1. Introduction
This paper presents a theoretical framework describing the thermodynamics and phase transformation of a three phase component system consisting of ice particles, liquid droplets and water vapour. The instant rates of changes of water ($q_w$), ice ($q_i$) and vapour ($q_v$) mixing ratios are described based on the quasi-steady approximation, which assumes that the sizes of cloud particles stay constant (Squires 1952). This assumption allows for the analytical solution of the differential equation for supersaturation in mixed phase clouds (Korolev and Mazin 2003). The local thermodynamical conditions required for the equilibrium of liquid ($q_w=0$), ice ($q_i=0$) and vapour ($q_v=0$) phases are analysed. It is shown that there are four different regimes of the partitioning of water between liquid, ice, and gaseous phases in mixed clouds. The Wegener-Bergeron-Findeisen (WBF) process is identified as being relevant to two of those regimes. The efficiency of the WBF process in characterizing the capability of ice crystals to deplete the water evaporated by liquid droplets is introduced here. It is shown that the WBF process has maximum efficiency at approximately zero vertical velocity. The analysis of the dependences of $q_w$, $q_i$ and $q_v$ on the vertical velocity, temperature, pressure and the integral radii of the cloud particles is presented. It is shown that the maximum rates of ice growth and droplets evaporation does not necessarily occur at $T=-12^\circ C$ where the maximum difference between saturation vapour pressure over ice and that over liquid is observed.

2. Rates of vapour, ice and liquid mass changes in mixed clouds
In the following discussion we consider an idealized adiabatic mixed phase cloud consisting of liquid droplets and ice particles suspended in water vapour. The interaction between ice particles, liquid droplets and water vapour occurs through processes of diffusional growth and/or evaporation. The processes related to particle-to-particle interaction, like riming, aggregation, and coagulation are not considered here. It is assumed that the spatial distribution of ice particles, liquid droplets, water vapour and temperature is uniform. The concentrations of liquid droplets and ice particles stay constant with time and the radiation effects are neglected. It is recognized that this idealization means that some of the quantitative results will not be directly applicable to real cloud systems and this will be addressed in section 7. However, such simplifications allow us to get a good theoretical understanding of mixed-phase phenomenon.

The rate of mass change of an ensemble of liquid droplets can be described as (e.g. Squires 1952, Korolev and Mazin 2003)

$$\frac{dq_w}{dt} = B_w S_w N_w \bar{r}_w$$

(1)

here $q_w$ is the liquid water mixing ratio; $S_w = \frac{e}{E_w} - 1$ is the supersaturation over water; $e$ is water vapour pressure; $E_w(T)$ is saturation water vapour pressure over water at the temperature $T$; $N_w$ and $\bar{r}_w$ are the concentration and average radius of the droplets, respectively; $B_w$ is a coefficient dependent upon $T$ and $P$ (an explanation of all the variables used in the text is provided in Appendix A).

Similar to the liquid droplets, the rate of mass change of ice particles can be written as

$$\frac{dq_i}{dt} = B_{10} S_i N_i \bar{r}_c$$

(2)

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Here, $S = \frac{e - E}{E_i} - 1$ is the supersaturation over ice; $E(T)$ is saturation water vapour pressure over ice; $B_{0}$ is a coefficient dependent upon $T$ and $P$; $N_{i}$ is the concentration of ice particles, $r_{i}$ is the characteristic size of ice particles; $c$ is a shape factor of ice particles characterizing “capacitance” in the equation of the growth rate.

In the following consideration, the size $r_{i}$ will be defined as a half of a maximum dimension of the ice particle. For this definition of $r_{i}$, the shape factor varies in the range $0 < c \leq 1$, being equal to 1 for ice spheres. For simplicity in the frame of this study, we assume $c = \text{const}$. This simplification does not disturb the generality of the following consideration. However, for an ensemble of ice particles having different shapes one should consider the average value of the product $c r_{i}$.

The products $N_{w} r_{w}$ and $N_{i} r_{i}$ (or the first moments of particle size distributions) will be referred to as the “integral radii” of the droplets and ice particles, respectively.

The supersaturation over ice can be related to the supersaturation over water as

$$1 - \frac{\xi}{\xi_{w}} = \frac{S_{w}}{S_{i}}$$

(3)

where $\xi(T) = \frac{E_{w}(T)}{E_{i}(T)}$. Then, substituting Eq.3 into Eq.2 yields

$$\frac{dq_{i}(t)}{dt} = (B_{i} S_{w} + B_{w}) N_{i} r_{i}$$

(4)

Here, $B_{i} = \xi c B_{0}$, and $B_{w} = (\xi - 1) c B_{0}$.

The supersaturation $S_{w}$ in Eqs. 1 and 4 can be approximated by the quasi-steady supersaturation (Korolev and Mazin, 2003)

$$S_{qsw} = \frac{a_{0} u_{z} - b_{w}^{*} N_{w} r_{w}}{b_{w} N_{w} r_{w} + b_{i} N_{i} r_{i}}$$

(5)

The remarkable property of the quasi-steady supersaturation is that the actual supersaturation $S_{w}$ approaches with time to $S_{qsw}$ calculated for current values of $N_{w}$, $N_{i}$, $r_{w}(t)$, $r_{i}(t)$, $T(t)$ and $P(t)$, i.e.

$$\lim_{t \to \infty} S_{w}(t) = S_{qsw}(t)$$

(6)

For clouds with typical integral radii $N_{w} r_{w}$ and $N_{i} r_{i}$, the difference between $S_{w}$ and $S_{qsw}$ usually becomes less than 10% within a time period $3 \tau_{p}$, where

$$\tau_{p} = \frac{1}{a_{0} u_{z} + b_{w} N_{w} r_{w} + (b_{i} + b_{w}^{*}) N_{i} r_{i}}$$

(7)

is the time of phase relaxation (Korolev and Mazin 2003).

