## The cloud expansion due to the Stefan outflow at nonisothermal condensation in multicomponent vapour-gas mixture

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As we have shown (Kuchma and Shchekin, 2012; Kuchma et al, 2016), nonstationary heating (due to releasing the condensation heat) a vapour-carrier gas mixture around a growing droplet produces the Stefan gas outflow at larger distances from the droplet. For an ensemble of droplets in the cloud, such effect provides an increase of volume of the cloud, what can be treated as cloud thermal expansion.

Here we extend the results to general case of multi-component nonisothermal condensation. We consider a small single free droplet of radius R growing in the diffusion regime in the atmosphere of several condensable vapours. As had been shown (Kulmala et al, 1993; Martyukova et al, 2016), after fast transition stage, the droplet grows with a fixed composition and droplet temperature  $T_d$ . For a mixture of ideal gases, we can represent the local mean velocity u(r,t) of the flow of the gas mixture at the distance r and time t due to nonisothermal condensation into droplet as

$$u(r,t) = \dot{R} \frac{R^2}{r^2} \frac{T(r,t)}{T_d} \left( 1 - \frac{T_d}{v_l(\{x\}, T_d)\tilde{n}T_0} + \frac{T_d}{R^2 \dot{R}} \int_{R(t)}^r dr_l r_l^2 \frac{1}{T^2(r_l,t)} \frac{\partial T(r_l,t)}{\partial t} \right).$$
(1)

Here  $\dot{R}$  is the growth rate of the droplet radius in time, T(r,t) is the local temperature in the vapor-gas mixture,  $v_l({x}, T_d)$  is the mean volume per molecule in the liquid solution with *k* components,  ${x} \equiv {x_1, x_2, ..., x_k}$ ,  $x_i$  is the relative molecular concentration of *i*-th component,  $\tilde{n}$  is the total vapour-gas concentration as  $r \rightarrow \infty$ ,  $T_0$  is the initial value of system temperature.

It follows from eq.(1) at  $r \to R(t)$ ,  $\dot{R} > 0$  and  $\tilde{n}v_t << 1$  that

$$u(R,t) \approx -\dot{R} \frac{T_d}{\tilde{n} v_l \left( \left\{ x \right\}, T_d \right) T_0} < 0 , \qquad (2)$$

and the Stefan flow is directed to the droplet. For a stationary heat conduction with  $\partial T(r,t)/\partial t = 0$ , the local velocity u(r,t) stays negative everywhere, and the cloud compresses. The non-stationary effects give rise to the last positive term in eq.(1) able to change the sign of the velocity u(r,t) at larger r. The condition of enthalpy balance leads to the expression

$$u(r,t) \approx \frac{R^2(t)\dot{R}(t)}{r^2} \frac{T_d}{T_0} \frac{1}{\tilde{n}v_l(\lbrace x \rbrace, T_d)} \left(\frac{q(\lbrace x \rbrace, T_d)}{h_g(T_d)} - 1\right) (3)$$

which is valid outside the nonstationary diffusion shell around the droplet. Here  $q(\lbrace x \rbrace, T_d)$  is the mean evaporation heat per molecule in the liquid solution with composition  $\lbrace x \rbrace$ ,  $h_g$  is the enthalpy per molecule of the carrier gas. Since  $q(\lbrace x \rbrace, T_d)/h_g(T_d) >> 1$ , the velocity u(r,t) in eq.(3) is positive, and the Stefan flow now produces the thermal expansion of the cloud.

To describe all the details in the behavior of the velocity u(r,t) analytically, we took the case of non-stationary self-similar regime of multi-component non-isothermal droplet growth. Introducing self-similar variable  $z \equiv r/R(t)$  and assuming  $(T(z)-T_0)/T_0 \ll 1$ , we have from eq.(1):

$$u(z) \approx -\frac{\dot{R}(t)}{z^{2}} \left( \frac{1}{v_{l}(\{x\}, T_{d})\tilde{n}} + \frac{1}{T_{0}} \int_{1}^{z} dz_{1} z_{1}^{3} \frac{dT(z_{1})}{dz_{1}} \right).$$
(4)

The self-similar solution of the thermal conductivity equation has the form (Kuchma et al, 2015)

$$T(z) = T_{d} - \left(T_{d} - T_{0}\right) \frac{\int_{1}^{z} \frac{dz_{1}}{z_{1}^{2}} exp\left[-\frac{R\dot{R}}{2t}\left(z_{1}^{2} + \frac{2}{z_{1}} - 3\right)\right]}{\int_{1}^{\infty} \frac{dz_{1}}{z_{1}^{2}} exp\left[-\frac{R\dot{R}}{2t}\left(z_{1}^{2} + \frac{2}{z_{1}} - 3\right)\right]}$$
(5)

where  $\chi$  is the thermal diffusivity of the vapour-gas mixture, parameters  $R\dot{R}/t$  and  $T_d - T_0$  are determined by the boundary conditions at the droplet surface and are related to the droplet composition. Using eq.(5), we have from eq.(4):

$$u(z) \approx \frac{\dot{R}(t)}{v_{l}(\{x\}, T_{d})\tilde{n}z^{2}} \left[\frac{q(\{x\}, T_{d})}{h_{g}(T_{0})} \left(1 - e^{-\frac{R\dot{R}}{2t}(z^{2} - 1)}\right) - 1\right].$$
 (6)

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