and Chung 74 [12] developed a hybrid desalination process comprising freeze desalination (cold 75 energy from LNG) and membrane distillation processes. High-quality drinkable water 76 with a low salinity of 0.104 g/L was obtained in the freeze desalination process alone. 77 Combined with freeze desalination, the membrane distillation process was employed 78 to treat the brine discharged from the freeze desalination unit. Drinkable clean water 79 was successfully produced by the hybrid process with a total water recovery of 71.5%.

John, et al. [13] even proposed to purify the urban wastewater by the natural freeze.
With this method, no cold energy input was needed and 95% purification efficiency
was achieved. Once free cold energy is available, there is potential to generate clear
water by the freeze desalination method.

84 Some devices were specially designed for crystallizing the salt solution to generate 85 ice and solid salt simultaneously. A novel 151 freezing disk column crystallizer has 86 been built to separate the frozen salt solution into ice and salt under the eutectic freezing 87 point [14]. With this device, as a case study, a 35 w% aqueous sodium nitrate and a 12 88 w% copper sulfate stream fed into the disk column crystallizer were cooled down to 89 the eutectic freezing point to separate water and salt. Compared to conventional multi-90 step evaporation, the energy reductions by using freeze desalination are 30% of that of 91 sodium nitrate and 65% of that of copper sulfate [15].

92 Although it has been demonstrated that the freeze-melting process could be a 93 method for desalination, the block, layer, or falling film freeze processes are not energy-94 efficient. Large thermal resistance exists due to the wall of the heat exchanger, the water, 95 or the ice itself, which results in a low rate of heat transfer and long crystallization time. 96 So, a direct freezing method - spray freezing with a large area of heat transfer with a 97 low thermal resistance is preferred in the freeze-melting process. During the spray 98 freezing process, the water droplets are sprayed directly into the cold air to be frozen. 99 The large surface area per unit volume of water drops makes the rates of both cooling 100 and ice formation much faster.

101 When the seawater is pumped and sprayed into the cold air, the surface 102 temperature of water droplets falls below ice nucleation temperature, they solidify from 103 the outside and push the impurities to the center of the drop by crystallization front, 104 resulting in a higher concentrated liquid and nearly pure ice [16]. The outer shell 105 fracture, when droplets fall and contact with the cold ground. The built-up pressure 106 inside the droplet caused by phase change expansion can also rupture the ice shell, and 107 release the liquid impurities out of the droplet [17]. The excess water drained out from 108 a spray ice deposit becomes runoff that is more concentrated than the original seawater.

109 The spray freezing process was proposed to produce drinkable water in Alaska in 110 the 1960s, which achieved salt concentration in the ice 0.007 times lower than the 111 source seawater [2]. The spray freezing was also used to treat pulp mill effluent and oil 112 sands tailings pond water by Gao, et al. [18]. In Gao's experiment, greater than 60% 113 impurity reduction in the spray ice was obtained when 30% of the total volume of the 114 sprayed water was released as runoff. Another field experiment was conducted to 115 evaluate the efficiency of spray freezing to remove dissolved chemicals from the lake 116 of tailing water at the Colomac Mine. The experimental analysis showed that after the 117 initial 39% of the spray ice column had been melted, the efficiency of dissolved 118 chemical removal in the ice core reached up to 87-99% [19]. Tatarniuk, et al. [20] used 119 spray freeze separation to concentrate salt in snowmelt water gathered from snow

4

runoff water on the roads into higher concentrated reusable brine to recover and recycle salts, intending to drive down salt costs in the Canadian winter. The spray freezing could concentrate the water 1.3~1.4 times higher than the source water, with much purer ice mound left.

124 Although the spray freezing to purify water is proved to be feasible, a large-scale 125 continuously-running spray freezing system has not been proposed. The heat and mass 126 transfer mechanism during the spray freezing and the impact factors in this spray 127 freezing process have not been analyzed yet. Besides, in a large-scale continuously-128 running spray freezing system, the heat energy released from the water droplets during 129 its freezing was never utilized in the previous research. Actually, this energy is a kind 130 of heat that could heat the cold air and form natural air current which can be utilized in 131 a spray freezing tower to propel the turbines mounted on the top of the tower to generate 132 electricity (see Fig. 1). So, the main objective of this paper is to desalinate seawater by 133 spray freezing utilizing the free cold energy from the atmosphere (or from LNG) and 134 generate green power simultaneously. An economic analysis of this method compared 135 with freezing by a heat-exchanger is also undertaken in this paper.

136

#### 137 2. Principle of the spray freezing desalination

To freeze the sprayed seawater droplets and utilize the free heat energy released by the water, a 200m high spraying tower with turbines mounted at the bottom is introduced in this paper. The warm seawater droplets are sprayed from the top of the tower, the freely-falling droplets would release heat into the air inside the tower. The heated air in the tower would float upwards because of its lower density, which caused a natural air draft. The updraft can be used to propel the turbines to generate electricity.

At the upper part of the tower, the droplets will be in the liquid state. When the temperature of droplets reduces to the freezing temperature, they will be frozen and become ice particles. If the droplets are partially frozen, the mixture of ice and concentrated brine is collected by the ice container at the bottom and filtered. Pure ice will be separated from brine and stored to produce fresh water. The concentrated brine will drain off. This proposed spray freezing desalination is presented in Fig. 1. The geometrical parameters of this system are derived from [21].



152 Fig. 1. (a) Schematic diagram of the spray freezing desalination system, (b) flow directions of seawater 153 droplets and air.

154 The principle of desalination by freeze crystallization can be described by the 155 typical phase diagram of binary solution which represents the equilibrium lines between 156 solid and liquid states of materials. A schematic representation of the phase change 157 diagram of saline water (NaCl-H<sub>2</sub>O) is depicted in Fig 2. Line 1 defines the freezing 158 point of water at different sodium chloride mass content.



159

151

160 Fig. 2. Phase diagram for the NaCl solution as a function of temperature and salt concentration [22].

161 The starting point is on the left side of the eutectic point when the mass fraction is 162 lower than 23.3wt%. The unsaturated saline solution is cooled until reaching the 163 equilibrium point on Line 1. Further cooling will take the solution along Line 1 with 164 equilibrium temperature depressing and concentration in remain liquid increasing until 165 the eutectic point is reached at  $-21.1^{\circ}$ C for NaCl solution.

166 The freezing point depression,  $T_f$ , depends linearly on the concentration of the 167 salt and can be calculated as [23]:

 $T_f = -54.1126 \left(\frac{S_w}{1 - S_w}\right), -7.7 \le T_a \le 0 \ \mathcal{C} and \ 0 \le S_w \le 12.47\%$ (1)

169 where  $S_w$  is the salinity of the seawater (0.035 for the initial salinity before 170 desalination).

