CHEMICAL ENGINEERING TRANSACTIONS

VOL. 43, 2015

A publication of AIDIC

The Italian Association of Chemical Engineering
Online at www.aidic.it/cet

Chief Editors: Sauro Pierucci, Jiří J. Klemeš

ISBN 978-88-95608-34-1; **ISSN** 2283-9216

DOI: 10.3303/CET1543335

Scavenging of Soluble Radioactive Gases by Evaporating Rain Droplets from Inhomogeneous Atmosphere

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We analyze effects of inhomogeneous concentration and temperature distributions in the atmosphere, rain droplet evaporation and radioactive decay of soluble gases on the rate of trace gas scavenging by rain. We employ a one-dimensional model of precipitation scavenging of radioactive soluble gaseous pollutants that is valid for small gradients and non-uniform initial altitudinal distributions of temperature and concentration in the atmosphere, and assume that conditions of equilibrium evaporation of rain droplets are satisfied. It is demonstrated that transient altitudinal distribution of concentration under the influence of rain is determined by the linear wave equation that describes propagation of a scavenging wave front. The obtained equation is solved by the method of characteristics. We calculated scavenging coefficients for wet removal of gaseous Iodine-131 and tritiated water vapor (HTO) for the exponential initial distribution of trace gases concentration in the atmosphere and linear temperature distribution. Theoretical predictions of the dependence of the magnitude of the scavenging coefficient on rain intensity for tritiated water vapor are in good agreement with the available atmospheric measurements.

1. Introduction

Scavenging of radioactive atmospheric soluble gaseous pollutants by rain droplets is the result of gas absorption mechanism. Variation of altitudinal distribution of concentration of a soluble radioactive gas in the atmosphere due to rain scavenging changes also radioactivity distribution in the atmosphere. In the present study we analyze dynamics of soluble radioactive gas scavenging by rain taking into account the effects which were neglected in the previous studies, e.g. droplet evaporation, inhomogeneous altitudinal temperature and concentration distributions in the atmosphere and radioactive decay of soluble gases. The analysis is focused on radioactive soluble in water atmospheric trace gases which can be appreciably scavenged by rain of moderate intensity and duration, namely Iodine-131 and tritiated water vapor (HTO). Iodine-131 is a radioactive isotope formed in nuclear fission, and has a half-life period of 8.02 days. Iodine-131 was a significant contributor to the health hazards from open-air atomic bomb testing in the 1950s, and from the Chernobyl disaster, as well as being a large fraction of the contamination hazard in the Fukushima nuclear crisis (Steinhauser et al. 2014). The gaseous release fraction of Iodine-131 is typically as high as the particulate fraction. In the Fukushima accident emissions about 70 % of the released 131-I was gaseous. In the Chernobyl accident, about 25 % of the total reactor inventory of 131-I was released into atmosphere as vapour or particulate aerosol. Naturally occurring tritium is produced by cosmic radiation. However, the distribution of natural tritium in the atmosphere and hydrosphere was severely disturbed since the testing of thermonuclear weapons began in 1954 (Junge 1963). Concentration measurements in the atmosphere revealed the decrease of tritiated water vapor (HTO) concentration with height (Ehhalt, 1971).

Please cite this article as: Elperin T., Fominykh A., Krasovitov B., 2015, Scavenging of soluble radioactive gases by evaporating rain droplets from inhomogeneous atmosphere, Chemical Engineering Transactions, 43, 2005-2010 DOI: 10.3303/CET1543335

