## 0.1 Ice-supersaturation parameterization: A first-order approach

### 0.1.1 The basic framework

The standard cloud parameterization described above is based on the assumption that water is always at thermodynamic equilibrium between the vapor and liquid phases or between the vapour and solid (ice) phases. However, this assumption is not always met in the atmosphere, as some regions can be supersaturated with respect to ice.

The objective of the modified parameterization is to allow the model to realistically create and maintain such ice-supersaturated conditions. Given the coarse resolution of LMDZ (see Sect. **??**), we consider that a given gridbox can be partitioned between an ice-subsaturated region (a thermodynamic equilibrium state with $q < q_{sat}$), a clear-sky ISS region (a metastable thermodynamic state with $q > q_{sat}$), and a cloud fraction (in thermodynamic equilibrium state with total water $q > q_{sat}$). This new scheme is illustrated in Fig. 1b, where the three fractions are labelled *sub*, *ss* and *cld*, respectively. In the computing process, this new parameterization needs to track the cloud fraction from the previous time step so as to keep some memory as to whether condensation has occurred or not. The rationale for such a scheme is that an ISS fraction can be converted into a cloudy fraction but the opposite is not true as the cloud can only disappear through sublimation in subsaturated conditions or sedimentation of the ice crystals. The cloud cover computed at the previous time step, is used as semi-prognostic variable, by considering it as an additional tracer, advected by the dynamical core, along with the specific contents of vapor, liquid and ice. Hereafter, the advected cloud cover and the new cloud cover for the current time step are referred to as $\alpha_{cld,adv}$ and $\alpha_{cld}$, respectively.

We further assume that cirrus clouds form only by homogeneous nucleation. Heterogeneous nucleation is an important process in cirrus cloud formation, but remains a less well-understood physical process [**?**], and accounting for it would require representing INPs in the model, which is beyond the scope of this study but will be the subject of a new study. The tuning method applied in this study could contribute to compensate to some extent for the absence of heterogeneous nucleation by making homogeneous nucleation more efficient.

In the new scheme, ice clouds can form through homogeneous freezing when ice supersaturation exceeds a threshold $\gamma_{ss}\, q_{sat}$ that is approximated as a function of temperature as specified by (koop2000water) and (ren2005cirrus). The threshold of ice supersaturation ratio, $\gamma_{ss}$, is expressed as:

$$\gamma_{ss} = \gamma_0 - T/T_\gamma \tag{1}$$

where $T$ is the grid-box temperature (in K), assumed to be homogeneously distributed in the gridbox. The values of $\gamma_0$ and $T_\gamma$ are initially fixed to 2.349 and 259 K, respectively (see Table 1 and (ren2005cirrus)). There are also considered here as tuning parameters because i) the data used to determine Equation 1 may be subject to uncertainty and ii) we favor ”effective” values of the parameters so that LMDZ simulates realistic amounts of clouds and supersaturation.

We defined the permitted intervals for the parameters $\gamma_0$ and $T_\gamma$ by varying the intercept and the slope of the nucleation line within the dispersion range of the data points presented in Figure 1 in (koop2000water). The linear model of Equation 1 is constrained to remain entirely within the limits defined by the dispersion of the observed supersaturation thresholds between 170 K and 240 K, in order to cover the entire range of experimental uncertainties, including variations associated with different crystal sizes. Consequently, the resulting intervals for $\gamma_0$ and $T_\gamma$ represent physically plausible nucleation regimes that encompass the diversity of crystal growth behaviors documented in (koop2000water).

We characterize the three fractions of the gridbox through their respective cover $\alpha_k$ (dimensionless) and their gridbox-average total water $q_k$ ($\mathrm{kg\,kg^{-1}}$). Thus the following conservation equations apply:

$$\alpha_{sub} + \alpha_{ss} + \alpha_{cld} = 1 \tag{2}$$

and

$$q_{sub} + q_{ss} + q_{cld} = q_t \tag{3}$$

where $q_{cld}$ represents the total water content of the cloud, including both water vapor and ice. This parameterization in LMDZ is only applied for temperatures below −30°C. For temperatures warmer than −30°C, the mixed-phase cloud parameterization takes over, with a linear transition applied to avoid discontinuities in $\alpha_{cld}$ and $q_{cld}$.

