Review 1

This article presents in-cloud scavenging coefficients that are function of rain rate for different types of clouds (cumulonimbus and stratus). Their suggested formulas can be a good reference for simplified models. I have several questions that need to be answered or clarified before publication.

1. They said that their approach is a theoretical approach, but I think there are many parameterizations and assumptions used to build their in-cloud scavenging coefficients. Also, their formulas are based on only one set of specific representation of cloud microphysics.

Our approach is described as a theoretical one because we rely on a microphysical description of all the interactions between the three phases water aerosol particles. Moreover, the description of these microphysical mechanisms is based on a physical description and not on a set of experimental data.

Note that this work aims in validating the approach. The sensitivity to more specific parameters will be presented in a following work. We will for example derive a spectral scavenging coefficient (Λ_{cloud}(d$_{\text{ap}}$)). Indeed, both activation and collection processes are spectrally dependant. Moreover, we will investigate the influence of background aerosol size distribution and nature on the scavenging coefficient.

For example, in equation (14), they adopted some collection efficiencies that were previously developed or parameterized by someone else. What if different collection efficiencies are used? Will the in-cloud scavenging coefficients be significantly different? I wonder how general the formulas they proposed in this study are.

The two main mechanisms involved in the incorporation of aerosol particles into hydrometeors are the activation of aerosol into droplets and their collection by droplets.

Indeed, the modelling of activation that we consider is the κ-Köhler theory introduced by (Petters, M. D., & Kreidenweis, S. M.,2007). This model is still a reference to account for activation process.

Concerning collection of aerosols by droplets, this process in modelled by a theoretical model built by Depee et al (2019). This model relies on Lagrangian tracking of aerosol particles around droplets falling at terminal velocity. This model from Dépée et al. 2019 has been further validated on laboratory experiments (Dépée et al., 2021, Part I ; Dépée et al., 2020, Part II). Note that Dépée, Lemaitre, Monier and Flossmann are author of both articles. In certain extent, the model development by Dépée et al. was a prerequisite performed in prevision of present article.

2. Are solid hydrometeors taken into account in-cloud scavenging? For the cumulonimbus case, I believe there are some ice or snow in the upper levels, and they certainly contribute aerosol

removal. At the cloud base, there are no solid hydrometeors? all melted? In this warm temperature case, there might no snow. However, let's assume temperature is low enough, and only snow is the precipitating hydrometeors (no rain). In this case, one cannot use their formulas that only consider rain. Do you have plan to include snow?

First, we focus on the in-cloud scavenging, so the below-cloud scavenging - made by snow, rain or any other hydrometeor is not the purpose. Second, we consider the main mechanisms occurring between the cloud base and the cloud top, including ice nucleation (line 156 -157 : "To model heterogeneous ice nucleation, we consider all the mechanisms described by Vali et al., (2015). The Biggs formula (1953) is used to describe immersion freezing and the model of Meyers et al., (1992) for condensation and contact freezing, as well as deposition nucleation. All these mechanisms have recently been incorporated into the DESCAM model by Hiron and Flossmann, (2015).").
However, in DESCAM, there is no modelling of aerosol collection by ice crystals, because it depends on the crystal morphology which would require a description of the crystal type (Magono and Lee, 1965). Nevertheless, for the cumulonimbus modelled in the present article, we believe that the contribution of collection by ice crystals to the global cloud scavenging is a second order mechanism compared to both activation and collection by droplets. This is because the concentration of interstitial aerosol particles when ice water content calculated by DESCAM is already low due a previous incorporation into the droplet according to warm processes that have acted previously (fig. 6a).
For the cumulonimbus model, depending on the cloud criteria considered to determine the cloud boundaries, the hydrometeor are either in ice state (figure 7a&b) or in liquid state (figure 7c). No main influence of the nature of the precipitation at the cloud base is observed from these three cases. This is because the scavenging coefficient is calculated on the basis of $\phi_{ap,\, precip}(dap)$ (eq. 15) that is determined by the summation of the flux of particle in both ice and liquid (eq.8).

3. They explained why status clouds are more efficient in removing aerosols even though the rain rates are the same for status and cumulonimbus. Besides the explanation using equations, can you provide more physically based interpretation?

