1	Cachoeira Paulista, 21 April 2015,
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4 5 6	Reference: acp-2014-463: "The biomass aerosol influence on precipitation over the Central Amazon: An observational study" by Gonçalves et al.
7 8 9 10 11 12 13 14 15 16	Dear Dr. Philip Stier Editor of the Atmospheric Chemistry and Physics Enclosed please find the revised version of W. A. Gonçalves, L. A. T. Machado and P-E, Kirstetter paper entitled "The biomass aerosol influence on precipitation over the Central Amazon: An observational study", after the corrections/recommendations requested by the Reviewers. The paper was revised taking into account the reviewers comments. All remarks made by them have been addressed and are commented in the next pages. We would like to acknowledge the reviewer to the detailed analysis.
18 19 20	Sincerely,
21	W. A, Gonçalves
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Response to Reviewer:

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Thank you for your careful second review and comments to improve the quality of the manuscript, we really appreciated your revision. We hope now we were able to clarify all aspects and comments raised and the paper now is more complete and the main results more clearly presented. It is important to mention that after the English review the title of the manuscript was modified for "Influence of biomass aerosol on precipitation over the Central Amazon: An observational study"

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I) Comments from the editor

General issues:

- 1. This study uses BC as "aerosol tracer". It clearly is an aerosol but is it sufficiently
- 37 correlated with CCN to support this kind of analysis? Please present some evidence
- 38 from measurements or related literature.
- 2. The language of the manuscript remains an issue. Some parts are very well
- 40 written, e.g. most of page 4, others not so clear and need to be improved (e.g. page
- 41 2-3 or Conclusions). This has to be addressed.
- 42 3. The results sections should be limited to the presentation of the results and their
- 43 discussion. In this manuscript they include a significant amount of speculative
- 44 material. This should be clearly separated from the results and presented as
- 45 hypotheses in discussions or conclusions.
- 4. The conclusions are very strong yet highly speculative. It is important to make
- clear what has been shown in this study, what has been robustly backed up by other
- 48 evidence and what is speculation.

49 **Specific issues:**

- 50 1. Page 1: The affiliation numbering is incorrect.
- 51 2. Page 1: Abstract is missing
- 3. Page 1, L22-26: You refer to direct effects here but what you describe is a fast
- 53 feedback commonly referred to as semi-direct effects. This should be clearly
- 54 described and referenced.
- 4. Page 1, L28: You only focus on CCN. For deep convection you may want to discuss
- the role of IN early on.
- 5. Page 2, L6-11. The description and referencing of aerosol effects on precipitation
- 58 is not very clear. Rain suppression is described fairly simplistically while there is
- 59 quite a bit of controversy in the literature. Some of the references are not suitable as
- 60 primary reference, e.g. any rain suppression in Nober is by construction of the study,

- which can hardly be cited as an evidence of this effect and Kaufman is an interesting
- paper but maybe not the strongest reference on cloud microphysics.
- 6. Page 2, L12: "Diehl et al. (2007) suggested that the ice phase could be an important
- 64 factor in the rain process." The importance of ice has been realized a long time
- before Diehl. Please use primary references.
- 7. Page 2, L17-24: The description of the hypothesis presented in Rosenfeld et al.
- 67 (2008) is slightly confusing.
- 8. Page 2, L24-25: "Over a certain time, the cloud accumulates higher liquid water
- 69 and ice contents, favoring more intense rainfall rates and increasing electrical
- activity (Graf, 2004)." This is neither very clear not sufficiently backed up to be
- 71 presented as a fact.
- 9. Page 3, L19-21: "However, the CAPE dataset was evaluated and shown to have
- very useful information and capture the daily instability feature even though it was
- 74 not recorded at the most appropriate time." This is not very clear and needs to be
- 75 better explained. What was evaluated and what was found?
- 10. Page 3, L31: "Therefore, due to its potential for ice nucleation, the presence of
- 77 BC would favor the development of ice phase clouds, including deep convection."
- 78 This is very speculative and it seems very unlikely that BC regulates the occurrence
- 79 of deep convection as the formulation suggests.
- 11. Page 3, L33- Page 4, L4: This is another insufficient description of the semi-direct
- effects / fast feedbacks. The description needs to be improved and should be merged
- 82 with the above.
- 12. Page 4-5: The description of the indices RF, IRF, IF is insufficient. Explain their
- significance and their relation to the physical properties you try to represent. "1mm"
- is presented here without any explanation.
- 13. Page 6, line 15 and throughout manuscript: you use "elevation synonymous with
- 87 upslope. This is not necessary the case. Discuss.
- 14. Page 6, L17: "At this distance, the lower radar elevation band is around 1 km
- 89 high, which eliminates the ground clutter effect possibility. Thus, this area should
- 90 receive special attention under forest protection policies." I don't understand this
- 91 statement;
- 15. Page 6, L30-33: "Thus, the combination of a period of the year (wet season) with
- more rain and less aerosol concentration, and the other season (dry season) with
- 94 high BC concentrations and smaller amounts of rain but more intense precipitation
- events allowed us to reach interesting results." At the same time you point out one
- of the biggest difficulty to isolate any aerosol effect in your study emissions, thus