In mixed phase clouds $\tau_{p}$ is mainly defined by the integral radius of liquid droplets $N_{w} r_{w}$, and typically, it does not exceed a few seconds. This time is significantly less than the characteristic lifetime of mixed phase clouds ($\tau_{c}$), i.e. $\tau_{c} >> \tau_{p}$. Therefore, the supersaturation $S_{w}$ has enough time to relax to $S_{qsw}$. This justifies the use of $S_{qsw}$ as an approximation of $S_{w}$ in calculations of the rate of mass changes of ice and liquid.

Thus, substituting Eq.5 into Eq.1 gives the rate of change of liquid water mixing ratio,

$$\dot{q}_{w} = \left(\frac{a_{0} u_{z} - b_{w}^{*} N_{w} r_{w}}{b_{w} N_{w} r_{w} + b_{i} N_{i} r_{i}}\right) B_{w} N_{w} r_{w}$$

(8)

Similarly, substituting Eq.5 into Eq.4, and taking into account that $B_{i}^{*} b_{i} = b_{w}^{*} B_{w}$ (Appendix A) we obtain the rate of change of ice water mass

$$\dot{q}_{i} = \left(\frac{a_{0} u_{z} - \frac{1 - \xi}{\xi} b_{w} N_{w} r_{w}}{b_{w} N_{w} r_{w} + b_{i} N_{i} r_{i}}\right) B_{i} N_{i} r_{i}$$

(9)

The rate of changes of the water vapour mixing ratio ($\dot{q}_{v}$) can be found from the equation of mass conservation

$$\dot{q}_{w} + \dot{q}_{i} + \dot{q}_{v} = 0$$

(10)

Substituting Eqs. 8 and 9 into Eq.10 yields

$$\dot{q}_{v} = \frac{B_{w} B_{i}^{*} (a_{1} - a_{2}) N_{w} r_{w} N_{i} r_{i} - a_{0} u_{z} (B_{w} N_{w} r_{w} + B_{i} N_{i} r_{i})}{b_{w} N_{w} r_{w} + b_{i} N_{i} r_{i}}$$

(11)

The replacement of $S_{w}$ with its quasi-steady approximation $S_{qsw}$ (Eq.5) raises questions concerning the accuracy of this replacement and the factors limiting the use of Eqs. 8 and 9. The accuracy of the quasi-steady approximation has

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1 At the final stage of droplet evaporation, when $N_{w} r_{w}$ becomes small, the time of phase relaxation will be mainly defined by the integral radii of ice particles $N_{i} r_{i}$, which eventually results in an increase of $\tau_{p}$. 
been tested by comparing $S_w$, $q_i$ and $q_w$ derived from Eqs. 5, 8 and 9, respectively, with that calculated from a parcel model of a vertically moving adiabatic clouds. The parcel model used here is similar to that described in Korolev and Mazin (2003). Figure 1 shows the results of the comparison of $S_w$, $q_i$ and $q_w$ for a set of modelled clouds, where the integral radii of droplets and ice particles were changed in the range of $500 < N_w r_w < 5000 \mu m \ cm^{-3}$ and $0.5 < N_I r_I < 500 \mu m \ cm^{-3}$, respectively, and the vertical velocity varied in the range $-5 < u_z < 5 m/s$. The initial radius of droplets varied from 2$\mu m$ to 10$\mu m$. The diagrams shown in Fig.1 suggest that Eqs. 5, 8 and 9 reproduce $S_w$, $q_i$ and $q_w$ calculated from the parcel model with a good accuracy. These results create a basis for the use of Eqs.8 and 9 in the following analysis.

It was found that at the final stage of glaciation the deviation of $S_{gsw}$ from $S_w$ increases when the droplets begin to rapidly reduce their sizes during evaporation. This deviation occurs because the limiting condition for the quasi-steady approximation (Korolev and Mazin 2003).

\[ \frac{2A_w a_0 |u_z|}{b_w^2 r_w^4 N_w^2} << 1 \] (12)

is not satisfied, when the droplets become too small. The limiting condition Eq.12 is also relevant to the initial stage of activation of droplets in the pre-existing ice cloud, when the droplets are yet not big enough. Modelling points, where Eq.12 was not satisfied, were indicated by red triangles in Fig.1. The sizes of the droplets corresponding to the red triangles vary from 0.5 to 3$\mu m$, depending on $N_w$ and $u_z$. Other limiting conditions applied to the rate of changes of $P$, $T$, $u_z$ are discussed in detail in (Korolev and Mazin, 2003). It is shown that usually the conditions for $P$, $T$, $u_z$ are satisfied in mixed-phase clouds in the troposphere.

Equations 8 and 9 have a simple form and, presumably, they may be implemented in cloud models or used for parameterization in large scale models, with relative ease.

Figure 1. Comparisons of (a) $S_w$, (b) $q_w$ and (c) $q_i$ derived from Eqs. 5, 8 and 9, respectively, with that calculated from a parcel model of a vertically moving adiabatic mixed phase cloud. Modelling was conducted for $500 < N_w r_w < 5000 \mu m \ cm^{-3}$; $0.5 < N_I r_I < 500 \mu m \ cm^{-3}$; $-5 < u_z < 5 m/s$; $-40 ^\circ C < T < -5 ^\circ C$; $500 mb < P < 1000 mb$. Blue circles indicate points satisfying Eq.12. Red triangles indicate points outside the envelope of the limiting conditions for the quasi-steady approximation described by Eq. 12.
3. Effect of thermodynamics on $\dot{q}_w$, $\dot{q}_i$ and $\dot{q}_v$

In this section we consider the effect of $u_z$, $T$ and $P$ on the rates of changes of ice, liquid and water vapour mass under conditions typical for mixed-phase clouds.

