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Scavenging of radioactive soluble gases from inhomogeneous atmosphere by evaporating rain droplets

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ABSTRACT

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. We assume that conditions of equilibrium evaporation of rain droplets are fulfilled. 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. Scavenging coefficients are calculated 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 the radioactive atmospheric soluble gaseous pollutants by rain droplets is the result of gas absorption mechanism (see, e.g. Pruppacher and Klett, 1997). 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 (see, e.g. Chamberlain, 1991). 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 goal of this study is to determine evolution of concentration distribution of radioactive soluble trace gases in the atmosphere below the cloud under the influence of gas scavenging by falling rain droplets. The analysis is focused on three radioactive soluble in water atmospheric trace gases which can be appreciably scavenged by rain of moderate intensity and duration, namely radon (Rn-222), iodine-131 and tritiated water vapor (HTO).

Radon-222 (radon) is a naturally occurring radioactive noble gas of terrestrial origin that has a half-life of 3.8 days. Radon-222 is formed as progeny of uranium and thorium in rocks and soil. Rn-222 is emitted from the ground into the atmosphere where it decays and forms daughter products, isotopes of polonium, bismuth and lead. These products either remain airborne till they decay, or are deposited by rain and by diffusion at the ground. Radon concentration in air always decreases with height (see, e.g. Chamberlain, 1991; Williams et al., 2011).

Iodine-131 is a radioactive isotope formed in nuclear fission, either directly or by decay of a tellurium precursor, 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 (see, e.g. Pham et al., 2012; 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 (Kristiansen et al., 2012). In the Chernobyl accident,

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Nomenclature

a	raindrop radius, m
c	total concentration of soluble trace gas in gaseous and liquid phases, mole l^{-1}
$c^{(G)}$	concentration of soluble trace gas in gaseous phase, mole l^{-1}
$c_c^{(G)}$	concentration of soluble gas at cloud bottom, mole l^{-1}
$c_{gr}^{(G)}$	concentration of soluble gas at the ground, mole l^{-1}
$c^{(L)}$	concentration of dissolved gas in droplet, mole l^{-1}
d	raindrop diameter, m
H_A	Henry's law constant, mole l^{-1} atm^{-1}
k_1	growth constant, m^{-1}
k_2	coefficient in a linear dependence of temperature on coordinate, K m^{-1}
k_3	coefficient in linear dependence of solubility parameter on temperature, K^{-1}
$k_4 = k_2 \cdot k_3$	coefficient in a linear dependence of solubility on coordinate, m^{-1}
K_v	evaporation coefficient, $m^2 s^{-1}$
L	distance between the ground and cloud bottom, m
L_v	latent heat of evaporation, kcal g^{-1}
$m = H_A R_g T$	dimensionless Henry's law coefficient
R	rainfall rate, ms^{-1}
R_g	universal gas constant, $atm \cdot l \cdot mole^{-1} \cdot K^{-1}$
q_c	mass flux density of dissolved gas transferred by rain droplets, $mole \cdot m^{-2} \cdot s^{-1}$
t	time, s
T	temperature, K
u	terminal velocity of droplet, ms^{-1}
U	wash-down front velocity, ms^{-1}
U_0	wash-down front velocity for non-evaporating droplets, ms^{-1}
$x^{(G)}$	mole fraction of a soluble trace gas in gaseous phase
z	vertical coordinate, m

Greek symbols

β	coefficient of mass transfer, ms^{-1}
φ	volume fraction of droplets in air
λ	radioactive decay constant, s^{-1}
Λ	scavenging coefficient, s^{-1}

Subscripts and superscripts

0	initial value
c	value at the cloud bottom
gr	value at the ground
G	gaseous phase
L	liquid phase
v	vapor

about 25% of the total reactor inventory of 131-I was released to atmosphere as vapor or particulate aerosol (see, e.g. Steinhauser et al., 2014).

Tritium was discovered in water as hydrogen tritium oxide (HTO) by Grosse et al. (1951). 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 (see, e.g. Junge, 1963). Most of the tritium from the atmospheric nuclear weapons tests was formed as HTO. Tritium is also formed as a product of ternary fission in nuclear power reactors. Although most of the tritium is retained in the reactor fuel some may be released to atmosphere as HTO during fuel reprocessing. Concentration measurements in the atmosphere revealed the decrease of HTO concentration with height (see, e.g. Ehhalt, 1971).

Inspired by the studies of Chamberlain and Eggleton (1964) and Booker (1965), a number of theoretical and experimental investigations were carried out to determine the rate of soluble radioactive gas scavenging by single droplets and precipitation. Radioactive gas absorption by a single falling drop was investigated experimentally by Belovodsky et al. (1997). Scavenging of soluble radioactive gas by rain was studied theoretically by Dana et al. (1978), Atanassov and Galeriu (2011) and experimentally by Abrol (1990), Köllö et al. (2011) and Piskunov et al. (2012). Sorption of radioactive Iodine-131 by aerosols was investigated experimentally by Noguchi et al. (1988, 1990). The state-of-the art in the field of soluble radioactive gas absorption by single droplets and by rain was overviewed quite comprehensively (see e.g. by Piskunov et al., 2012; Nankara et al., 2012). These previous investigations are summarized in Table 1. Note that most published investigations of radioactive gas absorption by falling liquid droplets and by precipitation are devoted to HTO absorption by water. Since the lifetime of radioactive HTO is by orders of magnitudes larger than the total time of soluble gas scavenging, these studies do not take into account radioactive decay in the equation of mass balance. Theoretical models which allow us to take into account a radioactive decay during gas absorption by a single stagnant and falling droplet are presented in the Appendixes A and B, respectively. Taking into account radioactive decay is important e.g. in the analysis of absorption of radioactive gaseous iodine-131 because the lifetime of radioactive I-131 is of the same order of magnitude as the time of complete gas absorption by a droplet.

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., 2013b), 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

Table 1
Scavenging of soluble radioactive gases by a single droplets and rain.

Problem	Experimental or theoretical	References
Single droplet	Experimental	Booker, 1965; Belovodsky et al., 1997.
Single droplet	Theoretical	Booker, 1965.
Precipitation	Experimental	Dana et al., 1978; Köllö et al., 2011; Abrol, 1990; Nankara et al., 2012; Matsumoto et al., 2013; Piskunov et al., 2012; Gautam et al., 2013
Precipitation	Theoretical	Chamberlain and Eggleton 1964; Dana et al., 1978; Atanassov and Galeriu 2011; Patry et al., 2011