

2nd REFEREE

This paper investigates the influence of bacteria (*P. Syringae*) on both precipitation (precipitable and non-precipitable ice) and cloud electrification which is connected to the mass fluxes of hail and graupel within storm cells. This is an interesting aspect of heterogeneous ice nucleation presented in form of a modeling case study for a convective event in São Paulo. However, I cannot recommend this paper for publication in its present form since there are two major concerns that I would like to raise: first, the parameterization for the ice nucleation behavior of *P. Syringae* needs revision. Secondly, the estimation of lightning frequency as a function of precipitable and non-precipitable ice mass fluxes is not explained very well and thus it is not comprehensible to the reader. In principle, however, I think that work on this interesting case study should be continued and because of that I have listed points that I find critical as well as technical corrections and hope that this will be helpful to the authors.

Major points:

1) Ice nucleation parameterization

- p. 26150, l. 10-16: A more recent parameterization for homogeneous ice nucleation is given in Koop et al. (2000). Maybe you could compare between the parameterization by deMott (1994) and the one derived by Koop et al. (2000). Also, please cite Pruppacher et al. (1997).

A: RAMS already includes some simple parameterizations of supercooled water. This is not the focus of this study, and we maintained the parameterization without any further modification along the simulations. Koop et al. (2000) worked with supercooled water in a very specific parameterization case. They have shown from experimental data that the homogeneous nucleation of ice from supercooled aqueous solutions is independent of the nature of the solute, but depends only on the water activity of the solution.

We used the information from DeMott (1994) only in order to compare the IN concentration and its respective temperature range.

We added Pruppacher reference at the very beginning.

- p. 26150, l. 16-24: For allowing the reader to understand how the maximum concentration of ice nucleating bacteria per l of cloud water was derived, it would be good to proceed in two steps: first, a range of atmospherically observed or estimated bacteria concentrations in air should be given (Amato et al., 2005; Burrows et al., 2009). Then, from observed ice fractions for ice-active bacteria (Möhler et al., 2008; Yankovsky et al., 1981; references in Philips et al., 2009) a parameterization for immersion freezing of *P. Syringae* could be derived. The calculated IN scenarios could then compared to concentrations of ice-active bacteria in cloud water derived from snow samples. If within the model framework the activation of bacteria to CCN was possible, a comparison between this calculated number and observed cloud water concentrations would be even more interesting and would underline the predictive power of BRAMS.

A: The maximum concentration is given by Figure 2 is based on data from Amato's work where he presents data for total bacterial concentrations in cloud water. From data on the fraction of *P. syringae* in snow and rainfall (Morris et al 2008) we can estimate the fraction of the total cloud water bacteria that consists of *P. syringae*. Furthermore, Amato isolated *P. syringae* from cloud water, therefore this extrapolation is reasonable. Based on a wide range of data in the literature, we also know the fraction of cells in a population of *P. syringae* that are INA at a certain temperature. Hence, we used the ensemble of these values to establish a range of INA cell densities in our cloud models.

We understand the scenarios that you are describing. However S. Paulo is a subtropical city where there are no examples of snow in the surroundings. Our scenarios are the simplest ones. We plan to sophisticate our scenarios in further studies.

- p. 26150, l. 21-23: "As no observational data were available at temperatures colder than -12 C, the IN concentration for -10 C was used. . .". Please check references in Philips et al. (2009) for data at lower temperature and as pointed out before consider developing an enhanced parameterization from a wider selection of data. It is not clear to me how the ice nucleation parameterization could be derived based on the information given in Amato et al. (2005, 2007). Maybe a reference is missing here.

A: Extrapolation of INA rates were based on the described curves for *P. syringae* such as that presented in Orser et al (Orser C, Staskawicz BJ, Panopoulos NJ, Dahlbeck D, Lindow SE (1985). Cloning and expression of bacterial ice nucleation genes in *Escherichia coli*. *J. Bacteriol.* 184: 359-366.) where the activity of the proteins from strain Cit7 were expressed at a range of temperatures. This type of curve has been repeated for numerous strains of *P. syringae*.

- p. 26150, l. 24/25: “. . .P. Syringae IN concentrations were assumed homogeneous over the whole model domain. . .” Does this mean that the bacterial concentration is not only homogeneous over the horizontal dimension, but also over the vertical column? If yes, additional assumptions about the atmospheric distribution of the *P. Syringae* bacteria should be made.

A: Yes, it is homogeneous, as all other IN and CCN at (B)RAMS parameterizations. This supposition is based on the fact that the Boundary Layer below the cloud has a homogenous concentration of gases, temperature and aerosols as expected for micrometeorological standards. We added “vertical and horizontal” at the text.