Figure 2 shows the growth rate of ice and liquid in mixed phase clouds versus $u_z$ calculated from Eqs. 8 and 9 for $N_{w}\bar{r}_w = 1000\mu m \ cm^{-3}$ and $N_{i}\bar{r}_i = 10\mu m \ cm^{-3}$ at three different temperatures; -5C, -15C and -25C. As follows from Eqs. 8 and 9 $\dot{q}_w$ and $\dot{q}_i$ are linearly related to $u_z$. Figure 2 indicates that $\dot{q}_w$ is significantly more sensitive to changes of $u_z$ as compared to that of $\dot{q}_i$. As seen from Fig.2 $\dot{q}_i$ stays nearly constant, whereas $\dot{q}_w$ changes signs within the range of $u_z$ considered in Fig.2. Formally, the lower sensitivity of $\dot{q}_i$ versus $u_z$ as compared to $\dot{q}_w$ is explained by difference in the coefficients $B_w N_{w}\bar{r}_w$ and $B_i N_{i}\bar{r}_i$ in front of $u_z$ in Eqs. 8 and 9, respectively. The difference in coefficients is mainly defined by the difference in values of $N_{w}\bar{r}_w$ and $N_{i}\bar{r}_i$. Typically, in mixed-phase clouds $N_{w}\bar{r}_w >> N_{i}\bar{r}_i$ and $B_w << B_i$.

The physical explanation of the difference in sensitivity of $\dot{q}_i$ and $\dot{q}_w$ to $u_z$ follows from the fact that the humidity in mixed phase clouds is close to saturation with respect to water (Korolev and Mazin 2005, Korolev and Isaac 2006). Therefore, the fluctuations of water supersaturation in mixed-phase clouds generated by fluctuations of vertical velocity with amplitude few meters per second have minor effect on supersaturation over ice, and for practical purposes $S_i$ may be considered approximately constant, i.e. $\Delta S_i << S_i$.

Figure 3. (a) Dependence of $\dot{q}_w$ versus vertical velocity $u_z$ for three temperatures. (b) Zoomed area in the vicinity of the point with $u_z=0$ and $\dot{q}_w=0$. The $x$- and $y$-axes in (b) are the same as in (a). Calculations were done for three different temperatures -5C, -15C and -25C, integral radii $N_{w}\bar{r}_w = 1000\mu m \ cm^{-3}$, $N_{i}\bar{r}_i = 10\mu m \ cm^{-3}$, and pressure $P=680mb$.

Figure 3 shows the dependence of the rate of the water vapour mass $\dot{q}_v$ versus $u_z$ for $N_{w}\bar{r}_w = 1000\mu m \ cm^{-3}$, $N_{i}\bar{r}_i = 10\mu m \ cm^{-3}$ calculated for three temperatures. Equation 11 indicates that $\dot{q}_v$ is also a linear function of $u_z$. In Fig.3a all $\dot{q}_v$ curves calculated for different $T$ appear to intersect at the same point (0,0). However, a closer look at the region of intersection (Fig.3b) indicates that the $\dot{q}_v$ curves intersect each other at different points.

The rates $\dot{q}_w$, $\dot{q}_i$ and $\dot{q}_v$ depend on $T$ and $P$ through the coefficients $a_0$, $b_w$, $B_w$, $b_i$, $B_i$ and $\xi$. Figure 4 shows $\dot{q}_w$, $\dot{q}_i$ and $\dot{q}_v$ versus $T$ for three different pressures $P=900mb$, 500mb and 300mb and two vertical velocities of $u_z = -1m/s$ and 1m/s.
Figure 4a shows that the growth rate of ice has a maximum ($\dot{q}_{i,\text{max}}$) at temperatures below -12°C. The temperature corresponding to $\dot{q}_{i,\text{max}}$ is decreasing with the decrease of $P$, and the amplitude of $\dot{q}_{i,\text{max}}$ is increasing with the decrease of $P$. It should be noted, that in mixed phase clouds the temperature corresponding to $\dot{q}_{i,\text{max}}$ is not equal to -12°C, where the maximum difference is between saturation pressures over ice and liquid, but it may change in the range $-40<T<-12$ depending on $P$. For pressure $200<P<1000$mb the temperature for $\dot{q}_{i,\text{max}}$ varies in the range $-19<T<-12$C. Cases with $P<200$mb resulting in $T<-19$C for $\dot{q}_{i,\text{max}}$ have no significance for the mixed phase clouds formed in the troposphere. As seen from Fig.4a, the ice growth rates $\dot{q}_i$ at $u_z=-1$ m/s and $u_z=1$ m/s practically coincide. In other words, $u_z$ has minor effect on $\dot{q}_i$, which is consistent with the above discussion.

Figure 4b shows the dependence of $\dot{q}_w$ versus $T$ and $P$. Contrary to $\dot{q}_i$, the growth rate of liquid $\dot{q}_w$ has minimum $\dot{q}_{w,\text{min}}$, which, depending on $P$ and $u_z$, may be located anywhere in the range $-40<T<0$C. Comparisons of Figs 4a and 4b indicate that the temperatures corresponding to the minimum of the growth rate of the liquid phase ($\dot{q}_{w,\text{min}}$) and the maximum growth rate of the ice mass ($\dot{q}_{i,\text{max}}$) are different.

