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The Method for Effective Co_2 Purification in the Atmosphere

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The Method for Effective Co_2 Purification in the Atmosphere

Svetlana Amirova [α] & Tamara Tulaikova [σ]

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I. Introduction

The idea of weather modification by precipitation seeding was generated earlier (Langmuir, 1937). The most popular methods used today for precipitation enhancement are the sprinkling of hygroscopic particles or a special solutions for 'warm' clouds and the introduction of glaciogenic substances into 'cold' clouds from an airplane (Mather 1997, Shmeter 2005, Drofa 2006) and etc. According to IPCC Fifth Assessment Report the predicted climate trend indicates the global overheating (Edenhofer, 2014) up to the end of century, and all known natural restore mechanisms have limited capacities in regards to the amount of incoming pollution and also they operate within specific time constants according to (Kleidon, 2010). We recall that the sum of water vapor and carbon dioxide makes a 95% of the greenhouse gases in a modern atmosphere. Mentioned aspects we used to propose new method for atmosphere purification and further climate recovery.

Our approach incorporates the possibility of stepwise CO_2 purification in two simple stages to be conducted in areas of the free atmosphere. The first stage involves spraying of alkaline compounds, such as *KOH* and etc., inside the cloud via an airplane to increase the *pH* in cloud droplets. The alkaline reagents significantly increase the solubility of co_2 in water, however rain droplets become saturated by atmospheric CO_2 during their gravitational fall. Due to the small co_2 concentration in air the probability of collision in cloud between small water droplets and co_2 molecules is low, so modified cloud droplets spend their alkali capacity later during rain. The rainy droplets provide effective transport of co_2 from the under-cloud atmospheric volumes to the ground and further more to soil, ground water and plants.

In addition to it, the special acoustic devices can be utilized to accelerate the coalescence of water droplets in modified clouds as possible second stage of proposed method according to (Tulaikova, 2010). The acoustically influencing for mists and fogs was widely used early (Boucher, 1960, Mednikov 1963) then industrial aerosols were the central purpose for the acoustical seeding, and common practice show the efficiency up to 99%. The background is the facts that acoustics provides high mobility for droplets, because they become be involving into air vibrations inside sound waves with the best coalescence as a results. The speed of acoustic-based vibrations and coalescence should be effective for cloud droplets due to high speed of acoustic waves (C ~ 340 m/sec) in comparison with typical atmosphere winds 2 – 20 m/sec. Previous acoustical experiments were only carried out near the ground earlier, but it could be most effective to place the sound generator directly inside a cloud in the region with oversaturated water steam by using a modern airplanes or helicopter.

II. The Features of Proposed Method for CO_2 Purification

The purification effect strongly correlates with changes of *pH* level in cloud water. In natural precipitation with acid or neutral *pH* the concentrations of the dissociated ions are relatively small. On the other hand, the natural ocean of our planet stores great mass of CO_2 due to alkali properties of the ocean water where is $pH \approx 8.3$. There is an established method to describe the insoluble and dissociated portions of the weak acid that remains after attaining the equilibrium of saturation for water by CO_2: $CO_2 + H_2O \Leftrightarrow H_2CO_3$, $H_2CO_3 \Leftrightarrow HCO_3^- + H^+$, and $HCO_3^- \Leftrightarrow CO_3^{2-} + H^+$. The equilibrium concentrations of dissolved and dissociated portions became $[H_2CO_3] = C_1 = 0.71$ mg/l; $[HCO_3^-] = C_2 = 3.3$ mg/l, also $[CO_3^{2-}] = C_3 = 10^{-3}$ mg//then $pH = 7$, and given data were listed for a cloud with 3 mkg/m^3

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of NH_3 (Yunge 1965, Rasool 1073). Therefore, co_2 solubility significantly grows at higher pH levels due to increase of H^+ concentration, and concentrations of ions $[HCO_3^-]$ and $[CO_3^{2-}]$ increase in 10 and 100 times accordingly by each unit of pH. The ratios of carbon in the first, second and third of the listed compounds are the following: $D_1 = 0.1935$; $D_2 = 0.1967$; and $D_3 = 0.20$. For given water volume U_w the carbon mass can be calculated as:

$$M_C^{Uw}(pH) \approx \left[\ 0.1935 \cdot C_1 + 0.1967 \cdot C_2(pH) + 0.2 \cdot C_3(pH)\ \right] \cdot U_w \quad (1)$$

For instance, a precipitation layer with a height of $h_w = 1000$ mm covers a unit surface of 1 m^2, so the corresponding mass of carbon is equivalent to $M(10) = N_S(10) \cdot 1000 \approx 850$ g at the $pH = 10$. Using this approach one can estimate the concentration of CO_2 that is removed from the atmosphere underneath the cloud by precipitation. Let's take the volume of purified air in the atmosphere $U_a = 10^3$ m^3 in precipitation, there $h_a = 1$ km is the altitude of the cloud calculated from the ground for a unit square surface 1 m^2. The reduced CO_2 concentration C_{a-} could be estimated by dissolving the carbon mass in alkaline precipitation water:

$$C_{a-} \approx \frac{0.1935 \cdot C_1 + 0.1967 \cdot C_2(pH) + 0.2 \cdot C_3(pH)}{0.2727} \cdot \frac{h_w}{h_a} \quad (2)$$