- aerosols, are highly correlated with the wet / dry season. Any variation could be an
- 98 effect of either. This needs to be discussed and considered throughout.
- 99 16. Page 7 L6: "After this value, the RF slightly increases." It seems unclear how
- 100 significant this increase is.
- 101 17. Page 7, L18-32: The attribution to a rain suppression mechanism here is fairly
- arbitrary. I cannot see any evidence to rule out scavenging as the dominant effect.
- This is certainly consistent with the higher rain fraction for lower BC concentrations
- in Fig. 4c. Please move speculative discussions from the results section and discuss
- 105 all possibilities.
- 18. Page 7, L29-30: "As the RF decreases for elevated BC concentrations, the wet
- deposition seems not to be the dominant effect on much polluted atmospheres,
- which gives support to the rain suppression theory." I don't understand this
- conclusion. Decreasing BC with increasing RF is entirely consistent with scavenging.
- 19. The description of the applied test for scavenging is insufficient: "Thus, the
- concept of this test was based on eliminating from our statistics all samples whose
- precipitation was observed over the EUCAARI site. This elimination excluded the
- samples whose precipitation could have cleaned the atmosphere, throughout the
- scavenging process." What data has exactly been excluded? The effectiveness of this
- screening will strongly depend on mixing time-scales (surface rain is not directly
- related to surface BC as the dominant scavenging mechanism will be activation
- scavenging, not below cloud scavenging). The scales of scavenging events are fairly
- small, introducing quite distinct gradients (c.f. Gryspeerdt et al., ACPD, 2015) so it is
- possible that this test will not work to screen out scavenging events. This needs to
- 120 be considered.
- 20. Page 8, L21-25: "This result could be an indication that convection is invigorated
- by higher BBA concentrations (Lin et al., 2006; Graf, 2004; Rosenfeld et al., 2008;
- Altaratz et al., 2010; Koren et al., 2012). Considering that high CAPE values are
- associated with stronger updrafts, the aerosol effect on the rainfall and in the
- severity of the convective processes could depend on the intensity of the vertical
- motions." I am not sure I am convinced by this argument. First, RF is purely
- geometrical it gives an intense rain fraction (horizontally). The link to cloud height
- or updraft strength is not obvious.
- 129 21. Page 8, L26-29: "The radiative effect that acts to stabilize the atmosphere is of a
- second order because even with high BBA the atmosphere is highly unstable, and
- thermodynamics, on this time scale, dominates over the radiative process. Also, the
- 132 feedback effect due to the radiative effect, which increases droplet evaporation, does
- not seem to be the predominant mechanism." I do not see any evidence or result
- corresponding to this statement. Please move from results section and provide
- evidence. Also, the effect of absorption is not limited to droplet evaporation. It could