171

# 172 **3.** A single water droplet freezes in the tower

173 3.1 A single water droplet velocity

174 As the heat and mass transfer rate between the water droplets and the surrounding 175 air is decided by the drops' velocity, the velocity of the drops must be analyzed first. In 176 this work, the water drops were assumed to be spherical [24] and fall vertically in the 177 one-dimensional model. The forces exerted on one droplet include gravity  $F_g$ , buoyancy 178 from the air  $F_b$ , air friction drag  $F_a$ . The force balance equation is:

179 
$$m_d \frac{dv_w}{dt} = F_g - F_b - F_a \tag{2}$$

180 Where  $v_w$  = velocity of a water droplet, t= time (s).

181 The mass of a water droplet  $m_d$  can be calculated by  $m_d = \pi d^3 \rho_w / 6$ , the 182 gravitational force of a droplet is  $F_g = \pi d^3 g \rho_w / 6$  and the buoyancy is  $F_b = \pi d^3 g \rho_a / 6$ . 183 The drag force by air friction  $F_a$  is expressed as  $F_a = \pi C_d \rho_a (v_a + v_w)^2 d^2 / 8$ . The 184  $\rho_w$  and  $\rho_a$  represent the density of water and air, respectively. Then the motion 185 equation of the water droplet can be written as [25]:

186 
$$\frac{dv_w}{dt} = g\left(1 - \frac{\rho_a}{\rho_w}\right) - \frac{3}{4} \frac{C_d}{d} \frac{\rho_a}{\rho_w} (v_a + v_w)^2$$
(3)

187 For liquid droplets, the following drag coefficient correlations were adopted [26]:

188 
$$C_{d} = \begin{cases} \frac{24.0}{\text{Re}}, \text{Re} \leq 1\\ \frac{24.0}{\text{Re}} (1 + 0.15 \text{Re}^{0.687}), 1 < \text{Re} \leq 1000\\ 0.44, \text{Re} > 1000 \end{cases}$$
(4)

189 where  $R_e$  is the Reynolds number,  $R_e = \rho_a (v_a + v_w) d/\mu_a$ .

190 The thermophysical properties of seawater and air needed for the numerical191 calculation are listed in Table 1.

#### Table 1

Tarameter quantities and correlations used.				
Parameter	Quantity/correlation	Units	References	
$S_w$	35	g/kg	[27]	
$T_{f}^{1,2}$	$-54.1126\left(\frac{S_w}{1-S_w}\right)$	°C	[23]	
$ ho_w$	$1000 + 0.8S_w$	$kg/m^3$	[23]	
$C_{p,w}^L$	$\frac{1.005 - 0.004136S_w + 0.0001098S_w^2}{-0.000001324S_w^3}$	Cal/g·°C	[23]	
$\Delta H_v^3$	2498510 (0°C, salinity 3.5%)	J/kg	[27]	
$\Delta H_s^3$	329928 (salinity 3.5%)	J/kg	[27]	
$P_{w,sat}^{3}$	602.4 (0°C, salinity 3.5%)	$P_a$	[27]	

Parameter quantities and correlations used.

$$\rho_a = \frac{P \cdot M_a}{8.3145 \times 10^3 T_a}$$
 $kg/m^3$  [28, 29]

$$M_a \qquad (1+Y_w) / \left(\frac{Y_w}{18.015} + \frac{1}{28.966}\right) \qquad kg/kmol \quad [28, 29]$$

$$D_{w,a}$$
 2.227 × 10<sup>-5</sup>  $\left(\frac{I_a + 273.15}{273.15}\right)$   $m^2/s$  [23, 29]

$$\mu_a \qquad \mu_0 \left[ \frac{416.16}{T_a + 393.15} \left( \frac{T_a + 273.15}{296.16} \right)^{1.5} \right] \qquad P_a \cdot s \qquad [23]$$

$$\mu_{0} = 1.8325 \times 10^{-5} P_{a} \cdot s$$

$$\alpha_{a} \qquad \frac{1}{(57736 - 585.78T_{a})} \qquad m^{2}/s \qquad [23, 29]$$

$$\kappa_{a} \qquad 0.024577 + 9.027 \times 10^{-5} T_{a} \qquad W/m \cdot {}^{\circ}\text{C} \qquad [23, 29]$$

<sup>1</sup> The equation is valid in  $-7.7 \le T_a \le 0^{\circ}$ C and  $0 \le S_w \le 12.47\%$ .

<sup>2</sup> The salinity,  $S_w$ , appears as a fraction in this formula (0.035 for Standard Ocean salinity).

<sup>3</sup> The accurate data is available from *http://www.teos-10.org*.

192

193 By solving Equation 3, the free-falling droplet velocity variation from the release in the static air is shown in Fig. 3 (the variation of  $v_a$  is discussed in section 4 in this 194 paper). Larger droplets tend to have a higher terminal velocity. The small differences 195 196 in droplet velocity between the present model and the model of Dehghani-Sanij et al. [30] are due to the different  $C_d$  estimation. The velocity variations are in good 197 198 agreement with the experimental results conducted by Chowdhury, et al.[31]. The 199 experiment demonstrated that velocity of water drops grew very rapidly at the 200 beginning and reached to the peak (terminal) velocity after a very short time period and 201 distance. When the droplets reaching the peak (terminal) velocity, they no longer 202 accelerate and keep at this velocity constantly. The terminal velocities of falling water 203 droplets in the air are also demonstrated by many researchers in their experiments [32-204 34].



8

205

Fig. 3. The freely-falling droplet velocity variation after the release in the static air.

207 3.2 A single water droplet freezing process

226

During the freezing process, the temperature inside of a droplet, in general, varies with time and the position. To simplify and solve the transient heat transfer phenomenon of water droplet, the Biot number, which is the ratio of heat convection at the surface to the internal heat conduction of a body, is first evaluated.

212 
$$Bi = \frac{h_{conv} \frac{d}{6}}{k_d}$$
(5)

213 Where the  $h_{conv}$  is the convection heat transfer coefficient and the  $k_d$  is the 214 droplet (water or ice) thermal conductivity. A small Bi represents lower resistance to 215 conduction within a body. For Bi<0.1, the temperature gradient within the droplet can 216 be neglected and uniform temperature within the body is regarded for the droplet.

One may notice that the Bi number would be slightly larger than 0.1 before the phase change stage when the water droplet diameter is larger than 2.5mm. However, in the calculation of Bi number, the circulation and mixing inside a freely falling water droplet, which could enhance internal heat transfer within a body, is not considered yet. The Reynolds number of the internal motion within a droplet is defined as [17, 35]:

222 
$$Re_{int} = \frac{v_a d}{2\left(1 + \frac{\mu_w}{\mu_a}\right)\frac{\mu_w}{\rho_w}}$$
(6)

223 Where  $\mu_w$  and  $\mu_a$  is the dynamic viscosity of droplet and air, respectively.

224 Considering the internal motion, the effective thermal diffusivity is expressed as225 [17, 35]:

$$\alpha_{w,eff} = \frac{k_w}{\rho_w C_{p,w}^L} = \alpha_w (1 + 0.01 R e_{int}) \tag{7}$$

227 The  $\alpha_{w,eff}$  is more than 10 times larger than the  $\alpha_w$  of non-internal-motion 228 seawater, which means that the effective thermal conductivity  $k_w$  is much larger. Thus, 229 even if the water droplet is bigger than 2.5mm, the Bi number is still smaller than 0.1 230 if the internal motion within the droplet is considered. Furthermore, the thermal 231 conductivity of ice is about four times larger than that of seawater  $\alpha_w$ , the Bi number 232 is still smaller than 0.1 when the ice is formed. Hindmarsh, et al. [36] also demonstrated 233 that a simple heat balance model is sufficient for the purpose of solving the internal 234 energy balance of the droplet, giving accurate results. Therefore, the temperature 235 gradient within the droplet is neglected in the whole freezing process of a droplet in this paper and a simple heat balance model was used. 236

The freezing process of a droplet can be divided into four stages: initial (liquid) supercooling, recalescence, solidification (phase change), and post-solidification (solid) stage [36, 37]. The theory was based on that the liquid droplet will not crystallize until it is supercooled and reaches the nucleation temperature  $T_n$  which is much lower than the freezing temperature  $T_f$ . Once the first crystal nuclei are formed, a rapid crystallization happens, resulting in a sudden temperature rise from  $T_n$  to  $T_f$ , which is called the recalescence stage.