The molar mass of CO_2 is 44 g, so the share of carbon in it is $D_4 = 12/44 = 0.2727$. The vertical CO_2 distribution was considered here almost uniform (Machida, 2003) up 2-3 km, excluding increase in 100-200 meters near ground. For the special case of the complete atmospheric purification in the volume under cloud, we assume that the CO_2 concentration is $C_{a-} \approx$ 420 ppm. The pH level of the droplet should be increased up to pH=10.3 for complete purification if the cloud located at 1 km above the ground. In general, clouds can be located at various altitudes h_a. For instance in case of the altitude $h_a = 6$ km the cloud medium should become pH=10.8 to get complete purification in lower area. Let's analyze a chemical approach to introduce KOH into clouds in the form of additional liquid aerosol. There is the dissociation reaction of KOH in water:

$$KOH \rightarrow K^+ + OH^-$$
$$H^+ + OH^- \Leftrightarrow H_2O \ , \ K_w = [H^+] \cdot [OH^-] \quad (3)$$

We note that the values of water dissociation constant are taken at variable temperatures: $K_w = 10^{-14}$ at $T = 20°C$ according to (Sillen 1964) or $K_w = 10^{-14.926}$ at $T = 0°C$ in cloud. The required KOH mass can be obtained approximately from the relation $n_{KOH} = K_w / [H^+] = [OH^-]$ according to algorithm of (3). Taking the equality of molar concentrations for KOH and OH from relation (3) we obtain the molar concentration of alkali $n_{KOH} = 10^{-4}$ mol/l then $pH = 10$, for example. The mass concentration value $N_{KOH} = 56 \cdot 10^{-4}$ g/l $\approx 5.6 \cdot 10^{-6}$ g/cm^3 results from molar one by multiplication to the molar mass of KOH. Let's comment upon the data. Suppose in a cloud the liquid water content is $W = 1$ g/m^3, it means that air volume 1 m^3 contains the 1 cm^3 of water, and listed data of N_{KOH} corresponds required alkali mass to air volume 1 m^3 ($5.6 \cdot 10^{-9}$ kg/m^3) to get mentioned pH-level in cloud droplets after evaporation/condensation and restructuring. For example, let the cloud has a volume 1 km^3 and $W = 1$ g/m^3, hence the required mass of KOH to add into this cloud should be equal to $5.6 \cdot 10^{-9} \cdot 10^9 \approx 5.6$ kg then $pH =$ 10 approximately. Exact calculations for KOH mass were performed with account of condensation processes onto aerosol particles in real cloud (Tulaikova, 2012). The average KOH concentrations as a function of pH are $N_{KOH} = 1.7 \cdot 10^{-5}$ g/cm^3 then pH=10.5, and $N_{KOH} = 3.5 \cdot 10^{-5}$ g/cm^3 then pH=10.8 in comparison with $N_{KOH} = 5.6 \cdot 10^{-6}$ g/cm^3 then pH=10.

We investigate a potential purification advantage of this method based or droplets assemble above the flat ocean surface. Firstly, due to small size of rain droplets the falling time exceeds the gas saturation time to provide perfect CO_2 absorption. The fall velocity $V_g(r)$ can be using the stationary speed of droplet gravitation sedimentation as follows: $V_g(r) \approx \sqrt{2rg\rho_w / \rho_a}$, where ρ_a and ρ_w are the densities for air and water respectively, r is a drop radius and g is the acceleration due to gravity. The falling time for droplet can be estimated as $t_h \approx h_a / V_g(r)$ with the initial cloud altitude of $h_a = 1$ km, these calculations show $t_h = 257$ sec then $r =$ 1 mm for example. On the other hand, we can estimate the saturation time t_{aw} for the falling droplets during the process of CO_2 solubility. The steady state concentration depends on the ratio of the air volume $U_r = 3\pi r^3/3$ for the drop and the area of its surface $S_r = 4\pi r^2$, and also depending on the gas exchange constant K_{aw} at the gas-water interface, the time of saturation is $t_{aw} \approx r/3K_{aw}$. The constant $K_{aw}(V)$ has been measured as a function of the velocity of air flow V over the water surface, according to (Broecher 1978, Wanninkhof 1996). We employed the following experimental measurements for constants of gas exchange at the gas-liquid interface: $K_{aw} \approx 4 \cdot 10^{-3}$ cm/s then $V = 3$ m/s; also $K_{aw} \approx 1.1 \cdot 10^{-2}$ cm/s if $V = 7$ m/s. Taking into account $K_{aw}(V)$ we obtain saturation time $t_{aw} = 6$ sec for droplet with a typical rain radius $r = 1$ mm. The largest radius in rain has been estimated as 3 mm due to drop disintegration, and at the beginning of the precipitation due to gravitational sedimentation the droplets have grown with radius $r \geq 100$ μm. For the largest droplet with radius $r = 3$ mm saturation time is $t_{aw} = 11$ sec and flying time $t_h = 148$ sec due to velocity

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$V_g \approx 7$ m/sec. As $t_{aw} << t_h$ there is enough time for gas saturation during droplet flight to the ground from the altitude $h \geq 1$ km.