2. Description of the model

2.1 Scavenging of radioactive gases with low solubility by non-evaporating droplets

Consider absorption of radioactive gas having a low solubility from a mixture containing inert gas by rain droplets falling in the atmosphere with the known initial non-uniform concentration and temperature distributions. Since the velocity of scavenging front propagation is proportional to the solubility of scavenged gases (see, e.g. Elperin et al. 2013), the velocity of temperature front propagation is by orders of magnitude larger than the velocity of a scavenging front propagation for radioactive gases having a low solubility, e.g. Iodine-131. Therefore, during the period of time when temperature front propagates from a bottom of a cloud to the ground and temperature distribution becomes homogeneous, the changes of the concentration profile are negligibly small. Consequently, scavenging of radioactive gases having a low solubility by precipitation in the inhomogeneous atmosphere can be analyzed assuming a homogeneous temperature distribution (Elperin et al. 2011, Elperin et al. 2013). The goal of this study is to determine the rate of radioactive soluble trace gases scavenging by falling rain droplets. In the case of gases having a low solubility the approximation of small gradient of concentration is always valid (Elperin et al. 2014). When the concentration gradient is small, the total concentration of soluble gas in the gaseous and liquid phases reads (Elperin et al. 2013, Elperin et al. 2014):

$$c = c^{(G)}\left[(1-\phi)+m\phi\right] . \tag{1}$$

The total mass flux density of the dissolved gas transferred by rain droplets is determined by the following expression:

$$q_c = m\phi \; u \; c^{(G)} \quad , \tag{2}$$

where u is the terminal fall velocity of droplets, $c^{(G)}$ is concentration of the soluble gas in a gaseous phase (in $[\,mole\; l^{-1}\,]$), m - dimensionless Henry's law coefficient. Equation of mass balance for radioactive soluble trace gas in the gaseous and liquid phases reads:

$$\frac{\partial c}{\partial t} = -\frac{\partial q_c}{\partial z} - \lambda c \; , \tag{3}$$

where z axis is directed from cloud bottom to the ground. Combining Eqs. (1) – (3) we obtain the following equation:

$$\frac{\partial x^{(G)}}{\partial t} + U_0 \frac{\partial x^{(G)}}{\partial z} + \lambda x^{(G)} = 0 , \tag{4}$$

where t is time, $x^{(G)}$ is mole fraction of soluble trace gas in the gaseous phase, $U_0 = mI$, I is rain intensity and λ is a radioactive decay constant. In the case of the exponential initial distribution of soluble gas in the atmosphere the initial and boundary conditions for Eq. (4) read:

$$t = 0 \; , \; x^{(G)} = x_c^{(G)} \cdot exp(k_1 \cdot z) , \tag{5}$$

$$x^{(G)} = x_c^{(G)} e^{-\lambda t} \;\; \text{for} \;\; z = 0 \; , \tag{6}$$

where $x_c^{(G)}$ is mole fraction of soluble gas at the cloud bottom at $t = 0$. Solution of Eqs. (4) – (6) yields the following expression for the scavenging coefficient $\Lambda = -\left(x^{(G)}\right)^{-1} \cdot \partial x^{(G)} / \partial t$:

$$\Lambda = \begin{bmatrix} k_1 \cdot U_0 + \lambda & z > U_0 \cdot t \\ \lambda & z < U_0 \cdot t \end{bmatrix} . \tag{7}$$

The dependence of scavenging coefficient vs. rain intensity for Iodine-131 wash out by rain is showed at Figure 1.

2.2 Scavenging of highly soluble radioactive gases by evaporating droplets in the atmosphere with non-uniform altitudinal concentration and temperature distributions

For highly soluble gases the velocity of scavenging front propagation is by orders of magnitude larger than velocity of temperature front propagation. Therefore, it can be assumed that during the time required for complete scavenging of radioactive gases by precipitation, the changes of temperature profile can be neglected.