The scavenging by stratus clouds is not more efficient than by cumulonimbus. However, if you parametrize the cloud scavenging by the rainfall rate, which is generally performed in the literature, it is observed that for a same rainfall rate the scavenging coefficient of status is higher than the one of cumulonimbus. Indeed, as the relative humidity is higher in cumulonimbus, there is more condensation and the same mass of aerosol collected (or activated) in diluted in a larger mass of water. However, this is balanced by the cumulonimbus rainfall rate which is on average 25 times higher than the stratus one.

4. I wonder if aerosol removal by falling hydrometeors within clouds can be called in-cloud scavenging. In-cloud scavenging usually indicates the removal by activation or collection. Based on their approach that examines the mass change of aerosols at the cloud base, I wonder if it is okay to include the effect of wash out in the in-cloud scavenging process.

The definition of what is an in-cloud scavenging could be discussed. The definition used here is for application to the atmospheric transport modelling. In these models, it is challenging to have access to the microphysical values like the share of activated aerosol, or the droplet/ice crystal size distribution. Whereas these models can have access to the rainfall intensity (whether the precipitation is rain or snow) and good guess to the cloud base and top altitude by the observation or processing of the NWP. That is the reason why this pragmatic definition of the in-cloud scavenging is used: all processes involved between the base and the top of the cloud.

5. Can scavenging coefficients be the same for mass concentration and number concentration of aerosols in equation (1)?

Both definitions are equal as far as they are written spectrally (as we do) and with a uniform density of all the aerosol particles. In our study, we assume the aerosol particles consisted of ammonium sulphate, so this equality is true. If the aerosol particles density in not uniform, which is the case in the atmosphere mathematical we should indicate if it is a mass scavenging coefficient or a number one. To avoid confusion, we added the exponent m to the scavenging coefficient $\Lambda_{cloud}^{m}\left(d_{ap}\right)$ in case of application of our model to a population of aerosol with various density. Indeed, the scavenging coefficient we derived is based on mass fluxes (eq. 3).

6. I recommend improving their English writing through the manuscript.

The manuscript has been proofread by a scientist native English speaker. Thanks to his helpful contribution, we choose to associate him (Daniel Hardy) as a co-worker of this article.

Review 2

I - General comments

This manuscript introduces a new parameterization for the in-cloud scavenging of atmospheric particles. The proposed scavenging coefficient is derived from the analysis of simulation results of the DESCAM cloud-resolving model. The derived parameterization, representative at the cloud scale, can be adjusted to treat both stratus and cumulonimbus clouds. Given the considerable uncertainties that remain in the scavenging process in clouds and the way in which its impact on pollutant concentrations can be represented, this is clearly a subject of scientific interest and within the scope of atmospheric chemistry and physics. The general presentation of this interesting work is clear and straightforward, and the results are quite convincing. I think it would be interesting from time to time to expand on the discussion of the limitations associated with the choices made. I also think that some careful proofreading and changes to certain turns of phrase would be useful to improve the accuracy of the text.

Thank you for your general comments and the questions. We feel these will improve the clarity of few points of the article.

II - Specific Comments

1. Introduction :

l40: The reference cited, Petroff et al. (2008), relates only to dry deposition, whereas the sentence seems intended to cover all the deposition processes. Probably that some other relevant publications could be added here (e.g. Modeling the Processing of Aerosol and Trace Gases in Clouds and Fogs, Barbara Ervens, Chemical Reviews 2015 115 (10), 4157-4198, DOI: 10.1021/cr5005887)

We amended the reference and added Croft et al., 2010 and Ervens, 2015.

l51: The reference cited, Querel et al (2021), is focused on operational atmospheric transport model devoted to radionuclides dispersion and certainly not summarised "all these models".

This is true we modified to:
“Quérel et al., 2021 summarised few of them in Table 3 of their article.”

2. Definition and theoretical context :

l67: It might be useful to remember that the equivalence of the scavenging coefficient for number and mass is based on the assumption of homogeneous particle densities and morphology (and potentially other properties) for the size class under consideration. The scavenging coefficient should otherwise be distinguished.