### 0.1.2 Subgrid water vapor distribution

~~This new parameterisation of ISSRs and cirrus clouds is based on a decomposition of~~ The new cloud scheme relies on the same analytical PDF $P(q)$ ~~, which is used in the former parameterisation (see Sect. ??),~~ for the subgrid scale total water as the former one.

In order to account for the possible co-existence within the same grid cell, of clouds, sub-saturated and supersaturated regions however, $P(q)$ is decomposed into three sub-PDFs ~~$P_{sub}$, $P_{ss}$ and $P_{cld}$~~ as (Fig. 1)~~.~~ :

$$P(q) = \alpha_{sub}\, P_{sub}(q) + \alpha_{ss}\, P_{ss}(q) + \alpha_{cld}\, P_{cld}(q), \tag{4}$$

each three PDF $P_{sub}$, $P_{ss}$ and $P_{cld}$ ~~describe the total water content in the *sub*, *ss*, *cld* regions, respectively (Fig. 1b). The decomposition of $P(q)$ is formalized as follows:~~

$$P(q) = \alpha_{sub}\, P_{sub}(q) + \alpha_{ss}\, P_{ss}(q) + \alpha_{cld}\, P_{cld}(q),$$

being normalized to 1.

The scheme introduces two new state variables: the cloud fraction and the humidity at saturation. The values of this state variables at the beginning of the cloud scheme, after large scale advection, are noted $\alpha_{cld,adv}$ and $q_{vc}$.

For $q \leq q_{vc}$ the air is unsaturated and with no clouds, so that

$$P_{sub}(q) = \begin{cases} P(q)/\alpha_{sub} & \text{if } q < q_{vc} \\ 0 & \text{otherwise} \end{cases} \tag{5}$$

For $q \geq q_{vc}$, cloudy and supersaturated air may co-exist. For the sake of simplicity, we assume that the PDF of the cloudy fraction is the total PDF multiplied by a constant factor $N$ so that:

$$P_{cld}(q) = \begin{cases} 0 & \text{if } q < q_{vc} \\ N\,P(q)/\alpha_{cld,adv} & \text{otherwise} \end{cases} \tag{6}$$

and

$$P_{ss}(q) = \begin{cases} 0 & \text{if } q < q_{vc} \\ (1-N)\,P(q)/\alpha_{ss} & \text{otherwise} \end{cases} \tag{7}$$

~~where $\alpha_{sub} = \int_0^{q_{vc}} P(q)\,dq$ and $\alpha_{ss} = 1 - \alpha_{sub} - \alpha_{cld,adv}$ at~~

At the beginning of the ~~physical timestep, $q_{vc}$ is the lower bound ($q_{sat}$) of $P_{cld}$ at the previous physical timestep, advected by the dynamical core. $N$ is the coefficient of proportionality between $P_{cld}$ and $P(q)$. It is computed as follows:~~

$$N = min\left(1, \alpha_{cld,adv} / \int_{q_{vc}}^{+\infty} P(q)\,dq\right)$$

cloud scheme, the fraction of sub-saturated air simply reads

$$\alpha_{sub} = \int_0^{q_{vc}} P(q)\,dq \tag{8}$$

and $\alpha_{ss} = 1 - \alpha_{sub} - \alpha_{cld,adv}$.

$N$ ~~takes~~ should take values ranging from 0 to 1. ~~$N \in ]0,1[$ indicate that the three fractions coexist within the gridbox.~~ When $\alpha_{cld,adv} \leq 1 - \alpha_{sub}$, $N$ is computed easily from the normalisations of the PDF:

$$N = \frac{\alpha_{cld,adv}}{1 - \alpha_{sub}} \tag{9}$$

If $N = 0$~~corresponds to situations in which~~, only the *sub* and *ss* regions coexist~~, while $N = 1$ corresponds to situations in which~~.

If $N = 1$, only the *ss* and *cld* regions coexist in the gridbox.