Figure 4c shows that $\dot{q}_v$ is monotonically increasing for $u_z=-1$ m/s and decreasing for $u_z=1$ m/s. The absolute value $|\dot{q}_v|$ decreases with increase of $P$. Calculations show that for the considered values of integral radii, $\dot{q}_v$ becomes non-monotonic for low vertical velocities $|u_z|<0.1$ m/s.

In some papers it is speculated that the maximum rate of the phase transformation of liquid-to-ice is expected to occur at -12°C, where the maximum difference between saturation water vapour pressure between ice and liquid is observed. In fact, the diagrams in Fig. 4 suggest that the extrema of $\dot{q}_w$ and $\dot{q}_i$ varies in a wide range of temperatures depending on $P$, $N_w\bar{r}_w$, $N_i\bar{r}_i$ and $u_z$, and that under the same conditions the temperatures corresponding to $\dot{q}_{w,\text{min}}$ and $\dot{q}_{i,\text{max}}$ are different.

**Figure 4.** Dependence of the growth rate of (a) ice water mass $\dot{q}_w$, (b) liquid mass $\dot{q}_i$ and (c) water vapour mass $\dot{q}_v$ versus temperature $T$, for two vertical velocities 1m/s (solid line) and -1m/s (dashed line) and three different pressures 900mb, 500mb and 300mb. Calculations were done for integral radii $N_w\bar{r}_w=1000\mu$m cm$^{-3}$, $N_i\bar{r}_i=10\mu$m cm$^{-3}$.
4. Points of the phase equilibrium in mixed phase clouds

The interaction between solid, liquid and gaseous phases of water in mixed phase clouds is a complex process. The rate and direction of the partitioning of the water between three phases is defined by local thermodynamical \((T, P, u_z)\) and microphysical \((N_{w,fr}, N_{i,fr})\) characteristics of the cloud. In this section, we consider thermodynamical conditions required for the equilibrium of each of the three phases, i.e. when (a) \(\dot{q}_w=0\) liquid phase is in equilibrium; (b) \(\dot{q}_i=0\) ice phase is in equilibrium; (c) \(\dot{q}_v=0\) water vapour is in equilibrium.

4.1 Case \(\dot{q}_w=0\)

As follows from Eq.8 the equilibrium of liquid phase \(\dot{q}_w=0\) occurs, when the vertical velocity \(u_z\) becomes equal to a threshold velocity,

\[
u^*_z = \frac{E_w - E_i}{E_i} \eta N_{i,fr}\]

(13)

here, \(\eta = \alpha_2 B_{i0}/a_0\). A similar result was obtained in Korolev and Mazin (2003). The threshold velocity \(u^*_z\) separates the regimes of growth and evaporation of liquid droplets in mixed phase clouds, i.e. for \(u_z > u^*_z\) liquid droplets grow in the presence of ice, whereas for \(u_z < u^*_z\) droplets evaporate. As seen from Eq.13 \(u^*_z\) is always positive. Figure 5a shows dependence of \(u^*_z\) versus \(T\) for different \(N_{i,fr}\) and \(P\). As follows from Fig.5a \(u^*_z\) increases with the decrease of \(T\). For typical values of \(N_{i,fr}\) the magnitude of the threshold velocity \(u^*_z\) changes in the range \(10^{-2}\) m/s to \(10^0\) m/s in the temperature interval from -5°C to -30°C (Fig.5a). Such vertical velocities are common in the atmosphere, and they can be generated in clouds by turbulence or convection.

For a general case, including non-adiabatic conditions, the equilibrium of liquid phase in mixed phase clouds occurs when \(e=E_w\), which is equivalent to \(S_w=0\).

4.2 Case \(\dot{q}_i=0\)

Equation 9 yields a vertical velocity \(u^*_z\) required for equilibrium of ice phase in a mixed phase cloud

\[
u^*_z = \frac{E_i - E_w}{E_w} \chi N_{w,fr}\]

(14)
here, $\chi = a_i B_w / a_0$. Similar to $u_z^*$, the velocity $u_z^0$ separates regimes of growth and evaporation of ice particles in mixed phase clouds, i.e. for $u_z < u_z^0$ ice particles sublimate in the presence of liquid droplets, whereas for $u_z > u_z^0$ ice particles grow. As follows from Eq.14 $u_z^0$ is always negative. Figure 5b shows the dependence of $u_z^0$ versus $T$ for different $N_i \bar{r}_i$ and $P$. As seen from Fig.5b, $u_z^0$ increases with the increase of $T$. For typical values of $N_i \bar{r}_i$, the magnitude of the velocity $u_z^0$ changes in the range from $-10^5 \text{m/s}$ to $-10^3 \text{m/s}$ in the temperature range $-5 \text{C}$ to $-30 \text{C}$ (Fig.5b). Vertical velocities $-10 < u_z < -1 \text{m/s}$ may occur in compensating downdrafts in convective clouds. Downdrafts with a higher velocity are unlikely to be formed in the atmosphere. The diagram in Fig.5b suggests that at temperatures close to $0 \text{C}$, the absolute values of the vertical downdrafts required for reaching ice equilibrium do not exceed $-1 \text{m/s}$, and they can be generated by turbulence.

Generally, the equilibrium of ice phase requires $e = E_i$, which is equivalent to the condition $S_w = \xi - 1$ in terms of the supersaturation over water, or $S_i = 0$ in terms of the supersaturation over ice. It should be noted, that for a non-adiabatic case the local equilibrium of ice phase in a mixed phase cloud can be reached during mixing with dry out-of-cloud air.