The droplet continuously releases heat during its freezing process, the total energyreleased includes the convective heat loss, radiative heat loss, and evaporative heat loss:

$$q_{total} = q_{conv} + q_{rad} + q_{evap} \tag{8}$$

247 
$$q_{conv} = h_{conv}(T_d - T_a)$$
 (9)

$$q_{rad} = \varepsilon \sigma (T_d - T_a)$$
(10)

$$q_{evap} = h_{mass}(Y_{w,i} - Y_w)\Delta H_v \tag{11}$$

250 
$$Y_{w,i} = 0.622 \frac{P_{w,sat}}{P - P_{w,sat}}$$
(12)

251 Where the  $T_d$  is droplet (water or ice) temperature,  $T_a$  is the atmosphere temperature, 252  $\sigma$  is the Stefan-Boltzmann constant,  $\varepsilon$  is the droplet emissivity.  $Y_{w,i}$  is the humidity 253 mass ratio of saturated moist air at the water-air interface, which depends on the droplet 254 temperature  $T_d$ .  $Y_w$  is the humidity mass ratio of moist atmospheric air.

255 The convective heat and mass transfer coefficient  $h_{conv}$  and  $k_{mass}$  were 256 obtained from experimental data by Ranz and Marshall [38]:

257 
$$N_u = \frac{h_{conv}d}{k_a} = 2 + 0.6P_r^{1/3}R_e^{1/2}$$
(13)

$$S_h = \frac{h_{mass}d}{D_{w,a}} = 2 + 0.6S_c^{1/3}R_e^{1/2}$$
(14)

259 where  $N_u$  is Nusselt number,  $P_r$  is Prandtl number  $P_r = \mu_a / \rho_a \alpha_a$ ,  $S_h$  is Sherwood 260 number,  $S_c$  is Schmidt number  $S_c = \mu_a / \rho_a D_{w,a}$ ,  $R_e$  is the Reynolds number.

Due to the mass transfer, the droplet mass decreases continuously until theevaporation ends. The mass change of a droplet can be calculated by:

263 
$$(m_d)_t = h_{mass} (Y_{w,i} - Y_w) A_d \Delta t + (m_d)_{t+\Delta t}$$
(15)

258

$$\frac{dm_d}{dt} = -h_{mass} (Y_{w,i} - Y_w) A_d \tag{16}$$

where  $m_d$  is the mass of a water droplet and the  $A_d$  is the surface area of a water droplet.

According to [16], when a droplet surface temperature reaches the freezing point, ice nucleation begins at the surface, a solid shell grows rapidly around the surface (about 2/30-3/30 s) and then solidifies inwards. The evaporative mass transfer from the water to the ambient air can be neglected during the ice formation process around the droplet surface.

272 Before the solidification (phase change) stage, the energy balance for the droplet273 is:

274 
$$(m_d H_d)_t = (m_d H_d)_{t+\Delta t} + A_d q_{conv} \Delta t + A_d q_{rad} \Delta t + A_d q_{evap} \Delta t$$
(17)

275 and the final expression for the water temperature  $T_d$  is

276 
$$\frac{dT_d}{dt} = -\frac{h_{conv}A_d(T_d - T_a) + \varepsilon\sigma(T_d^4 - T_a^4)A_d + h_{mass}(Y_{w,i} - Y_w)A_d\Delta H_v}{m_d C_{p,w}^L}$$
(18)

277 During the solidification (phase change) stage, the ice is formed from the outside 278 and it pushes the impurities to the center of the drop, resulting in a higher concentrated 279 liquid and nearly pure ice. This concentrated solution would decrease the freezing 280 temperature until the new equilibrium is reached. Thus, the heat released by the droplet 281 will not only facilitate the phase change but also reduce the droplet (both the outside 282 ice and the inside concentrated solution) temperature. Since the diffusion of impurities 283 in the ice can be negligible [37, 39], the droplet temperature will follow the freezing 284 point depression line (see Fig. 2). Based on this theory, the energy balance for the 285 droplet is:

286 
$$(m_d H_d)_t = (m_d H_d)_{t+\Delta t} + A_d q_{conv} \Delta t + A_d q_{rad} \Delta t - \Delta H_s m_d f r_{ice}$$
 (19)  
287 and the final expression is:

$$288 \qquad \Delta H_s m_d \frac{dfr_{ice}}{dt} - \left(C_{p,ice} m_d fr_{ice} + C_{p,w}^L m_d (1 - fr_{ice})\right) \frac{dT_d}{dt} = A_d q_{conv} + A_d q_{rad} (20)$$

289 where  $fr_{ice}$  is the ice mass fraction in the droplet,  $\Delta H_s$  is the specific latent heat of 290 fusion.

Equation 19 could be solved with the freezing point depression equation (Equation
1 for seawater) with a forward difference time step method [37] or a chain rule method
[39]:

294 
$$\frac{dT_d}{dt} = \frac{dT_d}{dfr_{ice}} \frac{dfr_{ice}}{dt} = \frac{dT_f}{dfr_{ice}} \frac{dfr_{ice}}{dt}$$
(21)

295 For seawater, when the ice appears, the freezing point could be expressed as:

296 
$$T_f = -54.1126 \left( \frac{S_w / (1 - fr_{ice})}{1 - S_w / (1 - fr_{ice})} \right)$$
(22)

297 where  $S_w$  is the initial salinity of the seawater before crystallization.

The phase change stage ends when the temperature of the droplet or the concentration of the inside solute reaches the eutectic point (23.3% for seawater). After that the post- solidification stage starts. The droplet temperature can be expressed as:

301 
$$\frac{dT_d}{dt} = -\frac{h_{conv}A_d(T_d - T_a) + \varepsilon\sigma(T_d^4 - T_a^4)A_d}{C_{p,ice}m_d fr_{ice} + C_{p,w}^L m_d(1 - fr_{ice})}$$
(23)

302 To validate this numerical model, the freezing process of a sucrose droplet was 303 simulated. The comparison with the reference [39] and [40] was made in Figure 4. In 304 [39], the temperature variation of a solution droplet during the freezing process was 305 discussed. The solute mass fraction was analyzed in the following paper [40]. The boundary conditions were:  $T_a = -15$  °C, d = 1.56mm,  $v_a = 0.42m/s$ ,  $v_w = 0m/s$ . A 306 slight difference between the model in the reference [39] and [40] with the present 307 308 model was observed in this figure. In [39, 40] the temperature gradient of the surface 309 ice was considered and the inside concentrated solution was treated as a lumped matter.