~~\includegraphics[width=\textwidth, trim=0cm 0cm 0cm 0cm, scale=0.8] {Fig1.pdf}}~~

~~Total water distribution for the standard cloud parameterization in LMDZ6A (a) and the new ice supersaturation parameterization (b). The vertical black lines in a) and b) separate the undersaturated and saturated regions, and correspond to $q_{sat}$ and $q^{vc}$, respectively. In b) the orange curve represents~~

~~the $N \cdot P(q) = \alpha_{cld,adv} P_{cld}(q)$ curve. The situation illustrated in (b) is not an equilibrium state, but can be interpreted as the state at the beginning of the timestep before cloud physics is applied.~~

~~The separation of $P(q)$ into distincts parts is a new feature of this parametrization compared to the one introduced in Sect. **??**. For $q > q_{vc}$, $P_{cld}$ and $P_{ss}$ are assumed to be proportional to $P(q)$, otherwise they are equal to 0 (see Eq. 5–7). $q_{vc}$ is used to diagnose the dissipation of clouds (see Sect. 0.1.3) under the assumption that a decrease in the saturation threshold from one physical timestep to the next indicates a shift toward subsaturated state.~~

~~The formation of new clouds at each timestep is diagnosed by integrating $P_{ss}$ between $\gamma_{ss} q_{sat}$ and $+\infty$. In the subsequent physical timestep, the previous formed cloud fraction is conserved through $\alpha_{cld,adv}$, and its water is equally redistributed in *cld* and *ss* regions. This introduces additional water in the *ss* region, leading to an overestimation of ISSRs. This overestimation is adressed by changing the proportionality coefficient~~ $N$ ~~to $N_1$ =min$(1, \Phi_N\, N)$ for $q > \gamma_{ss}\, q^{vc}$ and to $N_2$ (see Eq. 11) for $q^{vc} \leq q \leq \gamma_{ss}\, q^{vc}$, where $\Phi_N$ is a tuning parameter that can take values ranging from 1 to 2. This treatment is not physical, but the introduction of the parameter $\Phi_N$ provides flexibility to the parameterization, allowing it to fit observations through tuning.~~

$$\cancel{N_2 = \frac{\alpha_{\mathrm{cld}} - N_1 \int_{\gamma_{\mathrm{ss}}\, q_{\mathrm{vc}}}^{+\infty} P(q)\, dq}{\int_{q_{\mathrm{vc}}}^{\gamma_{\mathrm{ss}}\, q_{\mathrm{vc}}} P(q)\, dq}}$$

~~This treatment, which consists of decomposing $P(q)$ in three components, as described above, must not lead to a loss or creation of water in the gridbox ($q_t$ must be conserved). A loss of water conservation may occur when the advected cloud fraction is greater than the maximum cloud fraction that can be diagnosed from $P(q)$, particularly in cases where $N = 1$. Under these conditions, cloud fraction tracking is not performed, and no ISSR is diagnosed because the cloud scheme reverts to the former cloud scheme of LMDZ. More precisely, the cloud fraction formed must not exceed~~ is then set to 1. This arrizes either when $\alpha_{cld,adv} \geq 1 - \alpha_{sub}$ or when $\alpha_{cld,adv} \geq \alpha_{cld,max}$, where $\alpha_{cld,max}$ is a maximum cloud fraction ~~$\alpha_{cld,max}$, which~~ that corresponds to the cloud fraction that would form if we were in a context of adjustment to saturation (i.e., conditions of equation **??**). ~~When $\alpha_{cld,adv} < \alpha_{cld,max}$, cloud fraction is computed using the new scheme . However, when $\alpha_{cld,adv} > \alpha_{cld,max}$,~~ In this case, the cloud scheme reverts to the former cloud scheme of LMDZ and the memory of the ~~cloud properties is lost, and the former diagnostic scheme applies. Therefore,~~ clouds from previous time step is lost.