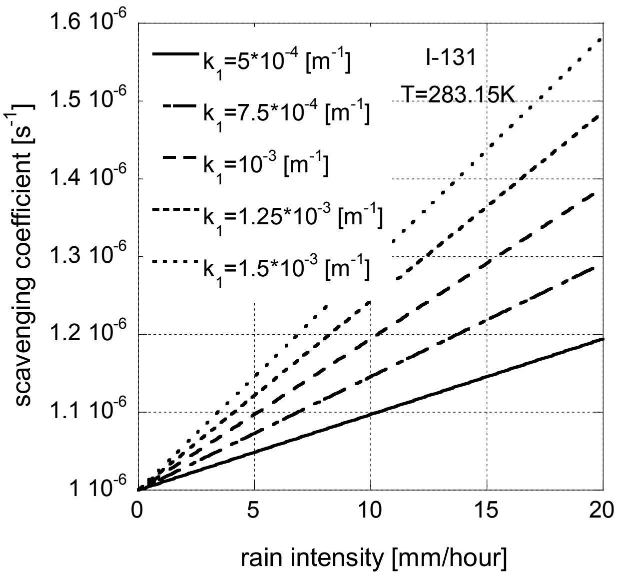

Figure 1: Dependence of scavenging coefficient vs. rain intensity for Iodine-131 wash out by non-evaporating droplets for different values of growth constant k_1 (Eq. 7)

Consequently, analysis of scavenging of highly soluble gases in the inhomogeneous atmosphere in this case reduces to solving equations of mass transfer in the atmosphere with the stationary inhomogeneous temperature distribution. Since in the considered problem the gradients of concentration and temperature are small, it can be assumed that the instantaneous concentration of the dissolved radioactive gas in a droplet is equal to the concentration of saturation in a liquid corresponding to the concentration of a trace soluble radioactive gas in the atmosphere at a given height (Elperin et al. 2014). Similarly, the instantaneous temperature of the droplet is equal to the local atmospheric temperature at a given height. Consider gas absorption by slowly evaporating falling rain droplets in the case of stationary linear altitudinal temperature distribution in the atmosphere. The dependence of the evaporating droplet radius vs. time is determined by the following expression:

$$a = \sqrt{a_0^2 - K_v t} \,, \tag{8}$$

where $K_v = K_{v0} + K_T \cdot z$, K_{v0} is a coefficient of droplet evaporation in the homogeneous atmosphere (see, e.g. Elperin et al. 2013), $K_T = -K_{v0} k_2 / T + (3 a_1 b_1 T_c k_2) / \left(b_1 T_c^3 + 1\right)^2$, $k_2 = (T_{gr} - T_c)/L$, T_c and T_{gr} - temperature at a bottom of a cloud and at the ground, L – distance between a bottom of a cloud and the ground, a_1 and b_1 are coefficients that depend upon thermodynamic properties of gas and liquid. The dependence of droplet fall velocity on droplet radius can be approximated by the following correlation for the terminal fall velocity:

$$u = c_1 \left[a(z)\right]^{\alpha} . \tag{9}$$

The values of coefficients c_1 and α can be found in Pruppacher and Klett (1997). Formula for volume fraction of evaporating droplets vs. droplet diameter reads:

$$\phi(z) = \left[a(z)\right]^3 \phi_0 \big/ a_0^3 \,. \tag{10}$$

Taking into account that $U(z) = m(T(z)) \cdot \phi \cdot u$, $m = m_0 - k_3 \cdot (T - T_0)$ and $T^{(G)} = T_0^{(G)} + k_2 \cdot z$, $m = m_0 - k_4 \cdot z$, where $k_4 = k_2 \cdot k_3$, we obtain the following expression for the dependence of the scavenging front velocity vs. coordinate:

$$U(z) = U_0 \cdot \left(1 - k_4 m_0^{-1} z\right)\left(1 - (\alpha + 3)\left(K_{v0} + K_T z\right) z / \left(2 c_1 a_0^{\alpha+2}\right)\right),. \tag{11}$$

The dependence of the velocity of scavenging front propagation vs. distance from a cloud bottom is showed in Fig. 2 for the initial temperature distribution in the atmosphere that is determined by nocturnal inversion and for rain intensity, equal to 14 mm/hour. The altitudinal temperature dependence increases scavenging velocity while droplet evaporation decreases scavenging front velocity. Competition of these two mechanisms – nocturnal inversion and droplet evaporation – leads to the increase of the scavenging front velocity at smaller distances from the cloud and to the decrease of the scavenging front velocity at larger distances from the cloud (see Figure 2).