Thus,  $C_{p,ice}m_d fr_{ice}$  in Equation 20 was not calculated in those papers (see Equation 20 in [39]). While, in this paper, as discussed before, the temperature gradient of the droplet (ice/water) was neglected, since the thermal conductivity of ice is about four times larger than the seawater. The protuberance of the solute mass fraction at the beginning of the freezing process (between 4.5-5s) was caused by the ice formed in the recalescence stage.





Fig. 4. The freezing process of a 5% sucrose droplet

#### 318 4. A system of droplets freeze in the tower

**319** 4.1 Collision and coalescence of water droplets

As discussed in the previous section, since the large droplets will reach a higher terminal velocity than that of the smaller droplets. When different diameters of water droplets are emitted by a spray nozzle, the larger droplets will collide with the smaller ones during the falling process in the tower. Some of the smaller droplets will be collected by the larger ones after the collision and resulting in coalescence. Those droplets collided without a coalescence will bounce away from each other. The probability of coalescence depends on the collision angle of two droplets [41].

The concept of Energy Tower that cools the hot desert air by spraying seawater on the top of the tower and thus generates a downdraft to produce power was proposed by [41]. In this paper, the collision frequency between two droplets with different velocity was expressed as:

$$\Theta = N_1 N_2 \pi (d_1/2 + d_2/2)^2 |v_{w,1} - v_{w,2}|$$
(24)

where *N* is the number of droplets per unit volume, *d* is the droplet diameter, and the subscript 1 and 2 indicate different diameters of the droplets. The probability of coalescence is set to be 1/2. In this energy tower, the calculation indicates that the model with collision consideration produced only 6% lesser energy (or the velocity of thedowndraft) in this 1000m-high tower compared with no collision consideration.

To simplify the mathematical process, the so-defined Sauter diameter was proposed by Makkinejad [42] to replace all the droplets with different sizes by a population of spherical uniform-size droplets in the tower (thus no collision would happen). The equalized spherical droplet diameter is  $d_s = \sum_i d_i^3 / \sum_i d_i^2$ . It was found that this model produces small deviations (<2.1%) in an industrial counter cooling tower. Therefore, a population of spherical uniform-size drops was assumed in the following section.

344

345 4.2 Heat and mass transfer between water droplets and air

346

Assumptions listed below are also suggested in this article:

347 1). The process is under the steady-state, which means the variables of the process
348 are unchanging in time under a given condition. The initial situation (a transient state
349 or a start-up period) is not considered;

350

351

2). One-dimensional compressible flow in the tower without crosswind;3). The distribution of water and air mass flow is uniform inside the tower;

352 Since the freezing process of seawater can be divided into four stages, the tower 353 will also be separated into sections to calculate the parameters of the air. But it should 354 be noticed that the recalescence stage will not happen in the freely falling seawater 355 droplets because the nucleation temperature is slightly higher  $(0.3\% \sim 0.4\%)$  than the 356 freezing temperature [17]. Then the tower is divided into two parts: i) in the upper part, 357 the water droplets still stay above the freezing point. Heat and mass transfer happen 358 simultaneously in this part; ii) in the lower part, because of the solid shell, the mass 359 transfer from the water to the ambient air can be neglected when the ice is formed 360 quickly around the surface.

361 Llano-Restrepo [28] proposed a set of equations to numerically calculate the heat 362 and mass transfer between water and air in a spray cooling tower neglecting the 363 radiative heat transfer. Those equations transform the temperature gradient against the time (s) to against z (m) by introducing L and A to substitute the  $m_d$  and  $A_d$ , 364 respectively. In the following equations, z is the vertical distance from the bottom of 365 366 a cooling tower upward,  $G_B$  is the mass flow rate of dry air per unit cross-section area of the cooling tower  $(kg/m^2 \cdot s)$ , L is the water mass flow rate per unit cross-section 367 area of the cooling tower  $kg/m^2 \cdot s$ , A is the interfacial area per tower unit 368 volume  $(m^2/m^3)$ ,  $A = N \cdot \pi d^2$ , and N is the number of water drops falling through a 369 unit volume per second,  $N = 6L/\rho_w \pi v_w d^3$ . With the addition of the radiative heat 370 371 transfer, those equations were given below, a more comprehensive analysis can be 372 found in [28]:

373 for the humidity mass ratio,  $Y_w$  is:

$$\frac{dY_w}{dz} = h_{mass} (Y_{w,i} - Y_w) A / G_B$$
(25)

375 for the water mass flow rate, L is

$$\frac{dL}{dz} = h_{mass} (Y_{w,i} - Y_w) A \tag{26}$$

377 for the air temperature,  $T_a$  is

376

$$378 \qquad \frac{dT_a}{dz} = \frac{h_{conv}A(T_w - T_a) + \varepsilon\sigma(T_w^4 - T_a^4)A + h_{mass}(Y_{w,i} - Y_w)A\int_{T_a}^{T_w} C_{p,w}^g dT}{G_B(C_{p,B} + Y_w C_{p,w}^g)}$$
(27)

379 for the water temperature,  $T_L$  is

380 
$$\frac{dT_{w}}{dz} = \frac{h_{conv}A(T_{w} - T_{a}) + \varepsilon\sigma(T_{w}^{4} - T_{a}^{4})A + h_{mass}(Y_{w,i} - Y_{w})A\Delta H_{v}}{LC_{p,w}^{L}}$$
(28)

381 Equations 25, 26 and 28 can be used for water droplets before the phase change 382 stage in the tower (it should be noticed that the minus sign was not shown in Equations 383 26 and 28 because of the differential direction). During the phase change stage, one can easily transform Equations 19-21 from  $\frac{dT_w}{dt}$  to  $\frac{dT_w}{dz}$ . But the Equation 27 that treated 384 385 the air as incompressible gas without considering the variations of air density and air 386 pressure in the cooling tower is not suitable for the model in this paper, because the 387 continuous density change of air in this model is the main reason for the formation of 388 natural draught during the seawater droplets freezing. And the volume of airflow 389 dominates the maximum quantity of seawater that can be frozen in the tower (see the 390 discussion section).