Note that in all cases, the cirrus cloud fractions are limited by the diagnosis of $P(q)$ in the former scheme, ~~and they~~ which were shown to be underestimated [**?**]. This comes from the high value of $\xi_{old}$ at cirrus cloud altitudes. To overcome this issue, we reduce the value of $\xi_{old}$ when a cirrus cloud was previously present in the gridbox~~. To do this, we assume~~ , assuming that the standard deviation of the total water distribution is inversely proportional to the cloud fraction in the gridbox~~, and modify the function $\xi_{old}$ to $\xi$ as defined in equation 10.~~ . The modified value reads

$$\xi(p) = \frac{\xi_{old}(p)}{1 + \beta \cdot \alpha_{cld,adv}} \tag{10}$$

This formulation is not directly supported by observations ~~, consequently, we made depending $\xi$ dependent on a tuning parameter and test its relevance. The function $\xi(p)$ for the new parameterization is then computed as follows:~~

$$\cancel{\xi(p) = \frac{\xi_{old}(p)}{1 + \beta \cdot \alpha_{cld,adv}}}$$

~~where~~ and $\beta$ is a tuning parameter ~~whose value is between 0 and 5. Case where~~ with lower bound $\beta = 0$ ~~corresponds to the condition in~~ (for which the old ~~parameterization takes over~~arameterization takes over). Values of a few units are typically found in the tuning exercise.

Finally, if $N \in ]0, 1[$, i.e. if $0 < \alpha_{cld,adv} < \alpha_{cld,max}$, the three fractions coexist within the gridbox. ~~Tests have shown that the maximum value of 5 is realistic, allowing for scaling that reduces the variance of $P(q)$ compared to the former parameterization. Once the three distributions $P_{sub}$, $P_{ss}$, $P_{cld}$, as well as the total water distribution $P(q)$ in the gridbox, are constructed from $q_t$, $\alpha_{cld,adv}$ and $q_{vc}$, the physical processes that modify these quantities are applied. The processes considered are~~ In this case, one has to account for the dissipation of ~~subsaturated~~ sub-saturated clouds, the formation of new clouds, the adjustment of in-cloud water vapor to saturation, and the mixing of clouds with their environment.

### 0.1.3 Sublimation, condensation, and phase ~~partionning~~partitioning

Sublimation and condensation are diagnosed from the distributions of water in the three regions.

First, the region of ice clouds where $q_{cld}$ is lower than $q_{sat}$ is sublimated and transferred to the ~~subsaturated~~ sub-saturated clear-sky region. The corresponding tendency on cloud fraction $(\Delta\alpha_{cld})_{sub}$, is equal to the integral of $P_{cld}$ between $q_{vc}$ and $q_{sat}$. This process is activated only if $q_{vc} < q_{sat}$, typically because the temperature of the gridbox has increased.

Then, the water vapor in the cloud is adjusted to saturation, meaning that the excess vapor deposits onto existing ice crystals if $q_{vc} > q_{sat}$, or the ice crystals sublimate if $q_{vc} < q_{sat}$.

Finally, the water vapor in the ISSR that is above the condensation threshold $\gamma_{ss} q_{sat}$ is condensed and transfered to the cloudy region. The corresponding tendency on cloud fraction is $(\Delta\alpha_{cld})_{cond}$ and is equal to the integral of $P_{ss}$ between $\gamma_{ss} q_{sat}$ and $+\infty$. The three distributions $P_{sub}$, $P_{cld}$ and $P_{ss}$, are modified according to these processes. More specifically, we set $P_{cld} = 0$ and $P_{sub} = P(q)$ between $q^{vc}$ and $q_{sat}$ to account for the sublimation process, and we set $P_{ss} = 0$ and $P_{cld} = P(q)$ above $\gamma_{ss} q_{sat}$ to account for the condensation process.

\includegraphics[width=\textwidth, trim=0cm 0cm 0cm 0cm, scale=0.8] {Fig1.pdf}

Figure 1: Total water distribution for the standard cloud parameterization in LMDZ6A (a) and the new ice supersaturation parameterization (b). The vertical black lines in a) and b) separate the undersaturated and saturated regions, and correspond to $q_{sat}$ and $q^{vc}$, respectively. In b) the orange curve represents the $N \cdot P(q) = \alpha_{cld,adv} P_{cld}(q)$ curve. The situation illustrated in (b) is not an equilibrium state, but can be interpreted as the state at the beginning of the timestep before cloud physics is applied.