The next method's advantage is great increases of the air/water interface in droplets assemble in comparison with flat surface of ocean. Further analysis bases on the Marshall-Palmer approximation for the droplets spectrum for rain as follows $\varphi(r) = n \cdot b \cdot \exp(-br)$. This empirical formula includes the constant b (cm^{-1}), and the droplet number concentration n (cm^{-3}) that depends on the precipitation rate I, in millimeters per hour (Borovikov 1961, Kobayashi 2007). Different types of precipitations can be described by previous empirical equation for the parameters b and n using in, as follows: drizzle has $b = 57 \cdot I^{-0.21}$ with $n = 5 \cdot 10^{-3} \cdot I^{0.21}$; rains have $b = 41 \cdot I^{-0.21}$, $n = 2 \cdot 10^{-3} \cdot I^{0.21}$; and storms have $b = 30 \cdot I^{-0.21}$ with $n = 5 \cdot 10^{-4} \cdot I^{0.21}$. The radii interval for the complete rain spectrum was calculated over a wide interval $r = 0.05$ - 3 mm. The volume of received precipitation water $U_I = AIT\beta$ was calculated as a sum of all of the falling droplets, there the water layer is I on a ground surface $A = 1$ m^2 during $T = 1$ hour for all calculations as mentioned in the examples. Here rates for drizzle, rain or storm are $I = 3$; 10 or 30 mm/hour, , but the β coefficient was introduced in order to compare the features for the precipitation types, $\beta_1 = 1$, $\beta_2 = 0.3$ and $\beta_3 = 0.1$, so $I_1\beta_1 = I_2\beta_2 = I_3\beta_3$. Then the percentage of droplets with a radius r_i within the unit water volume can be described using normalized number of droplet $q_i(r_i) = \varphi_i(r_i)/\Sigma\varphi(r)$. The sub-volume of the unit volume $U_I(r_i)$ for these droplets with equal radii can be estimated as $q(r_m) \cdot U_I$, also the number of droplets in each sub-volume consist of $N_m = 3q(r_m)U_I/(4\pi r_m^3)$. The total sum for the entire droplet intervals in all volumes of sub-volumes $U_I(r_i)$ are described by the following sum:

$$N = \frac{3AIT\beta}{4\pi} \sum_r \frac{q(r)}{r^3} \qquad (4)$$

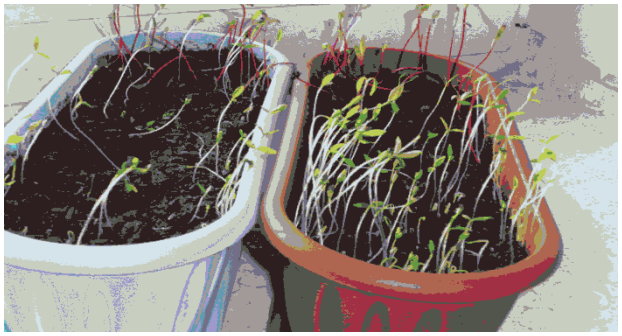
Calculations according to (4) provide the total number of falling droplets in mentioned water unit. For drizzle it is $N_d \approx 6.76 \cdot 10^{11}$, for rain $N_r \approx 4 \cdot 10^{11}$, and for storm $N_s \approx 2.45 \cdot 10^{11}$ for $AIT\beta = 3$ liters of water for drizzle, rain, or storm. These results prove that rain droplets could be interpreted as a porous medium with a large surface for gas/liquid interactions as compared to the ocean's plane surface. Then, each falling droplet runs at an air cylinder with a minimal volume length h and a ground area πr_m^2. The cylinder surface $S_m \approx h2\pi r_m$ means that air/water interface increases for rainwater and purified air contact. The total sum of this surface for all droplets in considered water unit is, as follows:

$$S \approx \sum_r N_{m,I} S_m = \frac{3AIT\beta h}{2} \sum_r \frac{q(r)}{r^2} \qquad (5)$$

provide the following numeric values: $S_d \approx 3 \cdot 10^8$ m^2 for drizzle, $S_r \approx 2 \cdot 10^8$ m^2 for rain, and $S_s \approx 10^8$ m^2 for storm. The corresponding calculations for expanded atmospheric air/water interface considering droplet set were done in precipitation volume $S_{hA} = 4 \cdot 10^3$ m^2 with altitude $h_a = 1$ km and ground surface $A = 1$ m^2 for precipitation time $T = 1$ hour. As a result, the formula (5) demonstrates the 10^5 times increase in air/water surfaces for purification possibilities as a result of rain. Due to their linear contribution in formulas for N and S, the values of T, h, I, and A can be multiplied by any numeral for real time, altitude, rain intensity or grand square. Due to the small co_2 concentration in cloud media the probability of collision between water droplets and co_2 molecules is low; but erosion of co_2 as well as a significant decrease in its concentration occurs in the sub-cloud precipitation volume.

An extra advantage of proposed approach is very positive plant response to the precipitation resulted from proposed method. To emulate the process of co_2 absorption in water droplets during an indoor experiment, the similar changes were made in alkaline solution during long-time diffusion according to formula $L_{lab} \approx (D \cdot t)^{1/2}$, diffusion coefficient is $D = \cdot 10^{-5}$ cm^2/sec, but L_{lab} there is a water layer depth. The KOH was dissolved in water and resulting mixture was kept indoors during several hours $t = L^2_{lab}/D$, as a result the solution has ions of $K+$; HCO_3^- ; CO_3^{2-}. The initial $pH = 12$ was kept in these experiments. After 9-hours of saturation time t, the plants in brown pot were given the resulting solution, but the same volume of pure water was added to control blue pot (left side) with the same plants, Fig.1(a-d). These two watering processes were repeated regularly every day during January 2014.

a


b

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c d

Figure 1 : The indoor plants after 0 (a), 19 (b) and 30 (c) days correspondingly; (d) is the next plants after 19 days in the same soils with the same watering procedures. The plants in a right (brown) flower pot were watered by solution with ions CO_3^{2-}; HCO_3^- ; K+.