To solve the one-dimensional compressible airflow in a high tower, a method for calculating this buoyant flow induced by the variation of air density was proposed in [43], validated by [44] and further developed by [45]. But the forces that coupling between discrete water droplets and continuous air were not considered in those models, which are not neglected in the model of this paper. The new method considering the forces from water droplets and the heat/mass transfer between water and air is expressed into two parts according to the freezing stage of the droplets:

**398** 1). During the phase change stage of the seawater droplets:

**399** The state equation of air:

400

403

$$\frac{dP}{P} - \frac{d\rho_a}{\rho_a} - \frac{dT_a}{T_a} = 0$$
<sup>(29)</sup>

401 The continuity equation of air after the water droplets fell below freezing402 temperature:

$$d(\rho_a v_a S) = 0 \Rightarrow \frac{d\rho_a}{\rho_a} + \frac{dv_a}{v_a} = 0$$
(30)

404 The momentum equation of air:

405 
$$dP + \rho_a v_a dv_a + \rho_a g dz + \frac{\lambda}{D} \cdot \frac{\rho_a {v_a}^2}{2} \cdot dz + F_w dz = 0$$
(31)

406 where  $\lambda$  is the friction coefficient on the wall of the tower,  $\lambda = 0.008428$  [43], 407  $F_w$  is the source term of momentum due to the forces of water droplets. 408 The  $F_w$  was given in [46] as:  $F_{w}dz = \frac{dv_{w}}{dt}\frac{m_{w}dt}{d\forall}dz = \frac{dv_{w}}{dt}\frac{L\cdot S\cdot dt}{d\forall}dz = \frac{dv_{w}}{dt}\frac{L}{dz/dt}dz = \frac{dv_{w}}{dt}\frac{L}{v_{w} + v_{a}}dz \quad (32)$ 409 410 where  $m_w$  is the water mass flow rate kg/s,  $d\forall$  is the control volume, S is the 411 cross-section area of the tower  $m^2$ . To solve those equations, two more numbers were introduced: 412 413 Mach number is defined as the ratio of local velocity v and the speed of sound:  $M = \frac{v_a}{\sqrt{\nu RT}}$ (33)414 415 where  $\gamma$  is the specific heat ratio of air,  $\gamma = C_{p,a}/C_{v,a}$ . 416 Froude number is defined as the ratio between inertial and gravity force:  $F_r = \frac{v_a}{\sqrt{aD}}$ (34)417 418 This momentum equation can be expressed finally as:  $\frac{dP}{P} + \gamma M^2 \frac{dv_a}{v} = -\gamma M^2 \cdot \frac{2}{\lambda F^2} \cdot \frac{\lambda}{2D} dz - \gamma M^2 \cdot \frac{\lambda}{2D} dz - \frac{F_w dz}{P}$ 419 (35)420 To make the formulae read easily, a shorthand for the right-hand side of Equation 421 35 is introduced as MoX. 422 The energy equation of air per unit mass: 423  $dW - dQ + dH + v_a dv_a + gdz = 0$ (36)where W is the work, Q is the heat transferred during the height of dz, H is the 424 425 enthalpy. 426 Since the work is zero, this equation can be rewritten as:  $C_n dT_a + v_a dv_a + gdz = dQ$ 427 (37)428 With neglecting mass transfer:  $dQ = dQ_{ice} = \frac{[q_{conv} + q_{rad}]A - \pi Dq_{wal}}{C}dz$ 429 (38)where  $dQ_{ice}$  is heat transferred from water droplets during the phase change stage. 430  $q_{wal}$  is the heat loss of the air through the tower wall and can be expressed as [47]: 431  $q_{wal} = h_{wal} \left( T_a - T_{T_a} \right)$ (39)432  $Nu_{wal} = \frac{h_{wal}D}{k} = 0.023P_r^{0.3}R_e^{0.8}$ 433 (40)where  $h_{wal}$  is the heat transfer coefficient, and  $T_{z,\infty}$  is the temperature of the 434 435 atmosphere at the height z.

436This energy equation can be expressed finally as:

437 
$$\frac{\gamma}{\gamma - 1} \cdot \frac{dT_a}{T_a} - \frac{dP}{P} = \gamma M^2 \cdot \frac{\lambda}{2D} dz + \frac{F_w dz}{P} + \frac{\rho_a dQ_{ice}}{P}$$
(41)

438 To make the formulae read easily, a shorthand for the right-hand side of Equation

439 41 is introduced as EX.

440 After solving Equations 29, 30, 35, and 41 above, we get:

441 
$$\frac{dT_a}{T_a} = \frac{\gamma - 1}{\gamma (M^2 - 1)} \cdot [(\gamma M^2 - 1) \cdot EX - MoX]$$
(42)

442 
$$\frac{dv_a}{v_a} = \frac{1}{\gamma (M^2 - 1)} \cdot [MoX - (\gamma - 1) \cdot EX] = -\frac{d\rho_a}{\rho_a}$$
(43)

443 
$$\frac{dP}{P} = \frac{\gamma - 1}{M^2 - 1} \cdot [M^2 \cdot EX - MoX/(\gamma - 1)]$$
(44)

444  $\frac{dT_a}{dz}, \frac{dv_a}{dz}$ , and  $\frac{dP}{dz}$  can be calculated from Equations 42, 43 and 44.

2). When the water droplets are still above freezing temperature, the continuityequation of air can be rewritten as:

447 
$$d(\rho_a v_a) = dL \Rightarrow \frac{d\rho_a}{\rho_a} + \frac{dv_a}{v_a} = \frac{dL}{\rho_a v_a} = \frac{h_{mass}A(Y_{w,i} - Y_w)}{\rho_a v_a} dz$$
(45)

448 The energy equation is:

$$\frac{\gamma}{\gamma-1} \cdot \frac{dT_a}{T_a} - \frac{dP}{P} = \gamma M^2 \cdot \frac{\lambda}{2D} dz + \frac{F_w dz}{P} + \frac{\rho_a dQ_w}{P}$$
(46)

450 
$$dQ_w = \frac{A[q_{conv} + q_{rad} + q_{evap}] - \pi Dq_{wal}}{G_B} dz$$
(47)

451 where  $dQ_w$  is heat transferred from water when water stayed above freezing 452 temperature.

To make the formulae read easily, a shorthand for the right-hand side of Equation46 is introduced as EXw.

455 After solving Equations 29, 35, 45 and 46 above, we get:

456 
$$\frac{dT_a}{T_a} = \frac{\gamma - 1}{\gamma (M^2 - 1)} \cdot \left[ (\gamma M^2 - 1) \cdot \left( EXw + \frac{dL}{\rho_a v_a} \right) - \left( MoX - \frac{dL}{\rho_a v_a} \right) \right]$$
(48)

457 
$$\frac{dv_a}{v_a} = \frac{1}{\gamma(M^2 - 1)} \cdot \left[ \left( MoX - \frac{dL}{\rho_a v_a} \right) - (\gamma - 1) \cdot \left( EXw + \frac{dL}{\rho_a v_a} \right) \right]$$
(49)

449

$$\frac{dP}{P} = \frac{\gamma - 1}{M^2 - 1} \cdot \left[ M^2 \cdot \left( EXw + \frac{dL}{\rho_a v_a} \right) - \left( MoX - \frac{dL}{\rho_a v_a} \right) / (\gamma - 1) \right]$$
(50)

459  $\frac{dT_a}{dz}, \frac{dv_a}{dz}$ , and  $\frac{dP}{dz}$  can be calculated from Equations 48, 49 and 50.