### 4.3 Case $\dot{q}_v = 0$

Equation Eq.11 provides a velocity required for the water vapour equilibrium

$$u_z^+ = \frac{\xi - 1}{a_0 \xi (B_w N_i \bar{r}_i + B_i N_i \bar{r}_i)} (B_w b_i - b_w B_i) N_i \bar{r}_i N_i \bar{r}_i$$

For $u_z < u_z^+$, the mass of the water vapour is increasing, i.e. $\dot{q}_v > 0$, whereas for $u_z > u_z^+$ the mass of the water vapour is decreasing, i.e. $\dot{q}_v < 0$. As follows from Eq.15 $u_z^+$ is always positive. Figure 5c shows the dependence of $u_z^+$ versus $T$ for different $N_i \bar{r}_i$ calculated for $N_w \bar{r}_w = 1000 \mu \text{m cm}^{-3}$. As seen from Fig.5c in mixed phase clouds $u_z^+$ is close to zero, and for typical $N_i \bar{r}_i$ and $N_w \bar{r}_w$ it is usually of the order of millimetres and centimetres per second.

In a general case, including non-adiabatic conditions the equilibrium of water vapor will be reached when $e = E_v$, where $E_v$ is the equilibrium vapor pressure,

$$E_v = \left(1 + S_w^{(v)}\right) E_w^{(v)}$$

Here, $S_w^{(v)}$ is the supersaturation yielding the water vapour equilibrium. In order to find $S_w^{(v)}$ substitute $q_v = 0$ in Eq.10, which yields

$$q_w = -\dot{q}_i.$$  Then, combining Eqs.1, 4, 5 and $q_w = -\dot{q}_i$ we obtain

$$S_w^{(v)} = \frac{B_i N_i \bar{r}_i}{B_w N_i \bar{r}_w + B_i N_i \bar{r}_i}$$

It should be noted that the equilibrium of water vapour in mixed phase clouds has a dynamic nature, i.e. water vapour released by evaporating droplets is depleted by growing ice particles.

### 5. Different regimes of the phase transformation in mixed phase clouds

Three points of the phase equilibrium $\dot{q}_w = 0, \dot{q}_i = 0, \dot{q}_i = 0$ and their associated vertical velocities $u_z^+, u_z^+, u_z^0$, discussed in the previous section, play a fundamental role in the understanding of the phase transformation in mixed phase clouds. The analysis of Eqs. 13, 14, and 15 yields that the inequality

$$u_z^0 < u_z^+ < u_z^+$$

is always satisfied for any $N_i \bar{r}_i$ and $N_w \bar{r}_w$ (Korolev 2008). The inequality in Eq.26 enables separating the phase transformation in mixed clouds into four regimes:

1. if $u_z < u_z^+$, then $\dot{q}_i > 0$, $\dot{q}_i < 0$ and $\dot{q}_w < 0$. In this case, both ice particles and droplets evaporate, whereas the mass of the vapour increases. In terms of the vapour pressure this case corresponds to the condition, when $e < E_i$.

2. if $u_z^+ < u_z < u_z^0$, then $\dot{q}_v > 0$, $\dot{q}_i = 0$ and $\dot{q}_w < 0$. Under these conditions ice particles grow, droplets evaporate, and the water vapour
mixing ratio increases. The water vapour pressure in this case changes in the range \( E_i < e < E_v \).

(3) if \( u_z^* < u_z < u_z^* \), then \( q_v < 0, \ q_i > 0 \) and \( q_w < 0 \). In this case, ice particles grow, droplets evaporate, and the water vapour mass decreases. This case corresponds to the water vapour pressure \( E_i < e < E_v \).

(4) if \( u_z > u_z^* \), then \( q_v < 0, \ q_i > 0 \) and \( q_w > 0 \). In this case, both ice particles and liquid droplets grow, and the water vapour mass decreases. Under this condition the water vapour pressure will be \( e > E_w \).

A conceptual diagram in Fig.6 illustrates the four regimes of phase transformation in mixed-phase clouds, based on the analogy between three fluid reservoirs ('liquid', 'vapour' and 'ice') connected with pipes and placed on different levels. The levels of the reservoirs is analogous to the water vapour pressure. Since \( E_w \) and \( E_i \) are constant, then depending on the vapour pressure \( e \), the flow of the water vapour may be directed either from both liquid and ice to the vapour (Fig.6d), or from the vapour to both the liquid and the ice (Fig.6a), or from the liquid to the ice through the vapour (Fig.6b,c).

![Figure 6](image)

Figure 6. Conceptual diagram of four different scenarios of phase transformation in mixed-phase clouds: (a) \( u_z > u_z^* \); (b) \( u_z^* < u_z < u_z^* \); (c) \( u_z^* < u_z < u_z^* \); (d) \( u_z < u_z^* \). The arrows indicate the direction of the mass transfer. The thickness of the arrows indicates conditional rates of the mass transfer.

6. WBF process

The WBF process is defined as the process, when “...ice crystals would gain mass by vapour deposition at the expense of the liquid drops that would lose their mass by evaporation” (Glossary of Meteorology, 2000). The thermodynamical conditions which are a result of the aforementioned condition occur when “the vapour tension will adjust itself to a value in between the saturation values over ice and over water” (Wegener, 1911, p.81). Based on the above statements it can be concluded that the WBF process is determined by a set of conditions: \( q_w < 0, \ q_i > 0 \) and \( E_i < e < E_w \). Thus, of the above four scenarios described in section 5, only cases (2) and (3) suit the definition of the WBF process, i.e. when ice particles are growing at the expense of evaporating liquid droplets. These cases correspond to the velocity range \( u_z^* < u_z < u_z^* \).