$q_{vc}$ is used to diagnose the dissipation of clouds (see Sect. 0.1.3) under the assumption that a decrease in the saturation threshold from one physical timestep to the next indicates a shift toward subsaturated state.

The formation of new clouds at each timestep is diagnosed by integrating $P_{ss}$ between $\gamma_{ss} q_{sat}$ and $+\infty$. In the subsequent physical timestep, the previous formed cloud fraction is conserved through $\alpha_{cld,adv}$, and its water is equally redistributed in $cld$ and $ss$ regions. This introduces additional water in the $ss$ region, leading to an overestimation of ISSRs. This overestimation is adressed by changing the proportionality coefficient $N$ to $N_1$ =min$(1, \Phi_N N)$ for $q \geq \gamma_{ss} q^{vc}$ and to $N_2$ (see Eq. 11) for $q^{vc} \leq q \leq \gamma_{ss} q^{vc}$, where $\Phi_N$ is a tuning parameter that can take values ranging from 1 to 2. This treatment is not physical, but the introduction of the parameter $\Phi_N$ provides flexibility to the parameterization, allowing it to fit observations through tuning.

$$N_2 = \frac{\alpha_{\mathrm{cld}} - N_1 \int_{\gamma_{\mathrm{ss}}\, q_{\mathrm{vc}}}^{+\infty} P(q)\, dq}{\int_{q_{\mathrm{vc}}}^{\gamma_{\mathrm{ss}}\, q_{\mathrm{vc}}} P(q)\, dq} \tag{11}$$

This treatment, which consists of decomposing $P(q)$ in three components, as described above, must not lead to a loss or creation of water in the gridbox ($q_t$ must be conserved).

### 0.1.4 Turbulence

We assumed that cirrus clouds have the shape of a prolate spheroid of volume $\frac{4}{3}\pi a^2 b$, where $a$ and $b$ are its semi-axes (see Fig. 2). The aspect ratio of cirrus clouds is thus defined as $\frac{b}{a}$, and is set to 3 based on sensitivity tests. Small-scale turbulence may mix the cloudy region in the gridbox with the subsaturated and ISS regions. Consequently, we took the turbulence into account by parameterizing it using a characteristic velocity, $v_{turb}$, and a characteristic length, $l_{turb}$. The $v_{turb}$ variable is determined as an approximation of the diagnosed Turbulent Kinetic Energy (TKE) and $l_{turb}$ is a tuning parameter with a typical value of the order of ten to a hundred metres but we used a default value of 50 m. It is also assumed that all the mass that is transferred from the cloud to the environment, or vice versa, occurs within an area around the cloud delimited by

\includegraphics[width=\textwidth] {Fig2.png}

Figure 2: Left panel: Prolate spheroid illustrating our assumption on the shape of clouds. Right panel: cross-section of a cirrus cloud (panel a)). Schematic distribution of the quantities $V_{cld}$, $V_{ss}$ and $V_{sub}$ in the cloud boundary layer. $L$ is the width of the turbulent mixing zone while $R$ is the distance from the centre of the cloud to the border of the environment around the cloud.

a length $L = \min(v_{turb}\Delta t, l_{turb})$. In the environment, this area is that between the boundary (distance $R$ from the centre) and that at distance $R + L$ from the centre (Fig. 2). In the cloud, it is the area between the boundary and that at distance $R - L$ from the centre. It also assumed that these two areas are well mixed by turbulence. The volume of the cloud environment, $V_{env}$, is the sum of the volumes of the subsaturated region, $V_{sub}$, and ISS, $V_{ss}$, which can be expressed through the introduction of a parameter $\omega$, such that:

$$V_{ss} = \omega\, V_{env} \tag{12}$$

and

$$V_{sub} = (1 - \omega)\, V_{env} \tag{13}$$

The impact of turbulence on interactions between clouds and the surrounding air depends on the thermodynamic state of that air and is treated differently for mixing between clouds and ISSRs and mixing between clouds and subsaturated clear air:

**Turbulent mixing between clouds and ISSRs:** In the presence of clouds near an ISSR, turbulence can enhance the diffusion of ice crystals from the cloud into the ISSR, thereby enabling the condensation of supersaturated water vapor, breaking the metastable state. Part of the ISSR becomes cloud, and total water in the cloud increases while that in the ISSR decreases. The cloud fraction tendencies over the time step are computed as follows:

$$(\Delta\alpha_{cld})_{turb,ss} = -\Delta\alpha_{ss} = \frac{V_{ss}}{V_{grid}} \tag{14}$$

$$(\Delta q_{ss})_{turb} = -\frac{q_{ss} \cdot V_{ss}}{V_{grid}} \tag{15}$$

$$(\Delta q_i)_{turb,ss} = -\frac{(q_{ss} - q_{sat}) \cdot V_{ss}}{V_{grid}} \tag{16}$$

where $V_{grid}$ is the total volume of the grid box.

**Turbulent mixing between clouds and sub-saturated clear sky:** For sub-saturated clear-sky regions, two situations are possible, depending on the value of $q_{eq}$ which is the equilibrium total water value after mixing clear and cloudy sky:

$$q_{eq} = \frac{V_{env} \cdot q_{sub} + V_{cld} \cdot q_{cld}}{V_{env} + V_{cld}} \tag{17}$$

- If $q_{eq} > q_{sat}$, meaning that there is enough ice in the cloud region to saturate the clear-sky air and still retain ice crystals, the portion of clear-sky that has been mixed transitions into cloud. The average cloud humidity decreases. These tendencies are computed as follows:

$$(\Delta\alpha_{sub})_{turb,sub} = -\Delta\alpha_{sub} = \frac{V_{sub}}{V_{grid}} \tag{18}$$

$$(\Delta q_{sub})_{turb} = -\frac{q_{sub} \cdot V_{sub}}{V_{grid}} \tag{19}$$

$$(\Delta q_i)_{turb,sub} = -\frac{(q_{sub} - q_{sat}) \cdot V_{sub}}{V_{grid}} \tag{20}$$

- If instead $q_{eq} < q_{sat}$, then all ice crystals sublimate without being able to saturate the clear-sky air. In this case, the cloud transitions to clear-sky, and the mean humidity in the clear-sky region increases. On average, the cloud humidity decreases. The corresponding tendencies are calculated as follows:

$$(\Delta\alpha_{sub})_{turb,sub} = -(1-\omega) \cdot \Delta\alpha_{sub} = \frac{V_{cld}}{V_{grid}} \tag{21}$$

$$(\Delta q_{sub})_{turb} = -\frac{(q_{sat} + q_i) \cdot (1-\omega) \cdot V_{cld}}{V_{grid}} \tag{22}$$

$$(\Delta q_i)_{turb,sub} = -\frac{q_i(1-\omega) \cdot V_{cld}}{V_{grid}} \tag{23}$$

To separate *clr* and *ss* and cloud boundary, we use $\omega = \alpha_{ss}/(\chi\alpha_{sub} + \chi\alpha_{ss})$, where $\chi$ is a tunable parameter. This formulation of $\omega$ accounts for the fact that there is a higher probability that the *cld* and *ss* regions are adjacent. In case of a homogeneous distribution in the cloud-free region, $\chi > 1$, otherwise $\chi = 1$.

Table 1: List of parameters in the ice supersaturation parameterisation. For each parameter, the range of values, the default or First Guess (FG) value and the physical process controlled are indicated. The names of the parameters as used in the tuning process are given in brackets.

| Name | Unit | Range | FG value | Controlled processes |
|---|---|---|---|---|
| $T_\gamma$ (TG) | K | [249, 269] | 259 | Saturation rate for homogeneous nucleation |
| $\gamma_0$ (TG0) | – | [2.3, 2.4] | 2.349 | Saturation rate for homogeneous nucleation |
| $\Phi_N$ (TN) | – | [1, 2] | 1.1 | Ratio between the $P_{cld}$, $P_{ss}$ and $P$ |
| $\beta$ (RQST) | – | [1, 5] | 3.0 | Width of $P$ |
| $\chi$ (KHI) | – | [1, 10] | 1.1 | Boundary size between ISSR, clear sky and cloudy region |
| l_turb (LTURB) | m | [25, 200] | 50.0 | Turbulent mixing length around the cloud |