The two definitions are considered equal when expressed spectrally, as we have done, assuming a uniform density of all aerosol particles. In our study, we specifically considered aerosol particles composed of ammonium sulphate, thus confirming this equality. However, in real atmospheric conditions, aerosol particle density is typically not uniform. In such cases, it becomes crucial to specify whether we are referring to a mass scavenging coefficient or a numerical one. To prevent any confusion, we introduced the exponent "m" to the scavenging coefficient $\Lambda_{cloud}^{m}(d_{ap})$ when applying our model to aerosol populations with varying densities. This adjustment ensures clarity, particularly since the scavenging coefficient we developed is based on mass fluxes (as shown in equation 3). This modification is added in each equation.

l90-92 and equation 5: For greater clarity, I suggest specifying that the cloud volume nu_Vdrop is a function of D_drop.

For the sake of clarity, we changed $\mathcal{V}_{\mathcal{D}_{drop}}$ to $\mathcal{V}(\mathcal{D}_{drop})$

l140-150 : Insofar as the equations provided are not explicit, I am not fully convinced that this is the most relevant way to present the processes considered in DESCAM? A simple text could probably be just as effective?

We have made a few changes to this part of the article. Each term in equations 9 to 13 corresponds more explicitly to a microphysical mechanism shown in Figure 2. Moreover the term : $\left.\frac{\partial\mathcal{N}(d_{ap})}{\partial\mathcal{S}}\right|_{\kappa-\text{Köhler}}\left.\frac{\partial\mathcal{S}}{\partial t}\right|_{dyn}$ of equation 9 has been replaced by $\left.\frac{d\mathcal{N}(d_{ap})}{dt}\right|_{hygro}$ in order to refer explicitly to the hygroscopic growth of the aerosol. Each subscript that doesn't explicitly refer to a microphysical mechanism in fig 2 is now explicitly introduced in the text (line 152->155):

“In these equations, except for the index $(\ \ |_{dyn})$ denoting variations due to atmospheric transport, each term corresponds to one of the microphysical processes outlined in Figure 2. For instance, $(\ \ |_{act,\ deact})$ denotes to activation and deactivation processes. The subscripts $co\ell\ell,\ hygro,\ frz,\ sub,\ coal,\ cond,\ vap$ and agg respectively indicate aerosol collection by droplets, hygroscopicity, freezing, sublimation, coalescence, condensation, vaporization, and aggregation processes.”

Nevertheless if the equations are kept, several typos need to be corrected :

- equation 9 : subscript Köhler is not introduced in Figure 2.

This section has been revised and Equation 14 has been added in the article to provide a clearer explanation of how the hygroscopic growth of aerosol particles is calculated. These particles are considered in thermodynamic equilibrium with the surrounding air. This equilibrium, influenced by the supersaturation within the parcel, is described using the k-Köhler theory.

$$\left.\frac{d\mathcal{N}(d_{ap})}{dt}\right|_{hygro} = \left.\frac{\partial\mathcal{N}(d_{ap})}{\partial\mathcal{S}}\right|_{\kappa-\text{Köhler}}\left.\frac{\partial\mathcal{S}}{\partial t}\right|_{dyn} \qquad \text{Equation 14}$$

- equation 11 : a "d" should be removed I guess, the subscript "fra" is not introduced.

True, a "d" should be removed: this typo is corrected. “frz” is now introduced.

Equation 14: Variables of the equation are not introduced.

The variables that were not previously introduced are defined bellow the equation. We added line 175 -176 just below the equation:

“In this equation $\mathcal{U}_{\infty,\,droplet}(\mathcal{D}_{drop})$ corresponds to the terminal velocity of a droplet of diameter $\mathcal{D}_{drop}$, ρ_{ap} is the density of the aerosol particle, and $\mathcal{RH}$ refers to the relative humidity of air in the parcel. “

3. Applications

l217-218 : All the DESCAM simulations appear to have been done with a kappa value representative of ammonium sulphate. But further on (l408) a comparison is proposed with previous works for caesium-137. Is the kappa value chosen representative of caesium-137?