The experimental plants are beetroots (10 grains), carrots (20 grains), and much number of parsley (2 Grams) in each of plant pots, see Fig.1 (a, b, c).The second experiment was done later with the same soil after deleting previous plants; the next portion ware planted, there are 20 grains of dill and 5 grains of cucumber for each plant pot. The same watering process was repeated regularly every day during February 2014, results are presented at Fig.1d. One can see a strong vegetation growth by enriched solution watering according to proposed method for all studied plants.

III. The Method Consideration

In general the proposed method can be applied for the whole Earth on the global scale. The surface area of our planet is $A_E \approx 5{,}1\cdot10^8$ km^2 and the average annual layer of precipitation is $h_w \approx 1{,}000$ mm. Using equation (1) one can estimate the mass of removed atmospheric carbon (M_C^1) in 1 meter of precipitation water measured per surface 1 m^2 at pH = 10 - 10.8, for details see 1st and 2nd rows of Table. The carbon ratio in CO_2 is 0.2727, hence the amount of carbon oxide is $M_{CO2}^1 = M_C^1/0.2727$ and its value listed at the 3th row of the Table. Note, the $M_{CO2}^1 = 5.9\cdot\cdot10^{-4}$ then pH=5.6 for typical rain. The calculated mass of formed carbon at the surface A_E of the whole planet is $M_C^A = M_C^1\cdot A_E = 0.849\cdot5.1\cdot10^{14} \approx 4.3\cdot10^{14}$ kg then pH = 10, as listed at the 4th row of the Table. The CO_2 value $M_{CO2}^A = M_{CO2}^1\cdot A_E = 3.11\cdot5.1\cdot10^{14} \approx 1.6\cdot10^{15}$ kg listed at the 5th row of the Table. In 2010 the global CO_2 emission reached an amount of $3.06\cdot10^{13}$ kg. For further calculations let's assume the global annual emission of CO_2 as the value $AE = 3.2\cdot10^{13}$ kg, for example. To compensate an annual CO_2 emission from the ratio AE / M^A_{CO2} we estimate the minimal area at the Earth's surface (A_p, %) to be used for proposed technology and the corresponding details are given at the 6th row of the Table. For our approach the proposed method could to be applied on 2% - 0.14% of planet surface to compensate the annual carbon emission. To sum up let's estimate the mass of alkali to add for desired modifications in clouds. The mass KOH (Q_{KOH}) can be estimated using previous data for n_{KOH} from (3). Considering the precipitation layer of 1 m/year for the planet surface A its necessary percent $(\%A)$ can be estimated from the Table. The added mass of alkali at pH = 10 level is calculated here:

$$Q_{KOH}^{10} = n_{KOH}\cdot 1\cdot A\cdot(A_p) \approx 5.6\cdot10^{-3}\cdot1\cdot5.1\cdot10^{14}\cdot0.02 \approx 5.7\cdot10^{10}\text{ kg} = 57\text{ mil.Tons} \quad (6)$$

Note that similar estimations are presented at the bottom row of the Table. In comparison with the fertilizers for soil the added mass of alkali is relatively small. A required modification of the clouds can be achieved with the help of aircraft by spraying alkaline aerosol particles on top of the clouds and also by lifting of alkaline gas from the ground (Tulaikova 2012).

Table 1 : The carbon mass (M^A_C) and CO_2 mass ($M^A_{CO_2}$) for the Earth's surface $A_E = 5.1\cdot10^8$ km^2; the minimal required surface (A_p) and mass of KOH (Q) to compensate an annual carbon emission

1) pH-level	10	10.5	10.8
2) M_C^1 per 1m^3, kg	0.849	4.05	12.06
3) M_{CO2}^1per 1m^3, kg	3.11	14.8	44.2
4) M^A_C, mil. Tons	$4.3\cdot\cdot10^5$	$2.1\cdot\cdot10^6$	$6.2\cdot\cdot10^6$
5)$M^A_{CO_2}$, mil.Tons	$1.6\cdot\cdot10^6$	$7.7\cdot\cdot10^6$	$2.3\cdot\cdot10^7$
6) A_p , %	2%	0.42%	0.14%
7) $Q_{KOH}\cdot$, mil. Tons	57	38	25

IV. Acoustical Impact Inside Cloud for Droplet's Coalescence

The most interesting perspective for today is a joint utilization of two methods at the same time, which means that hygroscopic particles and acoustic influence would be directly applied inside one cloud area. Dynamics of different hygroscopic particles show that only first 15 ÷ 20 seconds demonstrate very fast changes in the main cloud characteristics as the supersaturation and spectrum dispersion. This is

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accompanied by an increase of the radii inside small drops from 0.01 ÷ 0.1 μm up to 1 ÷ 3 μm. So, after the first major variations of these parameters they increase with the same tendencies very slowly, so significant changes in spectrum of cloud and precipitation increase up to time 30 – 50 minutes (Drofa, 2006). It could be understood as initiation of water vapor condensation onto surface of hygroscopic particle from surrounding, but next stage will be the evaporation from the host cloud droplets with further condensation to new particle, such reorganization need much time. The idea is that drops could be triggered by sound waves in the form of vibrations to provide the coalescence to a drop size of more than 100 μm, and after this point gravity will predominate.

