

Thanks for all the comments and recommendation to publish. We have considered every point and corrected the paper to include their points. The editor comments are in black, responds from the authors are in blue and revised text are in red.

One point is connected to the thickness of the DI and eq. 14 What means infinitesimal thickness mathematically precisely and how do you define a concentration in such a DI with a volume of or approaching 0? With this question, I also refer to the molecular budget, which I'm sure you have looked at, but which would be worth mentioning.

Appendix B has been added to descript the deviation of equation 14.

Appendix B: Derivation for non-equilibrium kinetics

850 The processes involved in the equilibrium of the gas-phase and the surface of a droplet (Fig. A5):
 1) Gas-phase diffusion from far away ($> \mu\text{m}$) from the droplet to the surface of the droplet, which is likely to be driven by turbulence and molecular diffusion; 2) Interfacial mass transport; and 3) Condensed-phase diffusion and chemical reactions;

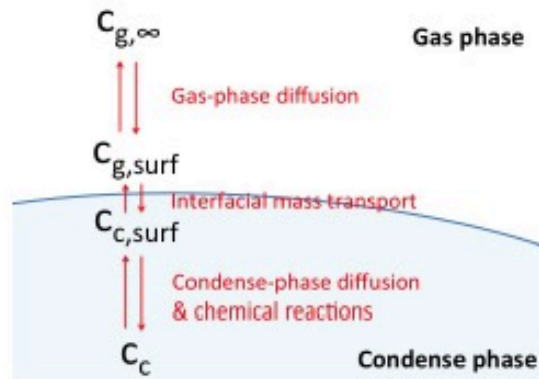


Figure A5. Processes involve in the equilibrium between gas-phase and condensed-phase, where $c_{g,\infty}$ is the gas-phase concentration in the SIA far away from the droplet, $c_{g,surf}$ is the gas-phase concentration at the surface (outside the droplet), $c_{c,surf}$ is the condensed-phase concentration at the surface (inside the droplet) and c_c is the average condensed-phase concentration.

Transport of gas-phase species from the SIA to the surface of the droplet can be described using
 855 Fick's law as diffusion flux, J_g :

$$J_g = -D_g \frac{dc_g}{dx} \quad (\text{B1})$$

where D_g is the gas-phase diffusivity, and $\frac{dc_g}{dx}$ is the concentration gradient at the droplet surface that $\frac{dc_g}{dx} = \frac{c_{g,\infty} - c_{g,surf}}{R_{eff}}$ with R_{eff} as the radius of the droplet. The concentration change in the condensed-phase can be expressed as

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$$\frac{dc_c}{dt} = \frac{A J_g}{V} = -\frac{A D_g}{V R_{eff}} (c_{g,\infty} - c_{g,surf}) \quad (\text{B2})$$

where A is the surface area of the droplet and V is the volume of the droplet. The first-order rate coefficient for the gas-phase diffusion process can be defined as $k_{dg} = \frac{A D_g}{V R_{eff}}$ (Sander, 1999). For an example, a liquid droplet with a radius R_{eff} the gas-phase diffusion rate coefficient $k_{dg} = \frac{3D_g}{R_{eff}^2}$.

The interfacial mass transport from gas-phase to condensed-phase can be expressed in terms of
 865 accommodation coefficient, α . The flux through the phase boundary into the droplet, J_b^{in} , is defined as:

$$J_b^{in} = \frac{\alpha \bar{v}}{4} c_{g,surf} \quad (\text{B3})$$

where the subscript b stands for 'boundary' and \bar{v} is the mean molecular velocity. The opposite flux, J_b^{out} , through the phase boundary out of the droplet can be expressed in the similar form as Eq. 870 **B3** that $J_b^{out} = \frac{\alpha_a \bar{v}_c}{4} c_{a,surf}$, where \bar{v}_c is the mean molecular velocity in condensed-phase and α_c is the condensed-phase accommodation coefficient. The net flux through the grain boundary, J_b , is the difference between the in and out flux.

$$J_b = J_b^{in} - J_b^{out} = \frac{\alpha \bar{v}}{4} \left(\frac{c_{c,surf}}{K} - c_{g,surf} \right) \quad (B4)$$

where K is the equilibrium constant, of which $K = c_{c,surf}^{eq}/c_{g,surf}^{eq}$. For example, for a gas-aqueous 875 interface, the ratio of aqueous-phase concentration to gas-phase concentration at equilibrium can be described as $c_{a,surf}^{eq}/c_{g,surf}^{eq} = k_H^{cc}$, where $c_{a,surf}$ is the aqueous-phase concentration at the surface and k_H^{cc} is the Henry's constant. The concentration change in the condensed phase due to interfacial mass transport can be expressed as:

$$\frac{dc_c}{dt} = -\frac{A J_b}{V} = \frac{A \alpha \bar{v}}{V} \left(c_{g,surf} - \frac{c_{c,surf}}{K} \right) \quad (B5)$$