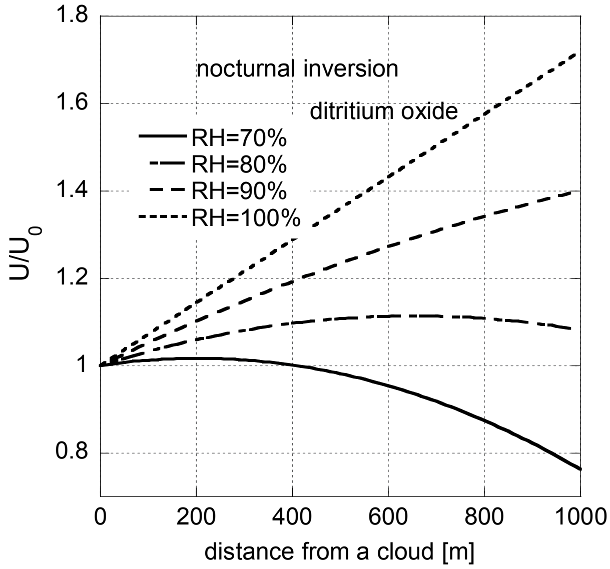

Figure 2: Dependence of scavenging velocity for absorption of tritiated water vapor (HTO) by evaporating droplets vs. distance from cloud bottom for various values of relative humidity.

Using equation of mass balance for the soluble radioactive trace gas in the gaseous and liquid phases and applying the approach outlined in Elperin et al. (2011) we arrive at the following equation for soluble trace gas distribution in the atmosphere:

$$\frac{\partial x^{(G)}}{\partial t} + U(z)\frac{\partial x^{(G)}}{\partial z} + \lambda x^{(G)} = 0 \,, \qquad (12)$$

where U(z) is determined by Eq. (11). The initial and boundary conditions to Eq. (12) are described by Eqs. (5) and (6).

3. Results

The dependence of scavenging coefficient for tritiated water vapor on rain intensity for the initial exponential distribution of radioactive trace gas concentration in a gaseous phase, calculated using the obtained analytical solution of Eq.(12) with the initial and boundary conditions (5) and (6), is shown in Figs. 3a-b. When temperature inversion determines altitudinal atmospheric temperature distribution, the value of the scavenging coefficient decreases with time for given rain intensity. On the contrary, when environmental lapse rate determines the altitudinal atmospheric temperature distribution, the value of scavenging coefficient increases with time for given rain intensity. Clearly, scavenging coefficient always increases when rain intensity increases. The dependence of the scavenging coefficient vs. altitude in the case of tritiated water vapor wash out by evaporating rain droplets for the exponential initial profile of soluble gas in the atmosphere is shown in Figures 4a-b. Inspection of Figures. 4a-b shows that when the environmental lapse rate determines altitudinal atmospheric temperature distribution, scavenging coefficient increases with height in a region between the scavenging front and the ground. On the contrary, when temperature inversion determines the altitudinal atmospheric temperature distribution, scavenging coefficient decreases with height in a region between the scavenging front and the ground. At the ground, the magnitude of scavenging coefficient increases with time when the environmental lapse rate determines the altitudinal atmospheric temperature distribution, and decreases with time for temperature inversion. Dependence of scavenging coefficient on altitude at different times is described by converging lines, when altitudinal atmospheric temperature distribution is determined by environmental lapse rate (see Figure 4a). On the contrary, when the altitudinal atmospheric temperature distribution is determined by temperature inversion, altitudinal dependence of scavenging coefficient at different times is described by the diverging lines (see Figure 4b). Notably, the altitudinal dependence of scavenging coefficient at different times during soluble gas scavenging by non-evaporating rain droplets in the atmosphere with inhomogeneous temperature distribution is described by parallel lines (see Elperin et al. 2014).