The propagation, absorption and attenuation of sound in clouds decrease with distance *(x)* in ideal air media, as follows:

$$P = P_0 \exp(-\alpha x),$$

$$\alpha = \frac{b \cdot \omega^2}{2\rho \cdot C_a^3},$$

$$b = \frac{4}{3}\eta + \frac{\gamma - 1}{C_p} \cdot a_T + \eta' \approx 2.5 \cdot \eta \qquad (7)$$

The absorption coefficient α of sound in the medium is expressed in m^{-1}. There ω is a sound circular frequency in air, η is the dynamic viscosity, the C_p heat capacity at constant pressure and the ratio of specific heat capacities is γ, the α_T is a thermal conductivity (m^{-1}). Theory predicts the absorption maximum in the range of few tens of Hz. Attenuation β may be expressed in sec^{-1}, then the value of α (m^{-1}) must be multiplied by the speed of sound. Measurements in the water mist then water liquid content is W = 2 g/m^3 for the more interesting frequency range for further consideration are: β = 3.5 dB/s for f = 58 Hz; β = 2.8 dB/s for f = 112 Hz; β = 3.6 dB/s for f = 150 Hz; β = 6.7 dB/s for f = 200 Hz (Mednikov, 1965). Measurements and calculations indicate further sound attenuation is not high $\alpha \sim 10^{-3} \div 10^{-2}$ dB/m in dry air, then the sound frequency is f = 3 ÷ 10 *kHz*. As the humidity increases up to 12 ÷ 20 % , the attenuation increases up to 0.17 ÷ 0.56 *dB/m*. For higher frequencies the attenuation does not depend strongly on the humidity and give the values: α = 0.15 ÷ 0.05 m^{-1} then f = 20 *kHz;* and etc. Using mentioned measured data we deduce that at low frequency (f = 50 - 200 Hz) the acoustical wave can propagate ~ 500 meters in typical clouds prior to decreasing in energy by a factor of 2. The x = 0.5 km is enough distance in cloud for initiation of the reorganization for precipitation to begin. In practice, the most effective glaciogenic particles run the distance of ~0.1 km in cloud from airplane prior to their evaporation or sublimation, such distance provides enough cloud reorganization for precipitation enhancement. The effects of the partial dissipation of irradiated sound waves during their propagation in a turbulence medium have been measured and considered in (Corner 1958, Ostashev 1992), for example. Clouds with plenty or prolonged rainfall are identified as *Ns, As, Cb* and *Cu*, they are more important for the utilization of proposed methods. Autumn and spring clouds are mixed with water droplets, ice crystals and snowflakes. The authors of paper (Connolly 2005, Connolly 2006) studied the formation and shapes of cloud fractions using both experiments and theories, they gave convenient empirical formula that can be employed in order to determine the percentage of water vapor, liquid, and solid phases in real clouds created by the adiabatic process.

The purposes of further analyses is to calculate an amplitude, *L(r,ω),* during the vibrations for typical droplet sizes inside acoustic wave to find no high optimal power. The optimal regimes below were analyzed and calculated for the ensemble of cloud droplets: the optimal frequency (*f*) should be low enough with an appropriate decrease in the acoustical power, *Q*. The power decrease tends to weight and size minimization these are desirable from a real utilization in helicopter. Typical clouds have droplet radii within the range from $r \approx$ 1 - 50 μm, and the objective for the acoustics is the additional motion of the droplet ensemble in receiving greater droplets with radii $r \geq$ 100 μm. Known model (8-9) was developed earlier for small droplet assembles (Fuchs 1064), it gave good description for special media like smoke or industrial fogs where radii r ≤ 1 μm are small. The Stokes friction for air flow at the droplet's surface provided particle's motion inside acoustic wave, which is physical core of this model. For more large cloud droplets this model wrong, because it predicts the equality for the velocities of air V_a and droplet V_w for small frequencies ω, see the first term in right side of result equation (9). The acoustic pressure is $P(t) = P_a \cdot sin(\omega t)$, ω is the circular frequency of the acoustic wave This model present equations with their solutions, as follows:

$$m\frac{dV_w}{dt} = 6\pi\eta r\left(V_a - V_w\right) \qquad (8)$$

$$\tau\frac{dV_w}{dt} + V_w = V_{a,\max}\sin(\omega t)$$

$$V_w = \frac{V_{a,\max}\sin(\omega t - \varphi)}{\sqrt{1+\omega^2\tau^2}} + \frac{\omega\tau V_a}{1+\omega^2\tau^2}\cdot\exp\left(-\frac{t}{\tau}\right) \qquad (9)$$

There relaxation time is $\tau \approx 0.22 \cdot \rho_w \cdot r^2/\eta$, $\tau \sim 10^{-7}$s for particles radius 0.1 μm, but $\tau \sim 10^{-5}$ s for r = 1 μm, it provides zero in the second term in right side of (9) for low frequency then t ~ π/ ω. As a result, the

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medium/droplet delay $\varphi = arctan(\omega\tau)$ is small here when $\tau\omega << 1$. This model was developed for small particles then $r << 1$ μm was the norm, but frequency was in *kHz* range. Therefore, using this model we found that the velocity amplitude of driving cloud droplets becomes equal to the speed of the air molecules $V_w \approx V_a$ in (9). Of cause, this is an incorrect model for large-sized droplets in clouds which need much driving force for their big masses.

