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Scavenging of soluble trace gases by falling rain droplets in inhomogeneous atmosphere

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ABSTRACT

We analyze non-isothermal absorption of trace gases by the rain droplets with internal circulation which is caused by interfacial shear stresses. It is assumed that the concentration of soluble trace gases and temperature in the atmosphere varies in a vertical direction. The rate of scavenging of soluble trace gases by falling rain droplets is determined by solving heat and mass transfer equations. In the analysis we accounted for the accumulation of the absorbate in the bulk of the falling rain droplet. The problem is solved in the approximation of a thin concentration and temperature boundary layers in the droplet and in the surrounding air. We assumed that the bulk of a droplet, beyond the diffusion boundary layer, is completely mixed and concentration of the absorbate and temperature are homogeneous and time-dependent in the bulk. By combining the generalized similarity transformation method with Duhamel's theorem, the system of transient conjugate equations of convective diffusion and energy conservation for absorbate transport in liquid and gaseous phases with time-dependent boundary conditions is reduced to a system of linear convolution Volterra integral equations of the second kind which is solved numerically. Calculations are performed using available experimental data on concentration and temperature profiles in the atmosphere.

It is shown than if concentration of a trace gas in the atmosphere is homogeneous and temperature in the atmosphere decreases with height, beginning from some altitude gas absorption is replaced by gas desorption. Neglecting temperature inhomogenity in the atmosphere described by adiabatic lapse rate leads to essential overestimation of the trace gas concentration in a droplet on the ground.

1. Introduction

Gas absorption by the falling rain droplets is of relevance in meteorology and environmental engineering. Rains play an important role in wet removal of gaseous pollutants from the atmosphere. Furthermore, scavenging of soluble hazardous gases by rains is an important part of self-cleansing process of the atmosphere (see, e.g. Flossmann, 1998; Pruppacher and Jaenicke, 1995).

Scavenging of atmospheric gaseous pollutants by rain droplets is a result of a gas absorption mechanism (Pruppacher and Klett, 1997). Comprehensive study of mass transfer during gas absorption by falling rain droplets is also required for predicting transport of hazardous gases in the atmosphere.

Concentration measurements of CO_2, SO_2, NH_3 and other trace gases in the atmospheric boundary layer revealed vertical (altitudinal) dependence of the concentrations (see Georgii, 1978; Gravenhorst et al., 1978; Georgii and Müller, 1974; Denning et al., 1995). Concentration of gases which are not associated with photosynthesis, e.g. SO_2 and NH_3, has a maximum at the Earth surface and decreases with height over the continents. The concentration of NH_3 over the continents decreases rapidly with altitude, reaching a constant background concentration in winter and warm days at the altitudes of about 1500 m above the ground and 3000 m above the ground, respectively (see Georgii and Müller, 1974; Georgii, 1978). On warm days in summer the ground concentration of NH_3 is considerably higher than that on the cold days in winter. Sulfur dioxide concentration in the ABL (atmospheric boundary layer) is higher during winter than during summer because of the higher anthropogenic SO_2 production caused by enhanced combustion of fossil fuels in winter. Diurnal and seasonal variations of CO_2 distribution with altitude occur due to the competition between different phenomena, e.g. photosynthesis, respiration and thermally driven buoyant mixing (see Denning et al., 1995). Information about the evolution of the vertical profile of soluble gases with time allows calculating fluxes of these gases in an the ABL. Vertical transport of

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Nomenclature

a	square root of the thermal diffusivities ratio, $\sqrt{a_1/a_2}$
c_p	specific heat, kJ kmole^{-1} K^{-1}
C_i	molar density at the bulk of fluid, mol m^{-3}
D_i	molecular diffusion coefficient, m^2 s^{-1}
D	square root of the diffusivities ratio, $\sqrt{D_1/D_2}$
H_A	Henry's law constant, mole litre^{-1} atm^{-1}
k	coefficient in Eq. (1)
$N_{Di} = -D_iC_i(\partial x_i/\partial y)$	molar flux density, mol m^{-2} s^{-1}
$N_{T_i} = -\lambda_i \partial T_i/\partial y$	heat flux density, W m^{-2}
m	distribution coefficient
m_0	distribution coefficient at temperature T_{20}
$Pe_{D_i} = kRU/D_i$	Peclet number for a moving droplet
$Pe_{T_i} = kRU/a_i$	Peclet number for a moving droplet
r	radial coordinate, m
R	droplet radius, m
t	time, s
T	temperature, K
U	translational velocity of a droplet, m s^{-1}
v_r, v_ϑ	velocity components, m s^{-1}
x	molar fraction of an absorbate
x_{b10}	initial value of molar fraction of absorbate in a droplet
x_{b20}	value of molar fraction of an absorbate in a gas phase at height H
x_{b200}	value of molar fraction of absorbate in a gas phase on the ground
$x_{b1}(t)$	molar fraction of absorbate in a bulk of a droplet
$x_{b2}(t)$	molar fraction of an absorbate in a bulk of a gas phase
$X_1(t) = x_1(t)/m_0x_{20}$	normalized molar fraction of an absorbate in a liquid phase
$X_2(t) = x_2(t)/x_{20}$	normalized molar fraction of an absorbate in a gaseous phase
y	distance from the surface of a droplet, m
$Y = y/R$	dimensionless distance from the surface of a droplet
z	coordinate in a vertical direction, m

