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Uptake of soluble gaseous pollutants by rain droplets in the atmosphere with nocturnal temperature profile

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A B S T R A C T

We analyze the uptake of gaseous pollutants by the rain droplets falling in the atmosphere with nocturnal temperature inversion. The rate of uptake of soluble trace gases by falling rain droplets is determined by solving energy and mass conservation equations. In the analysis we accounted for the accumulation of the soluble gas and energy 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 vicinity of the droplet surface. It is assumed that the bulk of a droplet, beyond the diffusion boundary layer, is completely mixed and distributions of concentration of the absorbate and temperature are homogeneous and time-dependent in the bulk. The problem 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 nocturnal temperature profiles in the atmosphere. It is shown than if the concentration of gaseous pollutants in the atmosphere is homogeneous and the absolute temperature in the atmosphere increases with altitude, a droplet absorbs gas during all the period of its fall. In the case when the temperature–altitude curve comprises the nocturnal inversion and temperature fall segments, gas absorption by a falling rain droplet can be replaced by desorption and vice versa. Neglecting temperature inhomogeneity in the atmosphere caused by nocturnal temperature profile leads to significant underestimation of the concentration of gaseous pollutants inside a droplet on the ground. The calculations performed using temperature profiles measured by Corsmeier et al. (1997) showed that the underestimation of the concentration of gaseous pollutants in rain droplets at the ground can exceed 20%.

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. 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. Vertical transport of soluble gases in the atmospheric boundary layer (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 enhanced understanding of the cycle of soluble gases is also important for the analysis of global climate change (see e.g., Aalto et al., 2006). Clouds and rains play a significant role in vertical redistribution of SO_2, NH_3 and other soluble gases in the atmosphere (see, e.g. Zhang et al., 2006). Scavenging of soluble gases, e.g., SO_2, and NH_3 by rain affects the evolution of vertical distribution of these gases. At the same time the vertical gradients of the soluble gas concentration in the atmosphere affect the rate of gas absorption by rain droplets. Notably, 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.

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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 Wetherald, 1967) revealed the existence of a $6.5\,\mathrm{K \cdot km^{-1}}$ lapse rate. Evolution of the lapse rate during the last decades is discussed by Trenberth and Smith (2006).

Enhanced interest to a problem of nocturnal temperature inversion during the last decades can be explained by the importance of these meteorological conditions for the dispersion of air pollutants and for fog and frost formation. In his pioneering theoretical study Brunt (1934) showed that infrared radiative transfer behaves like a diffusive process leading to the exponential type temperature profile with a strong negative curvature. Brunt (1934) assumed that the heat flux at the ground is constant throughout the night. Later studies were devoted to forecasting of temperature profile evolution in the lower layers of the atmosphere, the height of the inversion layer and inversion strength (see, e.g. Anfossi et al., 1976; Surridge and Swanepoel, 1987; Anfossi, 1989; Gassmann and Mazzeo, 2001). Two latter parameters are related to the radiation heat flux from the ground to the night sky and the rate at which heat propagates through the atmosphere to the ground. It was demonstrated (see, e.g. Anfossi, 1989) that the height of the inversion layer can expand to hundreds of meters.

Influence of vertical distribution of the 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 the temperature (see, e.g. Reid et al., 1987). Accounting for vertical distributions of soluble gaseous pollutants and temperature in the atmosphere requires solution of energy and mass conservation equations which describe gas absorption by falling rain droplets.

Due to the differences in solubility of gases in liquids, mass transfer during absorption of a soluble gas by droplets in the presence of an 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 Wurzler (1998). 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), Chen (2004), Elperin and Fominykh (2005), Elperin et al. (2007, 2008, 2009).

Accumulation of the dissolved atmospheric gases in a falling water droplet during gas absorption is determined by a system of unsteady convective diffusion and energy conservation equations. An analytical solution of these equations requires application of rather sophisticated methods (see, e.g. Ruckenstein, 1967). Moreover, in the Earth atmosphere the problem is complicated by the vertical gradients of the absorbate concentration and temperature in the gaseous phase.

The effect of altitudinal distribution of the soluble gases in the atmosphere 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 Duhamel's theorem. Then the problem reduces to the numerical solution of a linear-convolution Volterra integral equation of the second kind. Note that in our previous study (Elperin et al., 2009) it was assumed that the atmospheric temperature distribution is uniform.

In the present study we investigate the effect of the nocturnal temperature profile in the atmosphere on the rate of gas absorption by falling droplets. In the case of the uniform vertical distribution of the trace gas and temperature in the atmosphere the solution obtained in the present study recovers the expressions obtained in our previous study (Elperin and Fominykh, 2005). The latter approach was validated by comparing the numerical solution with available experimental data for CO_2 and SO_2 absorption by a falling droplet (see Altwicker and Lindhjem, 1988; Amokrane and Caussade, 1999).

2. Description of the model

Consider absorption of a soluble gas from a mixture containing an inert gas by a moving droplet. At time $t=0$ the droplet begins to absorb gas from the atmosphere. Distribution of the concentration of the absorbate and temperature in the gaseous phase in the vertical direction is assumed to be known. Schematic view of a falling droplet with the attached frame of coordinates is shown in Fig. 1.

In the analysis we account for the resistance to heat and mass transfer in both phases and use the following simplifying assumptions: 1) we employ the approximation of the infinite dilution of the absorbate in the absorbent; 2) thicknesses of the diffusion and temperature boundary layers in both phases are assumed small compared with the droplet's size; 3) tangential molecular heat and mass transfer rates along the surface of a spherical droplet are assumed small compared with the molecular heat and mass transfer rates in the normal direction;

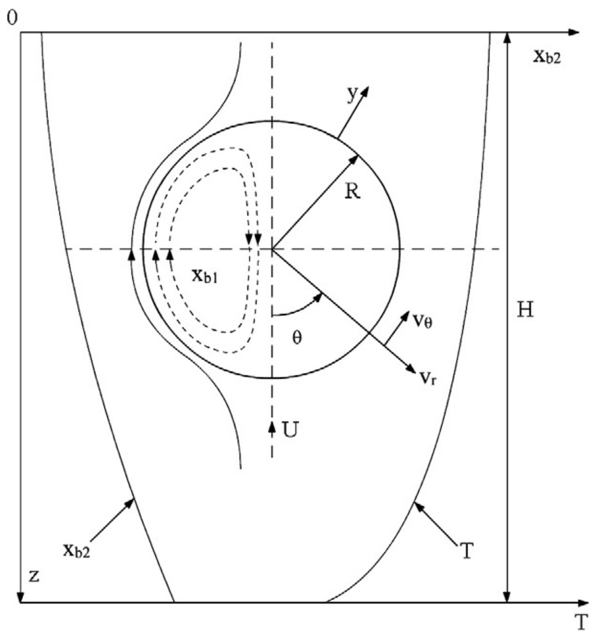

Fig. 1. Schematic view of a falling droplet.