- p. 26150, l. 26: Why is depletion, e.g. by scavenging, not considered?

A: Depletion/scavenging is not considered for the same reason given by the answer for the other referee. The cloud modeling (as well as in the reality) is continuously fed back by the new air masses, from the same original concentrations. Rain (i.e. scavenging processes) takes place in different position and age of the cloud life.

- p. 26150, l. 27: “All bacteria [. . .] induced ice formation.” This is contradictory to the statement in l. 18, where an ice fraction of 10^{-5} is assumed. Please either clarify or delete.

A: We will modify at the text. We assumed that the all bacteria, in these scenarios, from that fraction which led us a misunderstanding.

- p. 26151, l. 14: Setting the cutoff for the background ice nucleation at -8 C seems very arbitrary, especially if the authors' intention is to demonstrate that bacteria might have a significant influence in comparison to scenarios where only background IN are present. Does this threshold have a major influence on the results obtained under the assumed environmental conditions?

A: This is based on the RAMS default parameterization. All RAMS and BRAMS simulations use this threshold. Our article is trying to show exactly that this should be revised.

2) Cloud electrification parameterization

- p. 26151, l. 25/26: Please state how "precipitable" and "non-precipitable" ice masses are defined.

A: The terms precipitable and non-precipitable ice masses are then associated to the hydrometeor types likely to precipitate or not relatively to the convective core updraft, i.e., denser particles like hail and graupel can precipitate over the convective core and are defined as precipitable ice mass, while snowflakes and ice crystals cannot and are therefore defined as non-precipitable ice mass. This explanation was included in the text.

- p. 26152, l. 4: It is not clear where eq. 1 comes from, since the referenced publication by Barthe et al. (2007) is an abstract which does not contain any formula. Also, the functional form of the relation given in Deierling et al. (2008) is different from eq. 1 with $f = 9.0 \cdot 10^{-15} \cdot f_{np} \cdot f_p + 13.4$. Please give a detailed explanation and reference accordingly. Under which atmospheric conditions can eq. 1 be employed?

A: More references on the relationship between total lightning frequency (f) and precipitable and non-precipitable ice mass fluxes (f_p and f_{np}) were included in the text. We have referred Deierling et al.'s relationship $f = 9.0 \cdot 10^{-15} \cdot f_{np} \cdot f_p + 13.4$ directly as the equation used to

calculate f from our model data, and have included a brief description of the storms used to find this equation.

- p. 26152, l. 13 (eq. 3) and l. 20-24: In the publication by Deierling et al. (2008) it is stated that non-precipitable ice is identified with help of the horizontal divergence of the wind velocity. Was this criterion also employed for the numerical simulations in this paper? The non-precipitable ice flux is not given by eq. 3, but by summing up over the components as calculated with eq. 3. Please correct as in Deierling et al. (2008) the flux of non-precipitable ice is given by $f_{NP} = m_{NP} \cdot w$ which means that f_{NP} and f_P have the same units.

A: The hypothesis of equation 1 says that $f_{NP} = m_{NP} w$ (Deierling et al (2008); Blyth et al. (2001); Latham et al. (2004)). However, w cannot be directly inferred from Doppler radar data, so Deierling et al. used the horizontal divergence wind velocity to calculate f_{NP} as $f_{NP} = m_{NP} \partial w / \partial z = m_{NP} (\nabla_H \rho_0 \vec{V})$. This is fully explained in section 3.2 from Deierling et al. (2008) paper. Although we do have w at the model simulations, we had to infer w from horizontal wind divergence as in Deierling et al. to be consistent with their relationship ($F = 9.0 \times 10^{-15} \cdot f_P \cdot f_{NP} + 13.4$). We have included a few lines about this in the text to better explain the methodology used to calculate F .

Minor points:

- p. 26144, l. 9-11: The mentioned maximum IN concentrations (10^2 to 10^3 bacteria per l) do not match with the concentrations illustrated in Fig. 2, where IN concentrations correspond to maximum values of 102 to 104 bacteria per l. Please clarify.

A: We modified the text from “ 10^2 to 10^3 ” to “ 10^2 to 10^4 ”

- p. 26144, l. 11: Please point out that the S5 and S6 scenarios based on the RAMS ice nucleation parameterization was used as reference cases.

A: we modified it.

- p. 26144, l. 13: From the formulation "the chosen radiosonde data" it is not clear how this data is exactly related to the numerical simulations. Please clarify how this data was used for the initialization of the modeled temperature and humidity profile.