The new model is suggested here for large droplets inside clouds. Air molecules bombard great droplet surface at ½ front side to pass their impulse for droplet motion, the result is the creation of the driving force. It provide droplet vibrations back and forth in the acoustic wave; moves the droplets with an amplitude *L* that has a maximal displacement during a time for half the period, as follows:

$$L_{\max} = \int_0^{\pi/\omega} V(t)dt \qquad (10)$$

there *L* is the droplet amplitude, but *V(t)* is its velocity. The modern complete model of one-dimensional vibration for a cloud droplet in a viscous medium can be found, for example, in (Prokhorov 1997, Kumazaki 1999), as follows:

$$\left(\frac{4\pi r^3}{3}\rho_w + f_2\right)y'' + (f_0 + f_1)y' = F\cdot\sin(\omega t) \qquad (11)$$

$$f_1 = 6\pi\eta_a r + 3\pi r^2\sqrt{2\eta_a\rho_a\omega} \qquad (12)$$

$$f_2 = 3\pi r^2\sqrt{2\eta_a\rho_a/\omega} + \frac{2}{3}\pi\rho_a r^3 \qquad (13)$$

where are water and air density are ρ_w, and ρ_a; η_a is medium viscosity, and *F* is a driving force amplitude. The coefficients f_1 and f_2 correspond to the medium counteraction and are proportional to the speed and acceleration, respectively; f_0 corresponds to the inner mechanical losses that are, in reality, small. One can see that the Stokes's term, the first term of f_1 (12) is introduced into the motion equation (11). However, the second term in (12) provides the drag. The second part of f_2 characterizes the vibration of the joint mass of the medium surrounding a droplet, the first term in (13) corresponds to acoustic radiation losses. The solution of mentioned system (11-13) for the droplet velocity $V(t) = y'$ provides the following equation:

$$V = C\cdot\exp(-2ht) + \frac{B}{(2h)^2+\omega^2}\left[2h\cdot\sin(\omega t) - \omega\cdot\cos(\omega t)\right] \qquad (14)$$

where normalized acted force is $B \approx F \setminus m \approx F \setminus (1.333\pi r^3\rho_w)$. The attenuation parameter is *h* which follows from the initial equation *V=0* then *t=0;* also the integration-resulted term *c* is as follows:

$$h = \frac{f_0 + f_1}{f_2 + 4\pi r^3\rho_w/3}, \qquad C = \frac{B\omega}{\left[\omega^2 + (2h)^2\right]} \qquad (15)$$

The droplet amplitude for half of the period of acoustic waves is *L*, the final solutions are as follows:

$$L = \frac{B}{\omega^2 + (2h)^2}\left\{2\frac{2h}{\omega} - \frac{\omega}{2h}\left[\exp\left(-\frac{2h\pi}{\omega}\right) - 1\right]\right\} \qquad (16)$$

The driving force, *F,* should be determined at this point in order to obtain the numerical calculations, and it is necessary to set the predominant mechanism for the counteraction of acoustics using the weighted droplet in air to add the vibration. Here we assume the bombardment of the front surface of the large droplet by small air molecules; as well as the impulse transfer required to move the droplet to another location. The simplest formula of such a physical model describes the impulse transfer, $F\cdot\Delta t = \Delta p$. Side surface effects can be neglected for large droplet objects according to the next chapter consideration. The affecting force increases in proportion to the area of the front surface is ~ $2r^2$. However, the droplet mass grows with radius as ~ r^3 that is mass driving is predominate. Molecule forward or back motions occur during the time of half period of the wave $t_{1/2} = \pi/\omega = 0.5f^{-1}$. The volume of air molecules in the front of the droplet is $U_N = S\cdot\int_0^{\pi/\omega} V_a(t)dt = \pi r^2 V_a 2/\omega$, but the cross-section of water droplet is $S = \pi r^2$. The complete mass for air molecules in this volume is $M_a \approx N\cdot 28\cdot m_p\cdot U_N$. Here Avogadro's constant, and taking molecular mass for nitrogen is $2\cdot 14 m_p$. We put to use for estimations the average molecule's velocity in acoustical wave $V_a/2$. Driving force in acoustic wave according to second Newton's law could be found $F = M_a\cdot\Delta V/\Delta t \sim M_a\cdot V_a/t_{1/2}$, and it corresponds to the next equation:

$$F \approx (28 m_p N)\cdot r^2 V_a^2 = C_F\cdot r^2 V_a^2 \qquad (17)$$

where the coefficient is $C_F \approx 1$ in the SI system. The acoustic pressure, *P,* in the sound wave; the acoustic power, Q; the velocity, V_a, of the air molecules; their displacement, L_m; and the speed of sound C_a are connected as follows:

$$V_a = \sqrt{\frac{2Q}{\rho_a C_a}}, \quad P_a = \rho_a C_a V_a, \quad L_m = \frac{1}{2\pi f}\sqrt{\frac{2Q}{\rho_a C_a}} \qquad (18)$$