460 To validate this new method proposed in this paper, the temperatures calculated 461 by the present method are compared with those reported by Zarling [23] in Fig. 5. As 462 shown by Fig. 5, the variations of water temperature calculated by this new method are 463 in good agreement with the results reported in [25]. The slight difference may be caused 464 by different formulae used in two different methods (e.g. air/water viscosity, density 465 conductivity et al.). It should be noticed that the mass flow ratio of water (L) to air ( $G_B$ ) was set in that paper, but not by calculation. It can be seen that the temperature variation 466 467 of sprayed water is strongly affected by the flow ratio, which is also stated at the



#### 469 470

471 4.3 Energy generated by turbines

The pressure difference between the air inside the tower and the outside is the integral of the product of the difference of the air densities by the gravity acceleration with respect to height:

475 
$$\Delta P_{pot} = g \int_0^h (\rho_{z,\infty} - \rho_z) dz = g \int_0^h \rho_{z,\infty} dz - g \sum_0^h \rho_z \Delta h$$
(51)

476 where h is the tower height,  $\rho_{z,\infty}$  and  $\rho_z$  are ambient air density and internal airflow 477 density inside the tower at any height z, respectively. Although the air density in the 478 tower changes along with the tower height, this change can be neglected as long as the 479  $\Delta h$  is small enough. In this paper, a proper value of 0.2m is selected for  $\Delta h$ .

According to the International Standard Atmosphere [48], environment air
temperature decreases with the altitude increasing. The temperature at altitude *z* meters
above sea level is approximated by the following formula (only valid no more than ~18
km above Earth's surface):

$$T_{z,\infty} = T_{0,\infty} - T_{la}z \tag{52}$$

485 The pressure at altitude z is given by:

$$P_{z,\infty} = P_{0,\infty} \left( 1 - \frac{T_{la}z}{T_{0,\infty}} \right)^{g/RT_{la}}$$
(53)

487 The density of air can be calculated according to the ideal gas law:

488 
$$\rho_{z,\infty} = \rho_{0,\infty} \left( 1 - \frac{T_{la}z}{T_{0,\infty}} \right)^{(g/RT_{la}-1)}$$
(54)

489 where:

484

486

- 490  $P_{0,\infty}$  = sea-level standard atmospheric pressure, 101.325 kPa
- 491  $T_{0,\infty}$  = sea-level standard temperature, 288.15 K
- 492  $T_{la}$  = temperature lapse rate, 0.0065 K/m
- 493  $R = \text{air specific constant, } 287.05 \text{ J/ (kg } \cdot \text{K)}$

494 The specific potential energy due to heating is transformed to shaft power by 495 turbines, frictional losses in the tower, and lost to the environment. The total pressure 496 losses in the tower are the pressure potential minus  $\Delta P_{turb}$  [49]:

497 
$$\Delta P_{loss} = \Delta P_{pot} - \Delta P_{tur} = \Delta P_{pot} - \zeta \Delta P_{pot}$$
(55)

$$\Delta P_{loss} = \Delta P_{tur,in} + \Delta P_{br} + \Delta P_k \tag{56}$$

499 with

498

500

$$\Delta P_{tur,in} = \varepsilon_{tur,in} \cdot \frac{1}{2} \rho_{a,in} v_{a,in}^2 \tag{57}$$

501 
$$\Delta P_{br} = \varepsilon_{br} \cdot \frac{1}{2} \rho_{a,in} v_{a,in}^2$$
(58)

502 
$$\Delta P_t = \varepsilon_t \cdot \frac{1}{2} \rho_{a,out} v_{a,in}^2$$
(59)

503 
$$\Delta P_k = \varepsilon_k \cdot \frac{1}{2} \rho_{a,out} v_{a,out}^2$$
(60)

504 where  $\Delta P_{turb}$  is the pressure drop at the turbine,  $\zeta$  is the turbine pressure drop factor which is defined as the ratio of the pressure drop at the turbine to the total pressure 505 506 potential. The ratio  $\zeta$  is proposed to be 0.8 in [49], based on the previous work. In [49], 507 the pressure losses are suggested as:  $\Delta P_{turb,in}$  is the turbine inlet pressure loss with  $\varepsilon_{turb,in} = 0.14$ ;  $\Delta P_{br}$  is the pressure loss due to the internal bracing wheel (for 508 509 strengthening the tower) drag forces with  $\varepsilon_{br} = 0.25$ ;  $\Delta P_k$  is the pressure loss due to 510 exit kinetic energy loss with  $\varepsilon_k = 1.26$ .  $\Delta P_t$  is the air collector-to-chimney (airflow from horizontal to vertical) transition section pressure loss with  $\varepsilon_t = 0.268$  [45]. 511 512  $v_{a,in}$  and  $v_{a,out}$  represent the velocity of air at the inlet and outlet of the tower 513 respectively.

According to [45], the airflow through the turbine can be treated as incompressible air, which is accurate enough for a practical purpose. This indicates that the difference in air density at the inlet and the outlet of the turbine can be neglected.

517 The power extracted from the turbine generators under a turbine load condition518 can be expressed as:

519

$$Pow_t = \eta \cdot \Delta P_{tur} \cdot S \cdot v_{a,in} \tag{61}$$

520 where  $\eta$  is the mechanical efficiency of the turbine generators, which is proposed to 521 be 77% in [49].

522

# 523 5. Calculation procedure

- 524 The whole calculation process is divided into three steps:
- 525 Step 1: A guessed value for seawater quantity that the desalination system could

handle was given firstly, to calculate the corresponding flow of the cold air flowing into
the tower. The differential equations were calculated from the bottom of the tower to
the top of it.

529 Step 2: As the mass of the cold air induced into the tower by the pressure difference 530 is not known, a guessed value for cold air mass flow rate was given for the iteration 531 process. The Runge-Kutta fourth-order method [50] was applied to solve the Equations 532 25, 26, 28, 48, 49 and 50 simultaneously for the upper part in the tower (before the 533 phase change stage of the seawater). While for the lower (ice) part in the tower the Equations 26, 42, 43, 44 and the Equations 19-22 transformed from  $\frac{dT_w}{dt}$  to  $\frac{dT_w}{dz}$  need 534 535 to be solved simultaneously. When these equations were solved, the temperature 536 gradient against tower height was known. Then the mass of cold air can be calculated by Equations 51, 55 and 56. This value can be used as a reference value with the initial 537 guessed value together to get a new  $G_B$  by Newton iteration method. 538

539 Step 3: This new  $G_B$  was used to recalculate the temperature variation of water 540 droplets. If the water temperature on the top of the tower differed from the set 541 temperature of 2°C, a new water mass flow rate would be suggested. This new water 542 mass flow rate would be given to repeat step 1.

The final results would be obtained when the setting conditions were satisfied after
steps 1 to 3. The whole calculation process is shown in the following figure.





Fig. 6. The whole calculation process in this paper

548

# 549 6. Results and discussion

550 The experiments conducted by Gao [18, 51] showed that more than 60% of 551 impurity in the water was removed in the ice formed by the spray freezing. The ice was 552 about 70% of the total volume of feed water, while the rest was separated by gravity 553 and released as runoff. It was also demonstrated that a lower runoff fraction would 554 deteriorate the rate of impurity reduction because more brine pocket would be 555 entrapped in the ice powder. In another freeze desalination experiment [52], a mesh and 556 a filter cloth were used for gravity filtration of an ice slurry (a mixture of 1mm fine ice 557 grains and brine, ice fraction is more than 70%). Approximately 60% of the total feed 558 water was generated as the fresh water (at 0.5% NaCl concentration) after 50 min 559 filtration. Although the salinity of the generated water could be further lowered by 560 centrifugation, it is not discussed in this paper due to its energy consumption. From a 561 conservative point, the ice fraction after the spray freezing process was set to be 70%, 562 and 60% of the total feed water was assumed to be generated as fresh water. Thus, the 563 final temperature of water droplets is set to be -7.15°C based on Equation 21. The initial 564 seawater temperature was set to be 2°C, and the relative humidity of the atmosphere 565 was 80% [23].