A: We included a new picture of the radiosonde profile with all data for a homogenous initialization required for RAMS simulations.

- p. 26146, l. 19: In the study by Morris et al. (2008) also values for the bacteria concentrations in rain water are mentioned (up to 10^4 bacteria per l). Please add.

(A: We changed to: The maximum concentration of *P. syringae* observed in fresh snow fall is 10^5 bacteria l^{-1} and in rain is 10^4 bacteria l^{-1} (Morris et al., 2008).

- p. 26146, l. 21-25: The study by Möhler et al. (2008) found maximum ice-active fractions of 10^{-4} for *P. Syringae* and other bacteria species in the temperature range between -7 and -11C. This value could be added as a second reference for the ice-active fraction besides Orser et al. (1985).

A: We added it.

- p. 26147, l. 13-21: In this paragraph the role of mineral dust particles acting as CCN and IN is highlighted. As mineral dust belongs to the most abundant aerosol species in the atmosphere, and thus has a major influence on atmospheric processes it certainly deserves to be mentioned. However, for adding value to the manuscript and emphasizing that it is worthwhile to investigate bacteria, it would be necessary to briefly compare the role of mineral dust particles to that of biological particles (e.g., mention CCN activation thresholds, freezing ranges, etc).

A: We added information from Table 9.6 pg 219, in Pruppacher 1997.

- p. 26148, l. 7/8: “. . .increasing the total amount of electrical charge transferred and the charged centers, which in turn increases the lightning activity in the cloud.” How is lightning activity defined here? Is the relation between the number of charged centers and lightning really that straightforward? What role do storm cell structure, cloud dynamic processes and their respective time scales play?

Please explain.

A: Lightning activity (number of intracloud and cloud-to-ground flashes) only happens if you have at least two major charge centers of different polarity (i.e., one positive and one negative). These charge centers are large in its extension (horizontal and vertically) corresponding to the major cloud microphysics characteristics produced by the cloud, i.e., amount of graupel+hail and amount of ice crystals (aggregates, snow, pristine crystals, etc), and how it is distributed inside the cloud by its motions, vertical and horizontal. The major player is the vertical motion (up and downdrafts) that vertically segregates the smaller (less dense) and larger (denser) particles inside the cloud to higher and lower levels, respectively. As smaller and larger particles are charged with opposite signs, major charge centers are created that can produce lightning when they are large enough to break up the dielectric breakdown (in an attempt to neutralize the electrical potential energy between the centers). Cloud dynamics then do play an important role in the electrical charge structure of the cloud as it controls the vertical and horizontal motion of hydrometeors inside the cloud. This explanation was briefly introduced in the text.

- p. 26148, l. 26: Has the BRAMS model also been validated for other regions than the Amazon, i.e. for the urban area of São Paulo? If yes, please add references.

A:What exactly do you mean by validation? We have many articles using RAMS and BRAMS at the S.Paulo urban area such as:

Gonçalves, F.L.T., Massambani O., Beheng, K.D., Vautz, W., Schilling, M., Solci, M.C., Rocha, V., Klockow, D., 2000 “Modeling and

measurements of below cloud processes in the highly industrialised region of Cubatão- Brazil."Atmospheric Environment, vol. 34, pag.: 4113-4120.

Gonçalves, F.L.T., Malheiros, A. R., Freitas, R. S., Assunção, M. A. F., Massambani O 2002 *In-cloud and below-cloud numerical simulation of scavenging processes at Serra do Mar region, SE Brazil. Atmospheric Environment, vol.36, fasc.:33, págs:5245-5255.*

F. L. T. Gonçalves; W. N. Morinobu; M. F. Andrade, A. Fornaro, 2007. In-cloud and below-cloud scavenging analysis of sulfate in the metropolitan area of São Paulo, Brasil. *Rev. Bras. Meteorol.vol.22 (1),*

Edmilson D. Freitas, Christopher M. Rozoff, William R. Cotton and Pedro L. Silva Dias, 2007. Interactions of an urban heat island and sea-breeze circulations during winter over the metropolitan area of São Paulo, Brazil. Boundary-L. Meteorology, vol.122,(1), 43-65.

Freitas, E, Droprichinski, L.M., Dias, P.L.S., Andrade, M.F. , 2005. A simple photochemical module implemented in RAMS for tropospheric ozone concentration forecast in the metropolitan area of Sao Paulo, Brazil : Coupling and validation. *Atm. Env, vol. 39,n°34,pp.6352-6361*

• p. 26149, l. 8: ". . .according to Gonçalves et al. (2008)." In the referenced publication low level forcing is described as a "hot and wet bubble". Please describe in greater detail how the initialization is performed (temporal and spatial structure) and what part of the radiosonde data is used in this context.