Greek symbols

$\Theta_i = T_i/T_{10}$	dimensionless temperature
$\gamma = C_1/C_2$	molar densities ratio
δ_{D_i}	thickness of a diffusion boundary layer, m
δ_{T_i}	thickness of a thermal boundary layer, m
$\Delta_i = \delta_i/R$	dimensionless thickness of a boundary layer
λ	thermal conductivity, W m^{-1} K^{-1}
ξ	variable
η_i	similarity variable
ϑ	angle
$\tau = tUk/R$	dimensionless time

Subscripts

0	value at the height H in the atmosphere
1	liquid phase
2	gaseous phase
b	value in the bulk
r	radial direction
ϑ	tangential direction
s	value at gas–liquid interface

soluble gases in the ABL is an integral part of the atmospheric transport of gases and is important for understanding the global distribution pattern of soluble trace gases. An improved understanding of the cycle of soluble gases is also essential for the analysis of global climate change. Clouds and rains play essential role in vertical redistribution of SO_2, NH_3 and other soluble gases in the atmosphere. Scavenging of soluble gases, e.g. SO_2, NH_3 by rain affects the evolution of vertical distribution of these gases. At the same time the vertical gradients of the soluble gases concentration in the atmosphere affect the rate of gas absorption by rain droplets. Note that the existing models of global transport in the atmosphere (see, e.g. de Arellano et al., 2004) do not take into account the influence of rains on biogeochemical cycles of different gases.

Existence of the vertical temperature distribution in the atmosphere was discovered in 1749 by A. Wilson (see Wilson, 1826). Inspired by Wilson, numerous measurements and modeling of vertical temperature distribution in the atmosphere (see, e.g. Dines, 1911; Taylor, 1960; Manabe and Strickler, 1964; Manabe and Wetherald, 1967) revealed 6.5 K km^{-1} lapse rate. Evolution of the lapse rate during the last decades is discussed by Trenberth and Smith (2006). Vertical temperature profiles in atmosphere during nocturnal inversions were investigated experimentally and theoretically, e.g. by Anfossi et al. (1976), Surridge (1986), Surridge and Swanepoel (1987), Anfossi (1989). Analytical expressions for nocturnal temperature distribution in the atmosphere and for decrease of temperature at a ground during the night were determined by Surridge (1986) and Anfossi (1989). Influence of vertical distribution of temperature in the atmosphere on the rate of gas scavenging by falling rain droplets is explained by a strong nonlinear dependence of the solubility parameter (Henry's constant) for aqueous solutions of different gases on temperature (see, e.g. Reid et al., 1987). Accounting for vertical distributions of absorbate and temperature in the gaseous phase requires solution of heat and mass transfer equations which describe gas absorption by falling rain droplet.

Due to the differences in solubility of gases in liquids, mass transfer during absorption of soluble gas by droplets in the presence of inert admixture can be continuous-phase controlled, liquid-phase controlled or conjugate. Continuous-phase controlled mass transfer by falling droplets was discussed by Kaji et al. (1985), Altwicker and Lindhjem (1988), Waltrop et al. (1991) and Saboni and Alexandrova (2001). Liquid-phase controlled mass transfer was studied, e.g. by Amokrane and Caussade (1999) and Chen (2001). Mass transfer controlled by both phases was analyzed by Walcek and Pruppacher (1984), Alexandrova et al. (2004), Chen (2004), Elperin and Fominykh (2005), Elperin et al. (2007a,b, 2008, 2009), Kroll et al. (2008).

Accumulation of the dissolved atmospheric gases in a falling water droplet during absorption in the presence of vertical distribution of the absorbate concentration and temperature in a gaseous phase and circulation of fluid inside a droplet caused by shear stresses at the interface is determined by a system of unsteady convective diffusion and energy conservation equations with time-dependent boundary conditions. Analytical solution of these equations requires application of rather sophisticated methods (see, e.g. Bartels and Churchill, 1942; Ruckenstein, 1967).

The effect of altitudinal distribution of the absorbate in a gaseous phase on the rate of gas absorption by falling rain droplets was investigated by Elperin et al. (2009). The suggested approach includes applying the generalized similarity transformation to a system of transient equations of convective diffusion and a Duhamel's theorem. Then the problem reduces to a numerical solution of a linear convolution Volterra integral equation of the second kind.

In the present study we investigate simultaneous effect of the absorbate and temperature inhomogenity in a vertical direction in a gaseous phase on the rate of gas absorption by falling droplets.