Below we introduce the pulse power *J,* then $Q = J\cdot f$. After previous unification, the driving force or large droplets in the acoustic wave is as follows:

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$$F \approx \left(\frac{2C_F}{\rho_a C_a}\right) r^2 J \cdot f \,, \quad B=F/m_w \qquad (19)$$

The droplet mass is $m_w=4\pi r_w^3/3$, the droplet give the amplitude $L(f,r)$ in the from $L(f,r)$:

$$L(f,r)=\frac{Jf}{r}\cdot\frac{C_{F2}}{(2h)^2+(2\pi f)^2}\cdot\left\{\frac{2h}{\pi f}+\frac{\pi f}{h}\cdot\left[1-\exp\left(\frac{-h}{f}\right)\right]\right\} \qquad (20)$$

There coefficient is $C_{F2}=\dfrac{3C_F}{2\pi\rho_w C_a \rho_a}$.

Numerical calculations and the results are presented in Figure 2 according to proposed model (10-20). Figure 2 demonstrates examples for some optimized regimes for three acoustical ranges, as follows: (1) f = 20 Hz with lowest power Q = 175 W/m^2; (2) f = 50 Hz with Q = 800 W/m^2 and (3) f = 100 Hz with Q = 2500 W/m^2. Additional analysis were performed using the introduction of appropriate altitude changes relative to the main physical parameters in the model, as follows: air density (ρ_a), viscosity (η), sound velocity (C_a), and air temperature depending on km-altitudes. A decrease in necessary acoustic power using the 2 – 6 km location of cloud with sound source within the atmosphere was observed, so calculations indicated a 10-15% increases for the amplitude values, L, according to the acoustics applied to the cloud drops directly at a altitudes of 6 km above the ground. For a typical cloud droplet, the estimations indicate that gravity is negligible small (0.1%), as well as the drag force, due to the friction of air flow at the surface of the droplet.

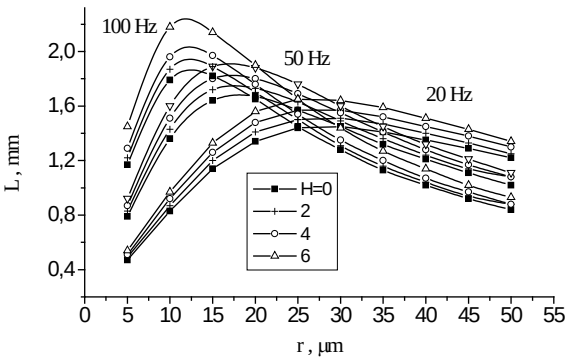

Figure 2 : Vibration amplitudes L for droplets radii 5 – 50 μm for their location at altitudes H = 0, 2, 4, 6 km. The regimes are: f = 100 Hz and Q =2.5 kW/m^2; f = 50 Hz and Q =800 W/m^2; f = 20 Hz and Q = 175 W/m^2

Now let's estimate the optimal average distance that should allow the droplets to collide. An average number concentration of droplets in a cloud occurs in the range from N ~ 60 – 1000 cm^{-3} and more according to various measurements, see for example (Borovikov, 1961). Here we assume that N ~ 500 cm^{-3} specifies the individual volume occupied by a weighted drop 1/N, and that its average distance is $L_i \sim N^{-1/3} \approx 1.26$ mm. The proposed regimes at Figure 2 provide necessary amplitudes for the typical ensemble of cloud droplets. The time requirement is L_i / r ≈ 1260/10 = 2 minutes or more to provide coalescence probability. The probability of the complete or partial amalgamation for two collision rainy droplets depends both on their sizes and velocities as listed below. The irradiation time should be more longer for low frequency affects.

V. The Acoustic Technique to Facilitate Precipitations

Sirens are believed to be most suitable generators for powerful sound in air medium (Hartman 1939, Gladishev 2000). The variant of siren is composed of a fixed stator with small holes in the periphery, and it has a stator located inside of the rotating rotor with the same numbers of holes. The holes of the rotor and stator periodically overlap, so the compressed air comes out from siren core from time to time. Outgoing air kicks to the walls of the resonator vibrating on own resonance frequency to put acoustical signal to the outer media. The siren frequency is given by the number of holes (or teethes) z, and the number of rotor circulation per second, n_c, as follows $f = zn_c$. For example, when n_c = 12.5 c^{-1} and z = 8, the frequency is f = 100 Hz; or the n_c = 6.25 c^{-1} then frequency is f = 50 Hz. This frequency is related to the rotor speed and the fundamental mode of the resonator to maximize power output for the signal. The siren was constructed and tested in experiments, so the frequency change of the rotation speed is done by coupling the motor and the transformer with the variable voltage in order to tune speed for experiments.