880 The first-order rate coefficient for the interfacial mass transport, k_b , to a droplet with a radius R_{eff} can then be defined as $k_b = \frac{3\alpha \bar{v}}{4} R_{eff}$. By assuming the fluxes of gas-phase diffusion, J_g , is equal to the interfacial mass transport, J_b , the rate of change of concentration in the condensed phase can be expressed as

$$\frac{dc_c}{dt} = \frac{A}{V} \left(\frac{R_{eff}}{D_g} + \frac{4}{\bar{v}\alpha} \right)^{-1} \left[c_{g,\infty} - \frac{c_{c,surf}}{K} \right] \quad (B6)$$

885 the term $\frac{A}{V} \left(\frac{R_{eff}}{D_g} + \frac{4}{\bar{v}\alpha} \right)^{-1}$, is often referred as the mass transfer coefficient, k_{mt} , for a chemical species transfer from air to liquid/solid. The mass transfer coefficient for chemical into a spherical droplet with radius R_{eff} is $k_{mt} = \left(\frac{r^2}{3D_g} + \frac{4R_{eff}}{3\bar{v}\alpha} \right)^{-1}$ and if the surface of the droplet is described as DI then the concentration at the grain surface, $c_{c,surf} = [HNO_3,DI]$.

Did I understand correctly, that the flux into the DI matches the flux from the DI into the bulk and that the concentration in the DI based on Henry is established? **Yes, that is correct.**

If so, I do not understand the statement on page 25 (825): "In general, the grain boundary concentration of nitrate defined by solvation into the DI is much larger than when it is defined by the combination of surface adsorption and co-condensation on ice." Does this imply that the fluxes are balanced?

The statement is refer to the boundary of the snow grain, the statement is now written as follow for clarity

"The concentration of the nitrate at the grain boundary, $U(R_{eff})$, have a much larger value when the interface between air and grain boundary is defined as 'Air-DI' (Eq. 13) than when it is defined as 'Air-Ice' (Eq. 7)."

The second aspect touches question 2 of report 1: You don not explicitly mention an upper limit of solubility in the bulk ice. That of the DI is given by Henry. That of the inner ice is given by the solid solution? If correct, I suggest to clearly state this (lines 216-230) to prevent the impression that the whole grain can become liquid-like as the DI holding such high concentrations of solutes.

It has been clarified in P.8 line 239

" 3) the DI has an infinitesimal thickness and the concentration of nitrate in the DI is acting as the boundary condition of the solid-state diffusion into the snow grain, which the solid-state concentration of nitrate in the bulk is limited by the solubility of ice. "

Some minor suggestions:

Page 3, line 90: Please define Co-condensation here.

Definition of co-condensation is added: “contributed by co- condensation, which is the simultaneous condensation of water vapour and trace gases at the air-ice interface, has an empirical relationship”

Page 4, line 124. I would not say that the idea of liquid co-existing with ice comes from the Domine paper. This is given by thermodynamics; Domine strongly argued for arrangement in pockets as you state later. I suggest to rather cite Cho or McNeill, ACP (2011)

The statement (P.4 line 113) is now cited to Cho et. al. (2002)

Page 7, line 210: I suggest to mention that the pH in the liquid content of the ice is not equal to the pH of molten snow.

The following sentence been added on P.7 line 197-199

“ Note that the range of pH measured by Udisti et al. (2004) is the pH of the melted sample, which might be different from the pH of the ice co-existed liquid. However, the pH of the liquid water co-existing with the ice cannot be measured with the current techniques yet. “

Page 9, line 275: The adsorption as shown by Ullerstam might be under saturated, but it is still in equilibrium. So, I can't quite follow this argument.

The sentences have now been rearrange to explain why the ice surface is not saturated and not in equilibrium. P. 8, line 256 – 263:

“Ullerstam et al. (2005b) have shown that for partial pressures of HNO₃ lower than 10⁻⁵ Pa the ice surface is not entirely covered with HNO₃, and therefore, undersaturated. The annual average atmospheric partial pressure of HNO₃ recorded at Dome C is ~ 10⁻⁶ Pa (Traversi et al., 2014) and is ~ 10⁻⁷ Pa at Halley (Jones et al., 2008), hence, the ice surface is unlikely to be saturated with HNO₃. A non-equilibrium kinetic approach is taken instead of an equilibrium adsorption as natural snowpacks are constantly undergoing sublimation and condensation of H₂O, especially at the skin layer, due to temperature gradient over a range of timescales from a fraction of seconds to days and seasons (Bartels-Rausch et al., 2014).”

Page 12, line 371:replace water with brine or solution.

“liquid water ” has been replaced with “liquid solution”