- 136 be an efficient mechanism to maintain convective inhibition for certain
- 137 stratifications.
- 22. Page 8, L29-31: "Probably, the high instability (high updraft) and the large
- 139 number of droplets inside the cloud ascend very fast, thereby reducing the
- evaporation." This again is speculation. Possible, but it could be also very different,
- depending on the cloud structure. Turbulent entrainment is likely to increase with
- updraft velocity and will also increase evaporation. Please provide evidence for any
- such statements you make.
- 23. Page 8, L31-33: "Although impossible to quantify, the wet scavenging also seems
- to be of second order, and would act in the opposite direction through the fact that
- precipitation did not decrease BC concentration at any point of the curve (Fig. 4b). "
- 147 This is an interesting observation but the question is if your sampling strategy would
- be able to detect scavenging from such low rain fractions of <2%.
- 24. Page 8, L33: "During the dry season only the upslope regions trigger convection
- and in the highly unstable cases it appears that BBA helps to increase ice nucleus,
- increasing precipitation." I cannot see any results supporting this conclusion.
- 25. Page 9, L14-29: This section contains a large amount of speculation on effects for
- which there is no evidence. There is discussion of stratiform and convective clouds
- but no formal separation is made. The similarity of the ice fraction data to the rain
- fraction data seems suggests that ice fraction and rain fraction are highly correlated;
- this alone could explain the behavior shown in Fig. 5. This needs to be discussed or
- 157 explicitly excluded.
- 26. Page 9-10: Discussion of rain cell size: the plots closely follow the behavior
- shown for RF and IF before so it appears that RF, IF and cell size may be strongly
- 160 correlated. While the relationship appears strong, there is no evidence of causality,
- i.e. that it is that aerosols actually cause the observed relationship. Generally, high
- and low aerosol conditions occur in different synoptic situations with very different
- cloud regimes (c.f. Gryspeerdt et al., 2013,2014). A snapshot analysis can only
- reconfirm this relationship. The speculative part of this section should be separated
- 165 from the results and be identifiable as hypotheses. Personally, I would require some
- evidence to be convinced by the proposed mechanisms.

II) Author's response

General issues:

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- 169 1. Black Carbon concentration is well correlated with Aerosol concentration in
- Amazonas. Several studies in Amazonia have shown this relationship. Bevan et al.
- 171 (2009) use 13-year time series of Along Track Scanning Radiometers (ATSR)
- derived aerosol optical depth (AOD) measurements to examine the role of aerosols
- in the interaction with biomass burning over the Amazon. AOD has a significant

positive correlation with the total number of satellite-observed fires. From the other side, many studies used aerosol particles greater than 80 nm as a proxy for CCN. Andrea et al. (2008) and Liu and Li (2014), commented the positive relationship between aerosol concentrations and CCN.

Figure 1 is a scatterplot between Black Carbon and aerosol particles greater than 80 nm. This figure is a courtesy from Dr Joel Ferreira de Brito and Dr. Paulo Artaxo. This is a result, still not published, from the ATTO (Amazonian Tall Tower Observatory), measured by SMPS (Scanning Mobility Particle Sizer). It can be clearly observed an almost linear relationship between Black Carbon and aerosol particles greater than 80 nm. This behavior justifies the utilization of Black Carbon concentrations as proxy of CCN concentration. Similar results were obtained during the GoAmazon with the G1 airplane and the HALO ACRIDICON-CHUVA campaign.



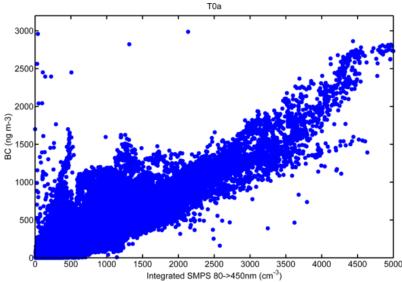


Figure 1 – Scatterplot between Black Carbon Concentrations and aerosol particles greater than 80 nm from TOAA for the entire year of 2014.

References:

Bevan, S. L., P. R. J. North, W. M. F. Grey, S. O. Los, and S. E. Plummer (2009), Impact of atmospheric aerosol from biomass burning on Amazon dry-season drought, J. Geophys. Res., 114, D09204, doi:10.1029/2008JD011112.

Andrea, S. D., Hakkinen S. S. K., Westervelt, D. M., Kuang, C., Levin, E. J. T., Kanawade, V. P., Leaitch, W. R., Spracklen, D. V., Riipinen, I, Pierce, J. R. Understanding global secondary organic aerosol amount and size-resolved condensational behavior. Atmos. Chem. Phys., 13, 11519–11534, 2013. doi:10.5194/acp-13-11519-2013.