Let's describe below the features of Bessel-form resonator to get pure monochromatic wave by high spectrum selection into fundamental mode of resonator. The idea is that the shape of the optimal resonator for a siren should have Bessel-formed walls to maximize the power of acoustic radiation to a fundamental harmonic within output beam. The fundamental harmonic distribution for energy in the radial direction is $J_0(kr)$ of propagated fundamental wave. The sound frequency is f, and sound speed C_v = *340 m/s*, for wavelength (Λ), when f = 100 Hz we have: $\Lambda = C_v/f = 3.4$ *m,* the wave vector $k_0 = 2\pi/\Lambda = 2\pi f/C_v$. The first zero-solutions for the Bessel function is x_0=*2.404826* for $J_0(k_0 r_0) = J_0(x_0) = 0$, such that the reflector's output radius, r_0, meters, can be modified to the formula $k_0 r_0 = x_0 =$ *2.404826,* and as follows:

$$r_0(f)=\frac{x_0}{2\pi\cdot f}\cdot C_v \approx \frac{130}{f} \qquad (21)$$

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The calculated data for the outlet radiuses of Bessel's resonators are: $f = 100$ Hz needs $r_0 = 1.3$ m; $f = 50$ Hz needs $r_0 = 2.6$ m and etc.

At the outlet of proposed reflector (at $x_0 = 2.4$), the edges should be folded outwards as a bell-shape to avoid edge diffraction with appropriate transfer of the radiated power into the higher harmonics. Technically, such a reflector could be manufactured using long and narrow strips of sheet of metal; strips could be assembled according to the principle of the fan and held together by transverse belts. The output siren powers can be estimated using data at Fig.2. These data should be multiplied to the output resonator square $S_{res} = \pi r_0^2$ then resonator output radius r_0 was calculated from equation (21), as follows:

$$Q_0 = 2.5 \cdot S_{res} \approx 13 \text{ kW then } f = 100 \text{ Hz};$$

$$Q_0 = 0.8 \cdot S_{res} \approx 21 \text{ kW then } f = 50 \text{ H} \qquad (22)$$

There are sirens with similar output power, and manufactured siren in (Tulaikova 2010) had power up to $Q_0 \approx 4$ kW. The experimental tests in mist areas demonstrate the coagulation effect, but the siren power should be increased by the factor 3 for $f = 100$ Hz according to (22). The simple way to do it is the increase of the number of holes by factor 3 or the same increase of hole's surface, so radiuses should be increased up to $1.7 \cdot r$. The air output flow near siren holes is $V \approx 300$ m/s and summarized holes surface $S_0 = 3$ cm^2. Calculation show that the siren needs the air pump about ~ 0.1 m^3/s. Let's consider ways of improving of acoustical effect and/or acoustic technique. To begin with we note, that the siren will be more effective then the average number N_{cl} of droplets in a cloud is higher then considered here number $N = 500$ cm^{-3}, so let's assume $N_{cl} \sim 1000$. The average droplet distances will be $L_m \sim 1$ mm and according to previous estimation: $L_m \sim N^{-1/3} \approx 1$ mm. The vibration amplitude from equation (20) is linearly proportional to the acoustic power necessity J. This means that such cloud has small output acoustic power from siren according to (22) and Fig.2: $Q_0/2 \approx 6.5$ kW.

Also considered here is the type of moved siren with air inside pumped by the strong oncoming of the air flow through additional cylinder with compression system (Karnovsky 1945). Such device can be located at airplane and oriented according to its axis, so incoming air flow is captured by compressed system to put air to sired rotated core, and also to the Bessel-resonator through summarized holes surface S_0. The axis of Bessel-form resonator should be oriented at opposite to moving direction. The model and calculation show high efficiency to get air pumping inside moved siren cylinder then airplane velocity is not supersonic.

The following acoustic action utilizes a helicopter. Small airplane is typically used in clouds for precipitation enhancement and so hygroscopic particles distributed into the cloud from an airplane. Note that a helicopter can serve this purpose much better, because it is less speedy and more maneuverable then an airplane. The helicopter has the following advantage in comparison to the airplane. The rotor blade of the helicopter rotates with a sufficiently large angular velocity, so the linear velocity at the end of each blade reaches the speed of sound. Typical helicopters blade has a length of 10 meters and its partial speed varies linearly from 0 to 300 m/sec, but suppose helicopter is moving forward with some speed, for example 100 km/h = 360 m/sec then supersonic motion of one blade is produced in small time periods. This effect takes place because there are the same directions of helicopter motion and one of its blades moving in the same direction with its own speed. It means that each of the blades overcomes the supersonic regimes producing the shock wave. The shock wave has a steep front and big difference of pressure and temperature comparable to surroundings. So such wave pushes the droplets to coalesce effective and also provides a rapid condensation. Fast condensation inside the shock wave can be seen in many photographs when airplane breaks the sound barrier.

VI. Conclusion

In this paper two staged approach for free atmosphere CO_2 purification is proposed. To begin with alkaline reagents were injected into natural clouds in order to increase their pH-level up to 10 or 11. Enhanced precipitation facilitated for the carbon transport from atmosphere to the ground. It was shown by corresponding calculations that there is a considerable increase of the gas/water interface for CO_2 absorption; and grass photos indicates the positive plant's reaction to water at initial $pH = 12$. Note that the proposed acoustic method is up to date and does not require extensive support. The models and calculations are presented for the regimes of acoustic power that are required to implement inside cloud according to proposed new low-frequency model. Acoustic method provides fast droplets coalescence inside nature clouds and sedimentation "to get rain at the right time". Additionally, artificial rains facilitate air purification and climate corrections. The resulting effect can compensate for annual carbon emission by applying method at 0.4 % – 0.1 % of the Earth surface.

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