566

## 567 6.1 Impact of environmental temperature on system performance

The atmospheric air temperature variation can greatly influence system performance. A spray freezing system with a 200*m* high tower under three different surrounding environmental temperatures is analyzed first. The 2mm is chosen as the Sauter diameter of the water droplets sprayed in the tower. It is reasonable because more than 80% of the droplets had a diameter between 1mm to 2.8mm in the experiments conducted by Gao [18, 51].

574 The variations of the water temperature are shown in Fig. 7. The typical 575 atmospheric temperature in the cold regions discussed in references is -26°C, -18°C, and 576 -10°C [17, 23, 30], those figures are also chosen in this paper. As stated above, the 577 water temperature at the top of the tower is set to be 2°C and it is set to be -7.15°C (70% 578 ice fraction) at the bottom. The longer distance of the water required to reach the 579 freezing point at -26°C is because of the larger mass flow ratio of feed water (L) to air 580  $(G_B)$  in that condition. More water needs longer distances to freeze when the air mass 581 flow rate is smaller. The seawater requires a distance of 10.5m, 8.3m and 5.0m to reach 582 the freezing point at -26°C, -18°C, and -10°C, respectively and the corresponding mass 583 flow ratio of feed water to air is 0.089, 0.053, and 0.019, respectively (see Table 2).

Although a higher mass flow rate of the water releases more heat to the air, more water droplets also hamper the airflow in the tower due to the drag forces between the water droplet and the air. Thus, the mass flow rate of the induced air would not increase proportionally with the increase of the water quantity.

588 When the ice is developed on the surface of the droplet, higher concentrated water 589 remains inside the droplet, leading to the reduction of its freezing point. Thus, the 590 temperature of the droplets continuously decreases until it reaches 7.15 °C. The 591 temperature decrease of water droplets is faster at -26°C at the lower half part of the 592 tower (between the 0m to about 100m above the bottom) because of the large 593 temperature difference between the cold air and water. At the height above 100m, the 594 change of water temperature becomes smooth due to the small temperature difference 595 between the cold air and water.





Fig. 7. The temperature variations of water droplets during the freezing process in the system.

598

599 The temperature and density variations of air in the tower are presented in Fig. 8 600 and 9, respectively. Figs 8 and 9 show that a higher temperature difference between the 601 air and water creates a higher heat transfer rate, a faster air temperature, and a higher 602 rate of density change, especially during the lower half part of the tower. The final 603 temperature of the air at the outlet of the tower is higher under the condition of the 604 colder atmosphere, which is caused by a larger mass flow ratio of feed water (L) to air  $(G_R)$  in that condition. This feature is also reflected by the variations of density in Fig. 605 9. The final density of the air is 1.281, 1.288, and 1.293  $kg/m^3$  at the atmospheric 606

temperature of -26 °C, -18 °C, and -10 °C, respectively. The corresponding final
temperature of the outlet air (200m above the bottom) is 267.7 K, 266.7 K, and 266.1 K,
respectively.



611 Fig. 8. The temperature variations of the induced air in the tower at different atmospheric temperatures612



613

610

Fig. 9. The density variations of the induced air in the tower at different atmospheric temperatures615

616 The main results of the freeze desalination system under different atmospheric617 temperatures are presented in Table 2. More air was induced into the tower to freeze

618 the sprayed water droplets in the colder atmosphere, which generated more fresh water. 619 At the same time more power was generated by the turbine. The maximum of the 620 generated fresh water flow rate was 27.7 kg/s at a temperature of -26°C. In this table, 621 the fresh water generated was set to be 60% of the feed water and the mechanical 622 efficiency of the pump for spray was set to be 0.85. The power consumption was 623 calculated by  $Pow_p = 0.85m_wgh$ . The third column was the result from the second 624 column multiplied with the cross-section area S and the fresh water generation rate 625 60%.

#### Table 2

Τ	Feed water flow rate	Fresh water generated	Airflow rate	Power generation/
Temperature	$(kg/m^2 \cdot s)$	(kg/s)	$(kg/m^2 \cdot s)$	consumption
-26°C	0.587	27.70	6.62	30.35%
−18°C	0.243	11.45	4.58	25.96%
−10°C	0.038	1.80	2.00	14.66%

626

627 6.2 Impact of droplets' diameter on system performance

628 The variations of droplets' diameters can also greatly influence system 629 performance. In the following discussion, the temperature of the surrounding 630 atmosphere is set to be -18°C. The impact of droplets' diameters on air temperature and 631 density is delineated in Fig. 10 and Fig. 11. As shown in Fig. 10, the air flows upwards 632 from the bottom of the tower with a temperature rising. Compared to larger droplets, 633 the temperature of air contacted with smaller droplets roses sharper at the beginning 634 due to the larger area of heat transfer per unit mass. With the temperature difference 635 between water and air being narrowed down quickly, the change of air temperature 636 becomes slow and reaches a relatively "stable" state. This feature is also reflected by 637 the variation of density in Fig. 11. The final density of the air is 1.265, 1.274, 1.288, 638 1.305 and 1.319  $kg/m^3$  with the diameter of 1, 1.5, 2, 2.5 and 3mm, respectively. 639 The corresponding final temperature of the outlet air (200m above the bottom) is 640 270.9K, 269.5K, 266.7K, 263.4K and 260.5K, respectively. As discussed above, the 641 final temperature of the outlet air would be higher when the droplets with smaller 642 diameter sprayed into the tower, which is caused by the larger mass flow ratio of water 643 to air (see Table 3).





Fig. 10. The temperature variations of the induced air in the tower with different droplets' diameters



- 646
- 647

Fig. 11. The density variations of the induced air in the tower with different droplets' diameters

648 The mass flow rate of the feed water that can be frozen by this system with 649 different diameters of water droplets is present in Table 3. As expected, the water mass 650 flow rate frozen by the system would be larger when smaller droplets sprayed into the 651 tower because the smaller droplets have larger heat and mass transfer area per unit mass. 652 Smaller droplets would also induce more air into the tower and generate more power. 653 Though smaller droplets led to greater output (fresh water and power generated)

- 654 compared to larger droplets, smaller droplets needed greater water pressure produced
- by the pump and were prone to jam in the sprinkler. Too small droplets may endure the
- risk of being blown out of the tower by the updraft air, which resulted in water loss.
- 657 Those advantages and disadvantages of using smaller droplets should be traded off in
- 658 a real project.

# Table 3

Results of the freeze desalination system with different diameters of water droplets

Droplets	Feed water flow rate	Fresh water generated	Airflow rate	Power generation/
diameter	$(kg/m^2 \cdot s)$	(kg/s)	$(kg/m^2 \cdot s)$	consumption
1 <i>mm</i>	0.436	20.54	6.14	34.85%
1.5 <i>mm</i>	0.349	16.44	5.40	29.61%
2mm	0.243	11.45	4.58	25.96%
2.5mm	0.150	7.07	3.75	23.05%
3mm	0.084	3.96	2.96	20.23%

659

From Table 2 and Table 3, it can be observed that the ratio of water mass flow rate to air mass flow rate was adjusted with the variation of the outside parameters. It cannot be set in this natural airflow induced by the pressure difference. The airflow rate is one of the most important factors that affect the fresh water output. The power generated by the air from turbines can compensate as large as one-third of the pump energy consumption, which was not recognized in previous research.