- 197 Liu, J., Li, Z. Estimation of cloud condensation nuclei concentration from aerosol
- optical quantities: influential factors and uncertainties Atmos. Chem. Phys., 14, 471–
- 199 483, 2014. doi:10.5194/acp-14-471-2014
- 200 2. The language of the manuscript was revised by a specialist (American Journal
- 201 Experts). We hope all issues have been covered. The certificate is as follows:



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- 3. Following the Reviewer suggestions, the results are separated and now presented
 in two distinct sections within the chapter 4. In this version, the Chapter 4 The
- effect of instability on the rainfall-aerosol relationship was split in two sections:
- 207 Section 4.1 Observational evidence and Section 4.2 Discussion of the possible
- 208 physical mechanisms.

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4. The conclusions were modified with eliminating the speculative part

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Specific issues:

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- **1.** The affiliation numbering was corrected in the manuscript. Affiliation was
- 215 updated.
- 216 **2.** The abstract was submitted in a separated ".pdf" file as asked by the editorial
- support by email. However, to avoid this kind of problem, in this submission the
- abstract and the complete manuscript are now in the same file.

- **3.** The text was modified.
- 4. In Line 71 the following discussion describes the IN effect on deep convective
- 221 clouds. Follow bellow this part of the text (from line 71 to 84)

- 223 "In addition to the influence of BBA on warm rain, ice-phase cloud development is affected in
- 224 polluted environments. In fact, laboratory measurements indicate a high capacity for ice
- nucleation by BBA (Petters et al., 2009). In recent years, some studies have suggested that ice-
- phase clouds are invigorated by the presence of aerosols from vegetation fires (Andreae et al.,
- 227 2004; Lin et al., 2006; Rosenfeld et al., 2008; Altaratz et al., 2010; Koren et al., 2012; Storer and
- Heever, 2013). Rosenfeld et al. (2008) have proposed a conceptual model based on the effect of
- aerosols on deep convective cells, one that is mainly associated to the microphysical effect.
- 230 According to the authors, due to the high concentration of aerosols in polluted environments,
- 231 raindrop nucleation would be slower than in unpolluted areas. This process delays the initiation
- of precipitation, leading the droplets and aerosols to ascend into the atmosphere and reach the
- frozen layer. In addition, these droplets and aerosols could act as ice nuclei and release latent
- 234 heat, which could increase the updrafts and invigorate the cloud dynamics (Lin at al. 2006;
- 235 Rosenfeld et al. 2008). However, even with this evidence, the cloud invigoration process by
- aerosols still needs to be better understood (Altaratz et al., 2014)."
- 5. As suggested, the reference: Nober et al., (2003) was removed from the
- 238 manuscript and new references related to cloud microphysics were added.
- **6.** We did intent that Diehl et al. (2007) is a primary reference on studying the ice in
- the rain process. However, the authors studied the effect of BBA on ice clouds. We
- agree that this sentence is a bit confusing and it was rewritten (the cited reference
- 242 was removed from the manuscript).
- **7.** The description was modified.
- **8.** The sentence was removed to make this point clear.
- **9.** Atmospheric soundings are operationally released at 00 and 12 UTC that do not
- correspond to the afternoon maximum convective activity. Therefore, we evaluated
- 247 if the available soundings, at these times, were representative of the atmosphere
- 248 instability at the initiation time of the convection. Figure 2 presents the CAPE
- 249 histograms employed in this research. It was observed that at the 00 UTC, histogram
- 250 presents a high occurrence of high CAPE values (>2600 J/kg). It is also noted that
- even less frequent, high CAPE values also occurs in the morning (08:00 LST 12
- UTC). This behavior could be explained by the fact that we observed from the
- soundings dataset that more than 70% of the days which presented CAPE values

higher than 2600 J/kg at 00 UTC high values were also identified in the next morning. Then, even not having atmospheric soundings for the afternoon, soundings for 00 and 12 UTC presented a considerable occurrence of high CAPE values, allowing us to use the available atmospheric soundings in this research. We have change the sentence in the manuscript for a better understanding as follows:

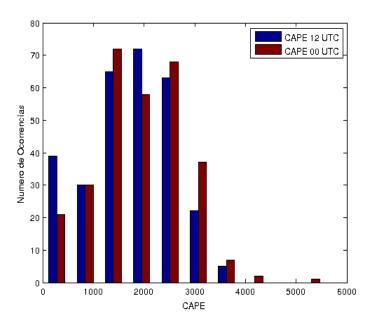


Figure 2 – Histrograms of CAPE values for 00 and 12 UTC in Manaus for the entire year of 2009.