A: The homogeneous initialization using one single radiosonde can also be forced by a hot and wet bubble, adding humidity and heat to the first level. Hot and wet bubble is more detailed described at this work.

- p. 26149, l. 23-25: "The configuration of additional microphysical parameters in the numerical simulations was adjusted according to values suggested in empirical studies." Please specify which parameters were adjusted according to what kind of empirical studies and add the corresponding references.

A:The empirical studies: additional micropysical parameters are those suggested by Meyers et al. (1992) to the ice-crystal formation by primary nucleation processes (see Figures 2,3 and 4) and those suggested by Hallett and Mossop (1974) from experiments that showed splintering of supercooled droplets impacting surface of riming ice particles.

- p. 26149, l. 25/26: ". . .parameters that directly impacted IN were the CCN concentration..." What is the relation between IN and CCN in the model for this statement to be made? CCN and IN concentrations are not necessarily related to each other.

A:They are not. We modified at the text.

- p. 26150, l. 1: ". . .shape parameter of the size distributions. . ." To which size distributions do the authors refer at this point? To the droplet size distributions? Or to all hydrometeor size distributions? Please clarify.

A: CCN affects only droplets. Therefore it refers only to droplet size distributions.

- p. 26150, l. 6: The authors assume "relatively clear atmospheric conditions". How can this be justified with regard to São Paulo being a highly polluted urban area?

A: As these simulations are just an intra comparison and most of the articles based on RAMS use shape parameter 2, we kept this set for our simulations. In future articles, we plan to refine these settings, adding shape parameter 6 as it is more compatible with S.Paulo polluted area.

- p. 26157, l. 12/13: “. . .BRAMS default IN concentration seems to play a secondary role at all, not affecting the total number of flashes. . .” Do you have any explanation for this behavior?

A: That is the main result of the article. Nucleation at higher/warmer temperatures seems to be more important than high IN concentration at colder temperatures. We should point out that this is a quite non linear processes (see Gonçalves et al., 2008).

- p. 26163, Table 2: Ice crystals may be partly belonging to precipitable ice and non-precipitable ice. Therefore it should be considered to differentiate between ice crystals of different sizes and crystals habits.

A: Yes, it is as explained above.

- p. 26170, Figure 3: The graphical representation of the temporal development of the different hydrometeor species is a very good idea in order to help the reader understand the results of your numerical simulations. However, it would be better if there were different graphs for each hydrometeor species since it is very difficult to distinguish between those species in the figure. Maybe you can restrict yourself to detailed representations of the hail and graupel populations and show integrated values for the other hydrometeor species in another graph.

A: We placed it in Tables 2 and 3. We can add more graphs, but it will be a higher number of them if we separate them at each hydrometeor type.

Technical points:

- Introduction: The introduction contains many interesting aspects of bacteria interacting with the atmosphere not only directly, but also via processes that are influenced by bacteria-induced ice nucleation in clouds. However, this section could benefit from a slight reorganization bringing together aspects that belong together in order to highlight the structure of argumentation as presented by the authors, with as a first point the role of bacteria as IN, then their natural abundance and subsequently the influence on both precipitation and rainfall.

A: The various atmospheric processes that bacteria can influence have been described in numerous papers. Here our goal is to present the information needed, in a logical sequence, to justify why we would investigate their role in the formation of lightning. Although there are different types of organization of the information that would achieve this goal, we do not feel that the suggestion of the reviewer would be adequate.

- p. 26144, l. 7-9, l. 14-16, l. 22: For the reader it might be easier to follow your line of argumentation, if you mentioned all aspects that were investigated (effect on total rain water, effect on cloud properties and precipitation, and total flash number) in the opening paragraph.

A: We placed it at the end of the Introduction, emphasizing that the main goal is the role of IN over flash rates (cloud electricity)

- p. 26146, l. 25/26: The highest concentration of biological IN found by Christner et al. (2008) was 120 IN per l, not 200 IN per l as stated in the manuscript. Maybe you could also mention the number of DNA containing cells ($1.5 \cdot 10^4$ - $5.4 \cdot 10^6$ cells per l) from this study.

A: ok. We added it.

- p. 26147, l. 1-13: In this paragraph a feedback mechanism involving bacteria and the hydrological cycle is mentioned. As this paragraph could serve more as an overview and a motivation for the research presented, it might be useful to put this paragraph at the beginning of the introduction.