No, in this part, the chosen Kappa value remains that of ammonium sulphate. The feedback from Fukushima accident (Kaneyasu et al., 2012) have highlighted that caesium-137 has been transported on sulphate aerosol for long-distance. Because in-cloud processing happened generally to such distance, the transport of caesium - 137 by sulphates is plausible.

We add line 421 – 423:

The comparison is made using the κ value of ammonium sulphate. This decision is based on the findings of the Kaneyasu et al. (2012) study, which demonstrated the long-distance transport of cesium-137 by these particles – a distance particularly relevant for in-cloud scavenging.

And add the reference to Kaneyasu.

Equation 15: If the scavenging coefficient proposed is integrated over the entire aerosol distribution, the dependence to "d_ap" should be removed.

Yes, this is corrected thank you:

$$\Lambda^{m}_{cloud} = -\frac{1}{dt}\frac{d\mathcal{M}}{\langle\mathcal{M}\rangle}\bigg|_{cloud} = \frac{\phi_{ap,\ precip}}{\langle\mathcal{M}\rangle\ .\ H_{cloud}}\Bigg| \qquad \text{Equation 1}$$

l253 : Could the authors precise how the thresholds are chosen? It appears they differ between the cumulonimbus and the stratus simulation.

These thresholds were chosen arbitrarily, mainly to observe their influence on scavenging when they vary over wide ranges of values for an hour period. The duration of 1 H is characteristic of both the duration of our microphysical models and the time step of the NWP used as input data for atmospheric dispersion models. Once critical supersaturation has been reached at a certain altitude, this altitude can be taken as the cloud base, and this altitude can be considered constant over this one-hour period. The need to a more or less constant cloud base definition is mentioned L316 : “However, this numeric concentration criterion, although more precise for theoretically assessing the scavenging coefficient, is not easily accessible in a crisis code. Nevertheless, detailed analysis of the results of these simulations seems to show that it would be wise to define the cloud base as being constant and equal to the altitude at which critical supersaturation was first reached, i.e., the altitude at which the cloud began its formation.”

l268: Could the need for a bijective relationship between the scavenging coefficient and a set of meteorological parameters be discussed? Is this just a practical choice, or is there a fundamental reason?

The usefulness of a bijective relationship is twofold. One aspect is the use of the model for inverse calculations. If the relationship between scavenging rate and meteorological data is bijective, it will be easier to estimate the discharge at the point of origin. The other aspect is the robustness of the model. A deposition scheme that correlates scavenging rate well with weather parameters implies a relationship that is robust to small variations in values.

Figures 8 and 10: The figure 6 shows rain rates that do not exceed 50 mm/h, but Figures 8 and 10 show values beyond 60 mm/h. Both correspond to the same simulation.

All these results are from the same simulation; however, the rainfall rates are not calculated at the same location. On figure 8, 9 and 10 the rainfall rates are determined at the cloud base of the associated figure. Whereas on figure 6 it is determined at the ground level. These variations of rainfall rates between cloud base and ground level are due to raindrop evaporation on these almost 3 km freefall.

By the why, one can also notice that even between the three cloud boundaries (Fig. 8, 9 and 10) there is a slight difference between the rainfall rates calculated at the expected cloud base. This difference is due to crystals collection by falling hydrometeor inside cloud when its base is incorrectly evaluated.

Figures 15, 16 and 17: The figure 12 shows rain rates that do not exceed 1.5 mm/h, but Figures 15, 16 and 17 show values beyond 2 mm/h. Both correspond to the same simulation?

Same answer as previously, the difference between fig 15, 16 & 17 and figure 12 is linked to droplet evaporation between cloud base and ground level.

4. Section "Comparison with the literature"

I suggest to change the title of this section or to split this section in two. The second part of this section is interesting but has nothing to do with a comparison with literature?

This is true we rename this section:

Comparison with the literature and unification of the scavenging coefficient scheme for a cumulonimbus and a stratus

l428-437: Is all this paragraph really useful to explain that the direct comparison of the parameterization derived for cumulonimbus is probably not really relevant?