For the vertical velocities \( u_z > u_z^* \), both ice and liquid particles grow simultaneously. Under these conditions the liquid droplets compete with the ice particles for the water vapour. It can be shown, that for \( u_z > u_z^* \) liquid droplets slow down the rate of growth of ice particles \( q_i \) (Korolev 2008). Figure 7 shows the rate of changes of the mixing ratio of ice \( q_i \) versus \( N_{w, \Gamma} \) for different \( u_z \). As seen from Fig. 7, for \( u_z > u_z^* \) the increase of \( N_{w, \Gamma} \) slows down the ice growth. In other words, when \( u_z > u_z^* \), ice crystals grow faster without droplets as compared to when the droplets are present. This type of behaviour of liquid droplets within the presence of ice particles is different from that described by the WBF process, which is when liquid droplets enhance the growth of ice particles by providing them with water vapour through their evaporation (Wegener, 1911, Bergeron 1935, Glossary of meteorology, 2000).

For a case when \( u_z > u_z^* \), both ice particles and liquid droplets simultaneously evaporate, which, again, does not match the definition of the WBF process, since ice particle evaporate in presence of liquid droplets (Glossary of Meteorology 2000). Figure 7 also shows that liquid droplets slow down the sublimation rate...
of ice particles when \( u_z < u_z^* \), i.e. when both droplets and ice particle evaporate.

**Figure 7.** The rates of changes of the mixing ratio of ice \( \dot{q}_i \) versus integral radius of cloud droplets \( N_w \bar{r}_w \) for different \( u_z \). As seen, for \( u_z > u_z^* \) the increase of \( N_w \bar{r}_w \) slows down the ice growth (the WBF process is declined), whereas for \( u_z^* < u_z < u_z^* \) the increase of \( N_w \bar{r}_w \) results in the increase of growth of ice. Grey colour shows the area where the WBF process is enabled. For \( u_z < u_z^* \) evaporating droplets slow down the rate of ice sublimation. Calculations were done for \( N_{i,i} \bar{r}_i = 10 \mu m cm^{-3} \), \( T = -10 C \); \( P = 890 mb \).

Earlier Reisin et al. (1996) came to a conclusion, based on the analysis of results of numerical modelling of convective clouds, that the WBF process “did not play a significant role in the rain formation process” and that in strong updrafts “ice particles grew by deposition, but did not cause the evaporation of the drops”. A similar statement can be found in Pruppacher and Klett (1997, p.549). Although it was understood that the WBF process may play a limited role in mixed phase clouds, at that point it was not clear as to what conditions activated the process. In the present section we demarcate three distinctly different regimes of the behaviour of the condensed water in mixed phase clouds and find the conditions for each of them.

For adiabatic parcels, the regimes of growth and evaporation of the droplets and ice particles can be well depicted by the ratio of the growth rates of liquid and ice \( \dot{q}_w / \dot{q}_i \). Dividing Eq. 8 by Eq. 9 yields,

\[
\frac{\dot{q}_w}{\dot{q}_i} = \frac{\left( a_0 u_z - \frac{1}{\zeta} b_i N_w \bar{r}_w \right) B_{w} N_i \bar{F} \cdot N_w \bar{r}_w}{\left( a_0 u_z - \frac{1}{\zeta} b_i N_w \bar{r}_w \right) B_{i} N_i \bar{F}}
\]

(19)

Figure 8 shows the dependence of the ratio \( \dot{q}_w / \dot{q}_i \) versus the vertical velocity \( u_z \) for \( N_w \bar{r}_w = 500 \mu m cm^{-3} \), \( N_{i,i} \bar{r}_i = 10 \mu m cm^{-3} \) at \( T = -5C \). Grey areas in Fig.8 indicated by numbers 1, 2, 3 and 4 correspond to four different regimes as described in section 5. The vertical dashed lines in Fig.8 show the threshold velocities \( u_z^* \) (Eq.11), \( u_z^o \) (Eq.12) and \( u_z^+ \) (Eq.15) separating the four regimes of mixed phase evolution.

For the vertical velocity \( u_z^* \), the growth rate of liquid water \( \dot{q}_w = 0 \) and, therefore, \( \dot{q}_w / \dot{q}_i = 0 \). For the velocity \( u_z^o \), the growth rate of ice \( \dot{q}_i = 0 \) and therefore, \( \dot{q}_w / \dot{q}_i \rightarrow -\infty \). The WBF process is enabled only in the range of vertical velocities \( u_z^* > u_z > u_z^o \) (areas 2 and 3 in Fig.8).

**Figure 8.** Ratio \( \dot{q}_w / \dot{q}_i \) versus vertical velocity \( u_z \) in a mixed phase cloud. Grey areas and numbers indicate areas where: (1) \( \dot{q}_i < 0 \) and \( \dot{q}_w < 0 \), both droplets and ice particles evaporate; (2 and 3) \( \dot{q}_i > 0 \) and \( \dot{q}_w < 0 \), liquid droplets evaporate, whereas ice particle grow (this condition corresponds to the WBF process); (4) \( \dot{q}_i > 0 \) and \( \dot{q}_w > 0 \), both droplets and ice particles grow. \( T = -5C \); \( P = 680 mb \); \( N_w \bar{r}_w = 300 \mu m cm^{-3} \); \( N_{i,i} \bar{r}_i = 10 \mu m cm^{-3} \).
During the WBF process the ice particles are growing not only due to the evaporating liquid droplets. The mass of the water vapour deposited on the ice crystals may be partitioned between evaporated and pre-existing liquid droplets within the cloud parcel water vapour. This growth regime corresponds to the condition $-1 < q_w / q_i < 0$ (area 3 in Fig.8). In other words, in area 3 in Fig.8 when $u^*_z < u_z < u^*_i$, ice particles consume more water vapour than was evaporated by the liquid droplets. When $u_z$ is approaching to $u^*_i$, the liquid droplets slow their evaporation down, whereas ice particles increase the growth rate. At $u_z = u^*_i$, liquid droplets reach equilibrium with water vapour and cease growing.