666

# 667 6.3 Limitations

668 Although a new mathematical model has been developed for heat and mass 669 transfer between compressible airflow and water droplets freezing in a tall tower, there 670 were some limitations in this model which could be improved in further study. Firstly, 671 the wind effects were not considered in this paper. Wind from the surrounding 672 environment may significantly affect the performance. It may blow the hot air from the 673 top of the tower down to the bottom. Secondly, in this study water droplets sprayed by 674 nozzle were treated as a population of spherical uniform-size droplets. Actually, the 675 droplets from the nozzle may not be in a uniform shape. The droplets collide and bounce 676 with each other due to the different velocities. The turbulent flow of the air and the 677 momentum transfer during the collision need to be analyzed further.

678

# 679 7. Economic analysis of this spray freeze desalination system

680 In this economic analysis, the case of water droplets with a diameter of 2 mm681 sprayed in the atmospheric temperature of -26°C was taken for comparison with other 682 desalination methods in the following part.

683 Firstly, this new spray freeze desalination method needs lesser power consumption

684 than some traditional methods. The power consumption required to produce 1  $m^3$ 685 fresh water in this system is approximately  $1.07 \ kWh$ , in which pump consumption is 686 the major part of the operational energy cost. Considering the power generated by the 687 turbines, this cost could be even lower by one third. If the altitude of the brackish water 688 is above the height of the desalination system (e.g. the tailing water at a mine), a pipe 689 could be made to channel the water to the tower and then benefit the operational cost of this freeze desalination system. In comparison, to produce 1  $m^3$  fresh water, a 690 typical multi-stage flash distillation method needs 3.5 kWh power [53], the reverse 691 692 osmosis method consumes 2-8 kWh power [7].

693 Secondly, this new spray freeze desalination method is more energy-efficient than 694 other freeze desalination methods because of the high rate of heat and mass transfer 695 between the water droplets and the air. A direct contact type seawater freeze 696 desalination method was developed in [54]. In this experiment, a flow of refrigerant 697 cooled by the cold energy from the regasification of LNG was injected into a seawater 698 tank to generate ice. A technical and economic evaluation of this system was made by 699 [55]. The optimum result of this system was that it generated 1.64 kg/s fresh water 700 by consuming 7.83 kg/s seawater while the cold energy was provided by 1 kg/s701 LNG regasification (about 827 kI/kg energy would be released by the LNG). Thus, 702 the cold energy needed in this system was 504.3 kI per kilogram fresh water. But in 703 the new freezing system proposed in this paper, only 375.4 kJ cold energy (calculated 704 from the enthalpy change of the airflow) was needed for generating one kilogram of 705 fresh water. A freezing crystallizer proposed by Attia et al. [56] needs 420 kJ energy 706 and a disk column freezing crystallizer developed by Van der Ham et al. [15] needs 707 1037-1282 kJ energy to produce 1 kg fresh water. From this point, if there is free 708 cold energy, the cold air (from the atmosphere or produced during the regasification of 709 LNG) can be utilized in the desalination system proposed in this paper which is more 710 energy-efficient than previous desalination systems discussed above.

711 Finally, according to the statistics data in [55], the price is about 1.5 USD for 1 712  $m^3$  freshwater and the industrial electricity price is about 0.15 USD for 1 kWh power. 713 The total fresh water generated by this system is 873,547  $m^3$  per year under the 714 assumption of no interruption of the free cold energy supply. The profit calculated by 715 deduction between the fresh water income and the pump cost is about 1,179,288 USD 716 per year (1,218,598 USD per year with power generated by the turbine). While the 717 construction cost of a 200m tower is about 5-10 million USD [57] depended on the 718 location, labor force cost and the materials of the tower. Then the feedback period of 719 this desalination system is about 4-8 years.

720

# 721 8. Conclusions

722

This paper investigated the feasibility of using spray freezing mechanism in a cold

environment to desalinate seawater. Because of the low energy consumption, no need
for huge pressure or membrane replacement work, and ignorable corrosion issues,
freeze desalination is an attractive method to desalinate the seawater. While, the spray
freezing method in this paper is more energy-efficient than the block, layer or falling
film freeze processes because it has higher heat and mass transfer area per unit seawater
and lower heat resistance.

729 In this paper, the freezing character of a freely falling seawater droplet was 730 analyzed first. The freezing process of a droplet was divided by several stages, the heat 731 and mass transfer of the droplet in each stage was investigated. An improved mathematical method was proposed to numerically simulate this freezing process. Then 732 733 the heat, momentum, and mass transfer between a system of water droplets sprayed in 734 the desalination system and the natural convective airflow induced by the hot water 735 were studied. An iterative process to solve the proposed differential equations was 736 introduced in this paper.

737 The results demonstrated the seawater desalination capacity was affected by the 738 surrounding atmospheric temperature and the diameter of the water droplets. Colder 739 atmospheric temperature and smaller droplets produce more fresh water. The 200m 740 high desalination system could generate 27.7 kg/s fresh water (at 0.5% NaCl 741 concentration) with the 2mm diameter of the sprayed water droplet in the atmospheric 742 temperature of -26°C. The induced natural convective airflow could generate one-third 743 of the energy consumption of the water pump in this system, while this free energy was 744 never respected in previous research.

745 This new spray freeze desalination method consumes lesser power. The power 746 consumption required to produce 1  $m^3$  fresh water in this system is about 1.07 kWh. 747 With consideration of the power generated by the wind turbines, this cost could be even 748 lower by one third. Compared with some traditional desalination methods, the newly 749 proposed spray freeze desalination method consumes less power while the same mass 750 flow rate of fresh water is generated. This new method is also energy efficient compared 751 to other freeze desalination approach because of the direct contact between the cold and 752 warm flow (low heat resistance) and the large area of heat and mass transfer per unit 753 mass of water. It needs only 375.4 kJ cold energy to produce one-kilogram fresh water.

The spray freeze desalination process can generate fresh water and produce green power simultaneously. It deserves to be considered by the desalination industry due to its high efficiency if free cold energy is available. The economic analysis of the system in different countries needs further study based on the local water, labor, electricity and land prices in the next step.

759

## 760 Acknowledgements

761 This study is financially supported by the National Natural Science Foundation of

- 762 China (Grant No. 51778511), the Hubei Provincial Natural Science Foundation of
- 763 China (Grant No. 2018CFA029), the Key Project of ESI Discipline Development of
- 764 Wuhan University of Technology (WUT Grant No. 2017001), the Fundamental
- 765 Research Funds for the Central Universities (WUT Grant No. 2019IVB082), and the
- 766 Scientific Research Foundation of Wuhan University of Technology (No. 40120237).
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**<sup>885</sup>** 670-6.

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# **Conflict of interests**

 $\boxtimes$  The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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