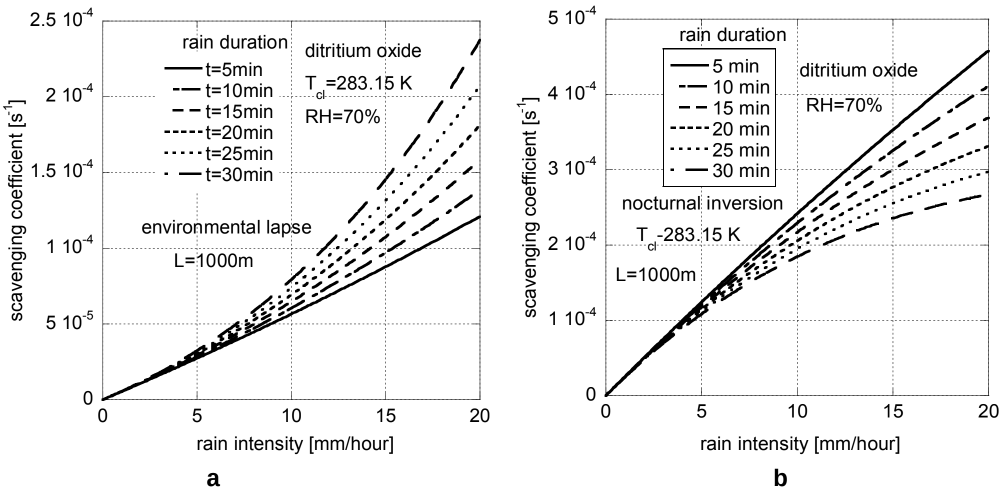

Figure 3: Dependence of scavenging coefficient vs. rain intensity for tritiated water vapor wash out by evaporating droplets. Temperature distribution is determined by a) environmental lapse; b) nocturnal inversion

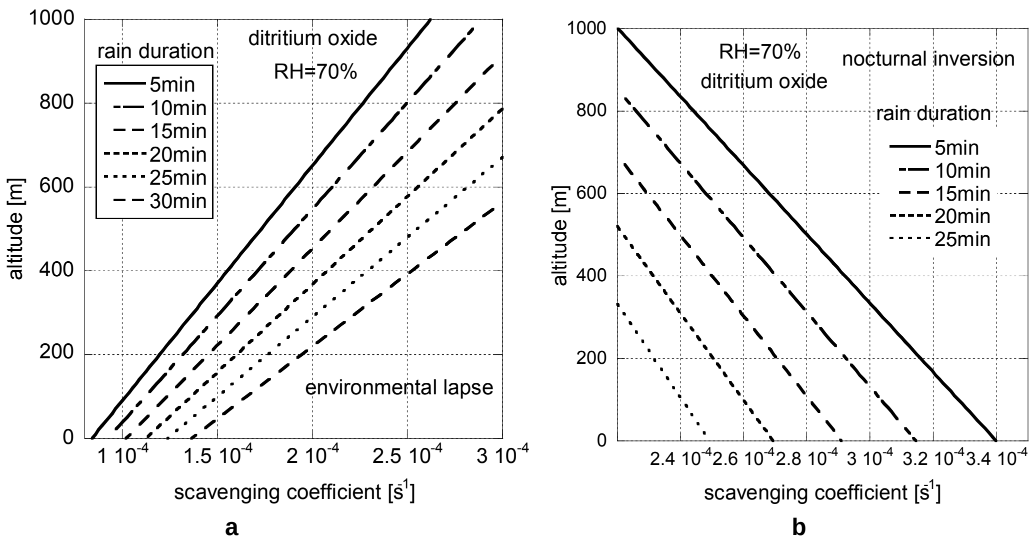

Figure 4: Dependence of scavenging coefficient vs. rain intensity for tritiated water vapor wash out by evaporating droplets. Temperature distribution is determined by a) environmental lapse; b) nocturnal inversion

Comparison of the obtained theoretical results with measurements of scavenging coefficients in the atmosphere for tritium-oxide washout by rain performed by Piskunov et al. (2012) is shown in Figure 5.