It seems necessary to explain the meteorological context of the Fukushima accident and why some previous scavenging schemes are better suited to stratus.

l438-471 : It seems to me that the general aim of this paragraph could be largely clarified. As it stands, the main part of the paragraph is an explanation of the lower scavenging coefficient derived for cumulonimbus on the basis of a scavenging coefficient depending only on rain intensity. From my point of view, this is interesting, but not directly the final objective which is to propose a common parameterization for cumulonimbus and stratus. This objective might be the title of the last section for instance?

True, the section has been renamed, and the introduction to the section has been adapted to the new title.

“Comparison with the literature and unification of the scavenging coefficient scheme for a cumulonimbus and a stratus”

Our calculations show that cumulonimbus scavenges less than stratus under the same rainfall intensity. How can we understand this and unify the two equations? This phenomenon can be attributed to the significantly higher level of supersaturation observed in cumulonimbus clouds (Figure 7.a) compared to that in stratus clouds (Figure 13.a). Hence, if the supersaturation is higher there is more condensation and the same mass of aerosol collected (or activated) in diluted in a larger mass of water. However, this is balanced by the cumulonimbus rainfall rate which is on average 25 times higher than the stratus one.

l453 : The variable s_v,w is not introduced?

This is a typo, $s_{v,w}$ is replaced with $\mathcal{S}$. (defined line 454: with supersaturation $\mathcal{S}$)

l468-471 : The way in which the correction coefficient is finally determined is not clear to me as it stands. Could the authors provide more details on this?

Text and equations have been rewritten for the sake of clarity. We hope that this section is now easier to understand.

As the Kelvin effect (linked to the curvature of the interface) and the solute effect become very quickly negligible after activation of the aerosol, this equation can be greatly simplified and reduced to: $\mathcal{D}_{drop} d\mathcal{D}_{drop} = \mathbb{C}.\mathcal{S}dt$, where $\mathbb{C}$ is a constant, enabling it to be integrated analytically, Δt the lifetime of the drop, to give: $\mathcal{D}_{drop} = \sqrt{\mathcal{D}_{drop,\,t_0}{}^2 + 2\mathbb{C}.\mathcal{S} \cdot \Delta t}$. Thus, to assess the effect of dilution of the aerosol in the droplet due to condensation, we can write:

$$\frac{\mathcal{D}_{drop}^{stratus^2}}{\mathcal{D}_{drop}^{cumulonimbus^2}} = \frac{\mathcal{D}_{drop,\,t_0}{}^2 + 2\mathbb{C}.\langle\mathcal{S}\rangle_{stratus} \cdot \Delta t_{stratus}}{\mathcal{D}_{drop,\,t_0}{}^2 + 2\mathbb{C}.\langle\mathcal{S}\rangle_{cumulonimbus} \cdot \Delta t_{cumulonimbus}} \qquad \text{Equation 2}$$

$\Delta t_{stratus}$ and $\Delta t_{cumulonimbus}$ are respectively the lifetime of the drop into a stratus and a cumulonimbus. Attention, in this equation, $\mathcal{D}_{drop}^{stratus}$ and $\mathcal{D}_{drop}^{cumulonimbus}$ are not the diameters of the droplets in the stratus and in the cumulonimbus, but the diameters they would have had, if only the condensation mechanism had caused them to grow. We are in fact seeking to assess how large will be the dilution of aerosol material in the droplets related to vapour condensation. There are other mechanisms modelled in DESCAM (such as coalescence or riming, **Erreur ! Source du renvoi introuvable.**) that lead to the growth of hydrometeors, without necessarily diluting the aerosols in the droplets. If there had only been the condensation mechanism, we could have used **Erreur ! Source du renvoi introuvable.** directly to assess this dilution. When precipitation begins, further simplification can still be made because $\mathcal{D}_{drop,\,t_0}{}^2 \ll \mathcal{D}_{drop}^{stratus^2} < \mathcal{D}_{drop}^{cumulonimbus^2}$. Finally, we can write:

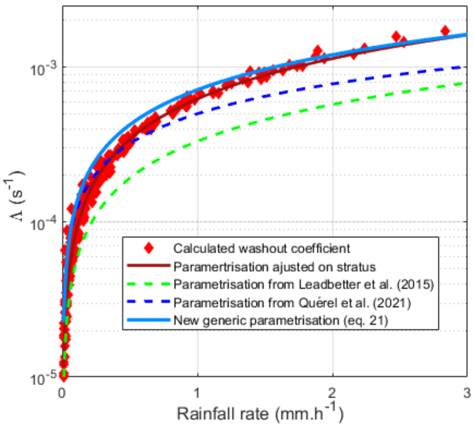
$$\frac{\mathcal{D}_{drop}^{stratus^2}}{\mathcal{D}_{drop}^{cumulonimbus^2}} = \frac{\langle\mathcal{S}\rangle_{stratus} \cdot \Delta t_{stratus}}{\langle\mathcal{S}\rangle_{cumulonimbus} \cdot \Delta t_{cumulonimbus}} \qquad \text{Equation 3}$$

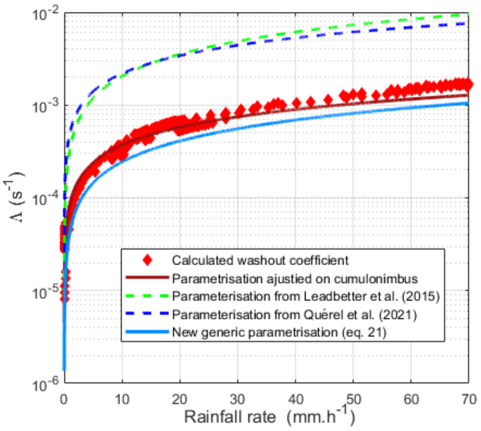
In this equation, the lifetimes $\Delta t_{stratus}$ and $\Delta t_{cumulonimbus}$ are therefore the durations necessary for the formation of precipitation under the cloud. These durations are assimilated to the duration of precipitation formation. For each of the types of cloud, we observe in **Erreur ! Source du renvoi introuvable.**.a and **Erreur ! Source du renvoi introuvable.**.a that these durations are very similar (≈2200 s), which allows us to write:

$$\frac{\mathcal{D}_{drop}^{stratus}}{\mathcal{D}_{drop}^{cumulonimbus}} = \sqrt{\frac{\langle\mathcal{S}\rangle_{stratus}}{\langle\mathcal{S}\rangle_{cumulonimbus}}} \qquad \text{Equation 4}$$

Figure 20 : The differences with Figure 18 are not obvious. It would be interesting to trace the previous version of the parameterizations here.

Yes, indeed. Both curves have been added to the figures 20.





III - Technical corrections

l32 : improving -> improve / impacting -> impact

modified

l43 : The reference to Laguionie et al. (2014) seems missing.

The reference is added.

Figure 2 : The variables written in red and blue are very difficult to read. Another color should be considerefor better visibility, the colors have been modifiedd.

for better visibility, the colors have been modified.

l172: In the legend some exponents appear as subscripts.

After a careful proof reading, no error of this kind appears on the legend of the Figure 3. Nevertheless, this legend have rewritten to be easier to read.

l174: A semi-column is missing and one should be replaced by colon.

See the previous answer.

Equation 15: What is the meaning of the vertical bar?

It is a typo the bar is removed.

Figure 7 : It seems the colors are not fully consistent (orange vs red) between the figure and the legend?

This is corrected, thank you;

l296-297 : 0.003 cm-3 instead of 0.003 m-3

Error corrected.

l366 : "their drop velocities" <-> "their velocities"

It is a typo the bar is removed.

l392 : "status" <-> "stratus"

This is corrected, thank you;

l408 : Section 2.1 should be section 2.3

This is corrected, thank you.

l414: Querel et al. (2017) is not referenced?

This is a typo, we wanted to refer to Quérel *et al.* (2021). It is corrected.

Quérel, A., Quélo, D., Roustan, Y., & Mathieu, A. (2021). Sensitivity study to select the wet deposition scheme in an operational atmospheric transport model. *Journal of Environmental Radioactivity*, *237*, 106712.

l423: Querel et al. (2015) is not referenced?

This is a typo, we wanted to refer to Quérel *et al.* (2021). It is corrected.

Quérel, A., Quélo, D., Roustan, Y., & Mathieu, A. (2021). Sensitivity study to select the wet deposition scheme in an operational atmospheric transport model. *Journal of Environmental Radioactivity*, *237*, 106712.