A: Yes, we did that.

- p. 26148, l. 3/4: "When the electrical potential between these [charged] centers are strong enough to break up the electric breakdown of air, lightning is initiated". Maybe you can elaborate a bit on this point, i.e., describe the basic process (ionization of air, creation of plasma channels, breakup) and mention that lightning can occur within clouds, but also between cloud bottom and ground.

A: We modified it

- p. 26148, l. 9-13: How is this paragraph on precipitation related to the issue of lightning that is discussed before and afterwards?

A: This paragraph was excluded.

- p. 26149, l. 20: ". . .complete development of both liquid and ice phases." What does the term "complete development" mean?

A: The simulated low level forcing was based on Walko et al. (1995). This simulation produces complete development of both liquid and ice phases which means that all phases can be presented since there is CCN and IN acting.

- p. 26150, l. 17: Change "(S3 scenario)" to "(S2 scenario)".

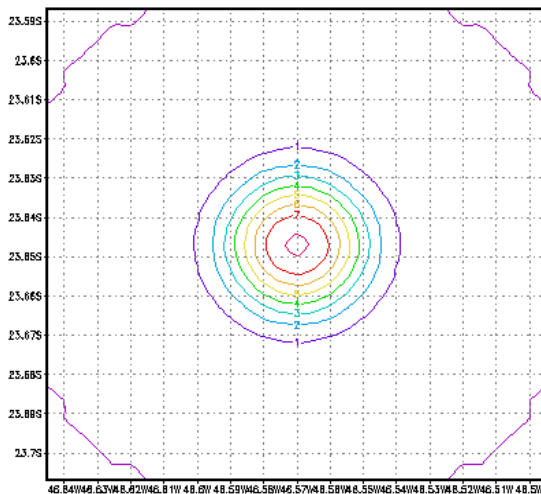
A; It has been done.

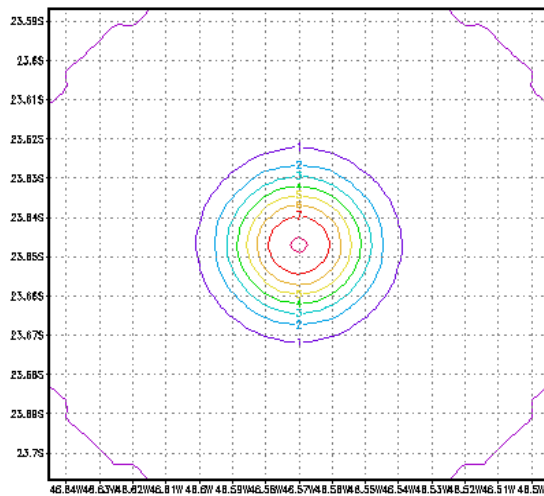
- p. 26153, l. 7: Change "Table 1" to "Table 2".

A: We modified it.

- Section 3.1: Regarding the structure of this section, it would be easier for the reader if general trends (e.g., decrease in rainfall) would given and then illustrated with a few examples. The authors could also focus more on the interpretation of their results.

A: the illustrations did not show any impressive visual difference as it is given below :





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- p. 26162, Table 1: Please add a note to the simulation S6 since the different freezing modes were only explicitly considered for the RAMS default parameterization, but not for the bacteria acting as IN.

A: S6 is a sum of S4 and S5 profiles, therefore, it contains both bacteria acting as IN plus RAMS default data. WE modified it at the text.

- p. 26164, Table 3: Please add units [g kg^{-1}]. In comparison to Table 2, rain is not listed as a hydrometeor in Table 3. If suitable, add maximum values and the corresponding time information.

A: We added it.

- p. 26165, Table 4: The phrase "lightning flashes" belongs to the fourth column.

A: We also modified it.

- p. 26168, Figure 1c: If available you could add a second figure showing the radar data for 12:00 GMT of March 3, 2003 in order to match the humidity and temperature data presented in Fig. 1b and thus to allow a better understanding

of the modeled "hot bubble" before the actually observed convective event at 18:00 GMT which is a point of time not within the modeled time period. Radar data for 15:00 GMT of March 3, 2003 might be used to compare the observed rainfall to the amount calculated in the numerical simulations.

A: We have more previous radar information. However, it does not add any new, presenting just few scattered points. We chose the higher cloud/precipitation developing at Figure 1c.

- whole manuscript: Please do a thorough check of punctuation and linguistic accuracy (i.e., complete sentences, spelling and similar).

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