4. Conclusion

We suggested a model for scavenging of radioactive soluble trace gases in inhomogeneous atmosphere by evaporating rain droplets. It is shown, that gas scavenging in the case of low gradients of radioactive soluble trace gas concentration and temperature in the atmosphere is determined by linear wave equation, that describes propagation of a linear wave in one direction without changing shape. The obtained equation was solved by the method of characteristics. The obtained results can be summarized as follows:

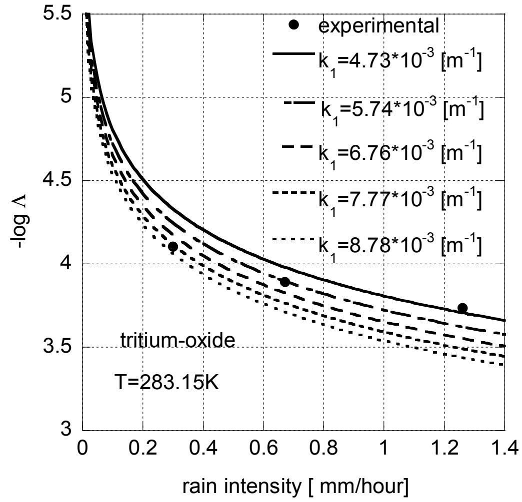

Figure 5. Comparison of theoretical predictions with atmospheric measurements of Piskunov et al. (2012) for tritium-oxide scavenging by rain

1. It is demonstrated, that if the initial altitudinal concentration distribution of a radioactive trace gas having low solubility in the atmosphere is exponential, scavenging coefficient in the region between the ground and a scavenging front is the sum of radioactive decay constant and a product of rain intensity, solubility parameter and the growth constant in the initial profile of concentration in a gaseous phase.
2. It is demonstrated that when initial temperature distribution in the atmosphere is determined by the environmental lapse, scavenging velocity is smaller than for the isothermal temperature distribution. Droplet evaporation causes further reduction of scavenging velocity. When the initial temperature distribution in the atmosphere is determined by nocturnal inversion, the competition of two factors – nocturnal inversion and droplet evaporation – leads to increase of scavenging velocity with coordinate at smaller distances from the cloud and to decrease of scavenging velocity at larger distances from the cloud.
3. It is demonstrated that altitudinal dependence of scavenging coefficient for radioactive soluble gas scavenging by evaporating rain droplets at different times is described either by converging or diverging lines depending on whether the altitudinal atmospheric temperature distribution is determined by the environmental lapse rate or temperature inversion.
4. The suggested model yields the same estimate of the scavenging coefficient during tritiated water vapor (HTO) washout by rain and the same dependence of the scavenging coefficient on rain intensity as the atmospheric measurements conducted by Piskunov et al. (2012).

References

Ehhalt D. H., 1971, Vertical profiles and transport of HTO in the troposphere. J. Geophys. Res., 76, 7351-7367.

Elperin T., Fominykh A., Krasovitov B., 2014, Non-isothermal scavenging of soluble gaseous pollutants by rain in the atmosphere with non-uniform vertical concentration and temperature distributions. Meteorol. Atmos. Phys., 125, 197–211.

Elperin T., Fominykh A., Krasovitov B., 2013, Scavenging of soluble gases by slowly evaporating droplets in inhomogeneous atmosphere, Chemical Engineering Transactions, 32, 1915-1920.

Elperin T., Fominykh A., Krasovitov B., Vikhansky A., 2011, Effect of rain scavenging on altitudinal distribution of soluble gaseous pollutants in the atmosphere. Atmos. Environ., 45, 2427-2433.

Junge C. E., 1963, Air Chemistry and Radioactivity, Academic Press, New York.

Piskunov V.N., Golubev A.V., Balashov Y.S., Mavrin S.V., et al. 2012, The effect of rain characteristics on scavenging rate of tritium-oxide from the atmosphere. Atmos. Environ., 62, 573-583.

Pruppacher H.R., Klett J.D., 1997, Microphysics of Clouds and Precipitation. Dordrecht, Kluwer Academic Publishers, Germany.

Steinhauser G., Brandl A., Johnson T.E., 2014, Comparison of the Chernobyl and Fukushima nuclear accidents: A review of the environmental impacts, Sci. Total Environ., 470–471, 800–817.