- **10.** We agree that it seems very unlikely that BC regulates the occurrence of deep convection. However, in an environment favorable to deep convection development (high unstable atmospheres), the results presented in this research indicate the invigoration of the pre-existing ice phase clouds. We agree that the way which the sentence was written does not lead to this conclusion. Then, the sentence was modified.
- **11.** The sentence was rewritten. Actually, the semi-direct effect was described in details in the Introduction. Therefore we just recall the importance in this effect in this part of the text.
- **12.** The rainfall and ice indexes were better described in the updated version.
- 13. In most of the sentences the word upslope was substituted by "elevated regions"or "elevated regions". Just the following sentence it was kept:
- "This suggests that in the absence of a large scale circulation that could support the precipitation, the upslope triggering plays an important role in the formation of rain cells."

- **14.** The last sentence ("Thus, this area should receive special attention under forest
- 275 protection policies.") was removed from the manuscript as we agree that it was out
- of the context.
- **15.** We agree that this sentence could lead to misinterpretations of the main results.
- 278 The main results of this research are within the dry period, when we analyzed the
- 279 differences of the precipitation characteristics in less and more unstable
- atmospheres. So, we have decided to exclude this sentence from the manuscript, as
- the main results are not related to comparisons between wet and dry seasons. The
- specification of that is throughout the text.

- **16.** This slight increase which we comment in the manuscript is not statistically
- significant. However, at this point, this sentence shows one of the reasons why the
- instability degree of the atmosphere could be modulating the aerosol effect on
- precipitation. In the right next bin (greater than 1600 ng/m³) the mean rain fraction
- is even greater. We understand that this could be explicit in the manuscript. So, the
- 289 sentence was modified.
- 290 17. We have moved the discussions of the possible physical mechanisms to the
- section 4.2. We agree that wet scavenging could be present in the results, mainly for
- low BC concentrations. The following original text is in agreement with that.
- 293 "The wet scavenging process, an important component responsible for the removal
- of aerosol in the atmosphere, could also contribute to the results observed, mainly
- for lower BC concentrations, at which the RF and IF reach their higher values.
- **18.** You are right; the sentence was corrected.
- **19.** The rain events which occurred exactly over the EUCAARI site were eliminated.
- In our database the time shift between the EUCAARI and radar measurements is less
- than then five minutes. However, even being small we agree that the effectiveness
- of this test will depend on the mixing time-scales, as you propose. Then, the
- 301 consideration of the time-scales difference was added to the manuscript.
- **20.** The high values of RF in polluted atmospheres were observed in more unstable
- 303 atmospheres. This suggests that the great RF values are linked to more intense
- precipitating systems. Despite this evidence, we agree that the analysis of a greater
- value of RF does not necessarily relates to more intense convection. The IF analysis
- were made in order to give support to these results. From the IF analysis, the
- presence of ice in the precipitating systems is greater in polluted and more unstable
- 308 atmospheres than for the unpolluted cases. This result is an evidence of the
- 309 convective invigoration, with more intense updrafts. Maybe the way that RF and IF
- were described in the manuscript did not clarify this aspect. In the current version,
- the text was modified and the link between RF and IF analysis was improved.

- **21.** As commented before, in the previous questions, a new section was created (4.2)
- Discussion of the possible physical mechanisms). There, we discuss the possible
- 315 physical mechanisms related to our results. In addition, we agree that: "Also, the
- effect of absorption is not limited to droplet evaporation. It could be an efficient
- mechanism to maintain convective inhibition for certain stratifications." We have
- 318 started this explanation saying that: "The radiative effect, which also acts to stabilize
- 319 the atmosphere ..."