In the case of $-\infty < q_w / q_i < -1$ (area 2 in Fig.8) only a fraction of water vapour evaporated by liquid droplets will be depleted by growing ice crystals, and another fraction will stay in the gaseous phase. In area 2 in Fig.8, when $u^*_z < u_z < u^*_i$, ice particles uptake less water vapour than that evaporated by the liquid droplets. When $u_z$ is approaching to $u^*_z$, the liquid droplets evaporate faster, whereas ice particle slow down their growth, and at $u_z = u^*_z$ ice particles stay in equilibrium with the water vapour and stop growing.

The genuine WBF process, when all water evaporated by the droplets is equal to that consumed by the ice particles, occurs when $q_w / q_i = -1$. Following Eq.10, the condition $q_w / q_i = -1$ is equivalent to $q_i = 0$, which for an adiabatic case is reached when $u_z = u^*_z$ (Eq.15).

In order to characterize the effect of evaporating droplets on the growth of ice particles we introduce the efficiency of the WBF process ($\omega$): The WBF efficiency is defined as the fraction of evaporated liquid water converted into ice ($\omega = q_i / |q_w|$) for $u^*_z < u_z < u^*_i$. For the velocity range $u^*_z < u_z < u^*_i$, the growth rate of ice $q_i$ becomes larger than that of liquid $|q_w|$, since ice grows both due to evaporating droplets and pre-existing water vapour. Therefore, for $u^*_z < u_z < u^*_i$ the WBF efficiency is defined as the fraction of evaporated liquid to the total water vapour converted into ice, i.e. $\omega = |q_w| / q_i$. Figure 9 shows the dependence of the WBF efficiency versus $u_z$. As seen from Fig.9, $\omega=0$, for $u_z = u^*_z$ and $u_z = u^*_i$, and it reaches maximum ($\omega=1$), when $u_z = u^*_z$.

![Figure 9. Efficiency of the WBF process versus $u_z$. The conditions are the same as in Fig.7: $T=-5^\circ C$; $P=680mb$; $N_{w} = 300 \mu m cm^{-3}$; $N_{i} = 10 \mu m cm^{-3}$](image)

![Figure 10. Dependence of the ratio $q_w / q_i$ versus vertical velocity $u_z$ for three different temperatures. Calculations were done for integral radii $N_{w} = 1000 \mu m cm^{-3}$, $N_{i} = 10 \mu m cm^{-3}$, and pressure $P=680mb$.](image)
The ratio $\dot{q}_w / \dot{q}_i$ is temperature sensitive. Figure 10 shows the behaviour of $\dot{q}_w / \dot{q}_i$ for the different temperatures in the vicinity of $u=0$. As seen from Fig.10 the dependence of $\dot{q}_w / \dot{q}_i$ on $u$ weakening with the decrease of temperature.

At $u=0$ the ratio $\dot{q}_w / \dot{q}_i$ becomes independent of integral radii $N_w \bar{r}_w$ and $N_i \bar{r}_i$, and it is equal to

$$\left( \frac{\dot{q}_w}{\dot{q}_i} \right)_{u=0} = - \frac{a_2}{a_1} \quad (20)$$

The ratio $a_2/a_1$ is a weak function of $T$ and $P$, and it changes in the range of $1 < a_2/a_1 < 1.08$, for the changes temperature and pressure in the ranges from $-40<T<0^\circ C$ to $300<P<1000mb$, respectively. Thus, for practical purposes, it can be considered, with a high degree of accuracy, that $\dot{q}_w / \dot{q}_i \approx -1$ at $u=0$ regardless $N_w \bar{r}_w$, $N_i \bar{r}_i$, $T$ and $P$. This finding results in the conclusion that the WBF process has a maximum efficiency $(\alpha=1)$ at $u \approx 0$ and to a first approximation it is not dependant on the microphysics of mixed phase clouds, temperature and pressure.

7. Limitations and assumptions

The consideration of an idealized mixed phase cloud as discussed at the beginning of section 2 resulted in the neglecting of some physical processes inherent in real clouds such as: (1) sedimentation; (2) aggregation of ice crystals; (3) riming; (4) nucleation of ice particles; (5) activation and evaporation of liquid droplets; (6) coalescence of droplets; (7) entrainment and mixing; (8) radiation effects.

The above processes affect the rates of phase transformation through the changes of $N_i \bar{r}_i$ (processes #1-4), $N_w \bar{r}_w$ (processes #5, 6), and the humidity and temperature (processes #7, 8). In the present work the instant rates of the phase transformation were considered for the diffusional stage. The characteristic timescale of the diffusional processes is determined by the time of phase relaxation $\tau_p$ (Eq.7). Typically, in mixed phase clouds $\tau_p$ is of the order of few seconds (Korolev and Mazin 2003). The magnitude of the characteristic timescales of processes related to aggregation, riming, coagulation, sedimentation radiative heating or cooling varies from minutes to hours (Prupacher and Klett, 1997) and it is much larger than $\tau_p$. Due to a naturally low concentration of ice nuclei, secondary ice nucleation in already pre-existing ice or mixed phase clouds is considered to be a relatively slow process, which is not expected to change $N_i \bar{r}_i$ within a few $\tau_p$.