- 321 **22.** This sentence was moved to the section 4.2 (Discussion Possible Physical
- Mechanisms) as we could not measure the droplet distribution.
- **23.** In the wet scavenging test explanation, we have commented that our screening
- 324 test did not account for the elimination of precipitation in regions outside the
- 325 EUCAARI measuring site. This explanation is presented in the manuscript. Probably,
- small RF (<2%) are within the cases which our test could not detect. We agree that
- this probability should be clarified in the manuscript. The sentence was modified.
- 328 **24.** Figure 2 (in the manuscript) presents the frequency histograms of radar
- reflectivity for different topography elevations for the rainy and dry seasons. In this
- figure we observe that most precipitation (radar reflectivity greater than 20 dBZ)
- occur over elevated regions. However, we agree that the sentence specified that
- convection is observed only over those regions. Then, this sentence was modified.
- **25.** We do agree that some of the explanations which are presented in this chapter
- could not be proved by our results. Then, as previously commented, this chapter was
- divided in two distinct sections: 4.1 Observational Evidences and 4.2 Discussion of
- the possible physical mechanisms. The discussions of the possible effects of biomass
- burning aerosols on stratiform and convective clouds are an example. These
- discussions were moved to the section 4.2.
- We do not agree that one of the results (rain fraction or ice fraction) should be
- excluded from the manuscript. One result complements the other. The results
- showed, for example, that in high unstable cases there is an increase of rain fraction
- under polluted conditions. As commented in the manuscript, unstable cases are
- more likely to favor the development of ice phase clouds. Then, this was the first
- evidence, by our dataset, that convective clouds are probably invigorated by aerosol
- loading. However, just with the rain fraction/black carbon relationship, the
- presence of ice phase clouds could not be proved. Then, we made the same analysis
- using the ice fraction. By this analysis we could check that our supposition was
- correct. In other words, in polluted and high unstable atmospheres, the ice phase
- 349 clouds are invigorated.

- Nevertheless, as the original chapter was divided in two distinct sections, we tried
- 351 to improve the explanations in order to clarify that the RF and IF analysis are
- extremely important to be together in the manuscript.
- **26.** We agree that we could not give more evidences when we discussed the possible
- mechanisms which are linked to the BC influence on the size of the precipitating
- cells. In our discussion we propose the entrainment as the principal mechanism to
- influence the growth of the precipitating systems in polluted and more unstable
- 357 atmospheres. This explanation should not be in our results, as we do not have
- measures of entrainment, for example. Then, it was moved to the new section 4.2
- 359 (Discussion of the possible physical mechanisms). Koren et al. (2012) observed
- 360 evidences of convective invigoration of intense precipitating systems occurring
- under polluted condition. In addition to these results, the authors also observed that
- the precipitating cells which occurred in polluted areas have their coverage area
- increased in around 20 %. This reference was also added to this explanation in the
- manuscript, in order to give support to our results.

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III) Author's Changes in Manuscript

General Issues

- **1.** The manuscript was changed to make this point clear as follow:
- "Notably, BC is a byproduct of the partial combustion of fossil fuels or a remnant from
- 370 wildfires (Ahmed et al., 2014), and it represents only approximately 5 % of the total
- 371 carbon concentration resulting from biomass burning (Formenti et al., 2001;
- 372 Grahamet al., 2003; Cozic et al., 2008). However, BBA dominates the aerosol
- concentration in the Amazon basin, allowing for the use of BC as an aerosol tracer. BC
- 374 concentration is well correlated with aerosol concentration in Amazon. Several studies
- in Amazonia have demonstrated this relationship. Bevan et al. (2009) used a 13-year
- time series of Along Track Scanning Radiometer (ATSR)-derived aerosol optical depth
- 377 (AOD) measurements to examine the role of aerosol in the interaction with biomass
- 378 burning over the Amazon. AOD has a significant positive correlation with the total
- 379 number of satellite-observed fires. Additionally, numerous studies have used aerosol
- 380 particles greater than 80 nm as a proxy for cloud condensation nuclei (CCN). Andrea
- et al. (2008) and Liu and Li (2014) commented on the positive relationship between
- 382 aerosol concentrations and CCN."
- **2.** The entire text was corrected by an specialist.
- **3.** The manuscript's structure was modified
- **4.** The conclusions were modified with eliminating the speculative part as follows:

"This study evaluates the relationship between precipitation and BC concentration using data from one year of ground observations. The results presented are innovative and independent; most prior studies have relied on satellite remote sensing. The methodology using observational data presented herein may contribute to the knowledge of the BBA effect on precipitating systems in the Amazon basin. One of the greatest difficulties regarding this issue has been filtering the aerosol effect from other important atmospheric features. Large-scale circulation or thermodynamic effects represent a major component underlying the strengthening of convection. An analysis of the contribution of each effect could not be performed observationally, but through theoretical simulations that are not completely parameterized. In this study, CAPE values were used as the atmospheric filtering component, which allowed us to divide analyses according to the degree of atmospheric stability. Important features, such as wet scavenging, synoptic scale influence, and droplet size distribution characteristics, need further study and improvement to extend this result. As BBA is predominant in the Amazon basin, BC was used as an aerosol tracer. Nevertheless, other types of aerosol are also present in the region and should receive more attention in new field campaigns. The El Niño configuration, as was observed during the dry season, is associated with reduced levels of precipitation and a decrease in the occurrence of rain cells. Even if this situation had decreased the rain cell population used to study the lifetime duration, a significant number of samples were analyzed for the evaluation of the aerosol-rainfall interaction, and this did not compromise the main results of this study that are associated with the convective scale.

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Despite the limitations of the database and the large set of independent variables, the results presented in this study were statistically significant and physically relevant. BBA releases into the atmosphere generally appear to contribute to a decrease in precipitation. It has been difficult to define the main factor responsible for this behavior because there are several effects, such as wet scavenging or atmosphere inhibitions, which cannot be excluded from the results and could also contribute to precipitation reduction. Nevertheless, we postulate a probable physical mechanism that could explain the observed results as follows. The decrease of RF and IF could be associated to the warm rain suppression mechanism linked to the radiative effect, or to an association of the radiative and microphysical effects together. The most important result obtained in this research was the difference in the rain features

analyzed during the dry season for less and more unstable atmospheres. The convective invigoration of polluted atmospheres was only observed in more unstable atmospheres. This appears to be a considerably significant result, based on the fact that the wet scavenging process acts in the opposite direction, reducing precipitation. The wet scavenging appears to be of a second order in the precipitation-aerosol relationship for elevated concentrations of BC. However, it was only possible to obtain a qualitative result because it was not possible to isolate this process for the precipitation inhibition cases and quantify the exact effect on the rain and ice fractions. Based on the results, we could again postulate a probable physical mechanism which could explain the observed behavior. We observed that during the dry season, most convection occurs in elevated areas. Thus, in more unstable cases, stronger updrafts inside the rain cells initiated over those elevated regions probably carry a greater quantity of droplets formed in a polluted environment to high tropospheric levels, producing in the clouds changes related to their microphysical properties, dynamics and thermodynamics. We were unable to measure the quantity of droplets formed due to the microphysical effect, and the vertical velocity within the precipitating systems was not available in the database used. However, the vertical velocity can be directly linked to CAPE values because the greater the atmospheric instability is, the stronger the updrafts are. This study does not define any specific BC concentration that could activate the cloud process, possibly increasing convective strengthening. Nevertheless, it has been shown that this process only occurs significantly when the BC concentration is higher than 1200 ng m-3.

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The indication of the influence of BBA on the size of the rain cells followed the same behavior observed for RF and IF. We again suggest that the effect is modulated by the atmospheric degree of instability. An important size threshold was found, and the relationship between BC concentration and rain cells area seems to depend on it. The influence of BBA on convective strengthening was observed for large rain cells. This effect is probably related to the reduced entrainment of dry air parcels into the convection, favoring a higher level of neutral buoyancy. The area increase was only observed for systems larger than 100 km2 for the more unstable cases in the dry period. Although the analysis of the influence of BBA on the longevity of the rain cells has been inconclusive, some evidence of this relationship should be mentioned. It is well known that the size of rain cells is positively correlated to their longevity. Thus, the results

- 453 presented in this study could be an indication that high concentrations of BC could lead
- 454 to longer lifetime rain cells, depending on the atmospheric degree of instability."