Therefore, it is expected that the effect of the processes #1-4, 6, 8 on the instant rates of $\dot{q}_w$, $\dot{q}_i$, and $\dot{q}_v$ will be negligible.

There are two main issues to consider: activation and evaporation of liquid droplets and entrainment and mixing. The entrainment and mixing with the dry out-of-cloud air changes the local relative humidity and temperature. Entrainment and mixing may result in a rapid decrease of the relative humidity followed by the evaporation of cloud particles to compensate for the deficit in humidity. This is an essentially non-equilibrium process, which cannot be described through the quasi-steady approximation. Since at timescales $t>>\tau_p$ the relative humidity in a cloud parcel relaxes to its quasi-steady value, the mixing is expected to have maximum effect on the steady state $\dot{q}_w$, $\dot{q}_i$, and $\dot{q}_v$ at $t<\tau_p$. Entrainment and mixing is a most common phenomenon in the vicinity of the cloud interfaces, and its effect decreases when moving away from the cloud boundaries deep into the cloud. Therefore, it is expected that the developed theoretical framework may not be applicable to the cloud regions in the vicinity of cloud boundaries with intensive entrainment and mixing.

The activation and evaporation of droplets imposes another limitation on the use of the quasi-steady approximation. During the initial stage of activation or the final stage of evaporation of droplets their sizes become too small, and that they do not satisfy the condition Eq.12 imposing the limitations on the use of the quasi-steady approximation. The activation of the droplets may occur in pre-existing ice clouds when updrafts $u_z > u_z^*$. The evaporation of droplets in a mixed phase cloud may happen during the process of cloud glaciation during ascent or descent, when $u_z < u_z^*$. The examples
of the cases which do not satisfy Eq.12 are
shown in Fig.3 with red triangles.
It should be noted that Eq.12 was derived in
assumption that \( \frac{N_w}{N_i} \gg \frac{i}{r} \). This condition is
typically satisfied in mixed phase clouds. For ice
clouds, a limiting condition can be obtained
from Eq.12 by simply replacing subscript “w” to
“i”.
Despite the limitations discussed above, the
quasi-steady approximation provides a relatively
accurate estimation of the instant rates of the
phase transformation (Fig.1). A more accurate
consideration of the phase transformation can be
achieved by utilizing the complete set of
differential equations describing all processes
mentioned at the beginning of this section. Such
elaboration may provide more accurate estimations of the threshold velocities \( u^* \), \( u^+ \),
\( u^- \) and the rates \( \dot{q}_i \), \( \dot{q}_e \) and \( \dot{q}_w \).

8. Conclusions
One of the goals of this work is to
demonstrate a complexity of interaction of liquid,
ice and gaseous phases in mixed phase clouds on a diffusional level. One of the
important outcomes of this work is that the interaction between three phases is not limited just by the WBF process, describing a one directional phase transition. In fact, mixed phase has several points of equilibrium and the phase transformation has different regimes and the WBF process is one of them. The direction and the rate of the phase transformation are tightly related to the local thermodynamical and microphysical properties of a mixed phase cloud.

It has been shown that for typical integral radii \( N_{i,f}, N_{w,f} \) the vertical velocities
separating different regimes of the phase transformation can be generated by turbulent fluctuations. In other words, during turbulent fluctuations the direction of partitioning of water between ice, liquid, and vapour are constantly changing direction, and occasionally following the WBF process. During turbulent fluctuations cloud parcels are continuously passing through different points of equilibrium, therefore resulting in a continuous change of the rate and direction of partitioning of the mass between ice,
liquid and ice phases. Since the integral radii of ice particles and droplets are continuously changing due to the diffusional growth and/or evaporation of droplets and ice particles, the vertical velocities defining the equilibrium between phases are also changing with time.

The following results were obtained within
the frame of this study:
1. It is shown that mixed phase clouds have
three basic points of phase equilibrium for liquid, ice and vapour phases, which separate the phase transformation in mixed phase clouds into four different regimes.
2. For typical \( N_{w,f} \) and \( N_{i,f} \) the growth rate of ice particles is significantly less sensitive to the vertical velocities in mixed phase clouds in comparison to liquid droplets.
3. The temperature corresponding to the maximum rate of the growth of ice and evaporation of liquid changes depending on \( P_i, N_{w,f}, N_{i,f} \) and \( u_z \), and it varies within a wide range. This temperature is not necessarily equal to -12C, where the maximum difference between saturation water vapour pressure between ice and liquid is observed.
4. Maximum efficiency of the WBF process, i.e. when all water vapour evaporated by the liquid droplets is deposited on the ice crystals, occurs at \( u_z = u^+_i \). It is shown that in mixed phase clouds \( u^+_i \sim 0 \).

References
Appendix A

List of Symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Units</th>
</tr>
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<tbody>
<tr>
<td>$a_0$</td>
<td>$g \left( \frac{L_v R_a}{c_p R_a T} - 1 \right)$</td>
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<td>m$^2$ s$^{-1}$</td>
</tr>
<tr>
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<td>$a_i B_i$</td>
<td>m$^2$ s$^{-1}$</td>
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<td>$a_i B_{i0}$</td>
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<td>$a_i B_i^*$</td>
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</tr>
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<td>$c_p$</td>
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<td>Symbol</td>
<td>Definition</td>
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