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Specific issues:

- **1.** The correct affiliation:
- 458 "W. A. Gonçalves1, L. A. T. Machado1 and P-E, Kirstetter2
- 459 [1]{ National Institute for Space Research-INPE/ Center for Weather Forecasting
- and Climate Studies-CPTEC, Cachoeira Paulista, SP, Brazil}
- 461 [2]{Advanced Radar Research Center/ University of Oklahoma and NOAA National
- 462 Severe Storm Laboratory, Oklahoma, United States}
- 463 Correspondence to: W. A. Gonçalves (goncalves.weber@gmail.com)"
- **2.** Following is the abstract:
- 465 "Understanding the influence of biomass burning aerosol on clouds and precipitation
- 466 in the Amazon is key to reducing uncertainties in simulations of climate change
- scenarios with regard to deforestation fires. Here, we associate rainfall characteristics
- obtained from an S-Band radar in the Amazon with in situ measurements of biomass
- burning aerosol for the entire year of 2009. The most important results were obtained
- 470 during the dry season (July-December). The results indicate that the influence of
- aerosol on precipitating systems is modulated by the atmospheric degree of instability.
- 472 For less unstable atmospheres, the higher the aerosol concentration is, the lower the
- 473 precipitation is over the region. In contrast, for more unstable cases, higher
- concentrations of black carbon are associated with greater precipitation, increased ice
- 475 content, and larger rain cells; the finding suggests an association with long-lived
- 476 systems. The results presented are statistically significant. However, due to limitations
- 477 imposed by the available dataset, important features such as the contribution of each
- 478 mechanism to the rainfall suppression need further investigation. Regional climate
- 479 model simulations with aircraft and radar measurements would help clarify these
- 480 questions."
- **3.** The text was modified as follows:
- 482 "In recent years, the scientific community has made great efforts to understand the
- 483 effect of aerosols on clouds and precipitation to reduce uncertainties in climate
- 484 prediction (Tao et al., 2012). Two main effects are well documented: radiative or semi-
- 485 direct, and microphysical or indirect effects. The first effect is related to BBA's high
- 486 capacity for absorption in the visible portion of the electromagnetic spectrum (Hansen

- at al. 1997; Ramanathan et al. 2001; Wake, 2012). This absorption could warm the 487 atmosphere (Koren et al., 2004; Randles and Ramaswamy, 2010; Koch and Del Genio, 488 2010; Jacobson, 2014) and produce atmospheric stabilization (Johnson et al. 2004; 489 Koren et al. 2008). The semi-direct effect alters the atmospheric temperature in the 490 491 boundary layer depending on the level at which the aerosol is presented (Randles and 492 Ramaswamy, 2010). Johnson et al. (2004) commented that in cases of absorbing aerosol located above the boundary layer, the effect is the opposite, i.e., that the 493 494 boundary layer suffers a radiative cooling. Boundary layer warming only occurs when the absorbing aerosols are constrained in the low atmosphere. The indirect or 495 microphysical effect is linked to the possibility of BBA particles becoming cloud 496 497 condensation nuclei (Roberts et al. 2001). As a result, it is expected that the quantity of cloud droplets would increase with the particle concentration (Rosenfeld, 1999; 498 Ramanathan et al., 2001; Andreae et al., 2004; Qian et al., 2009)." 499
- 500 New added references:
- Hansen, J., Sato, M., and Ruedy, R.: Radiative forcing and climate response, Journal of
- 502 Geophysical Research, 102(D6), 6831–6864, 1997.
- Johnson, B. T., Shine, K. P., and Forster, P. M.: The semi-direct aerosol effect: Impact
- of absorbing aerosols on marine stratocumulus, Q. J. Roy. Meterol.Soc., 130(599),
- 505 1407–1422, 2004.
- **4.** Here is the original explanation in the manuscript:
- 507 "In addition to the influence of BBA on warm rain, ice-phase cloud development is affected in 508 polluted environments. In fact, laboratory measurements indicate a high capacity for ice 509 nucleation by BBA (Petters et al., 2009). In recent years, some studies have suggested that ice-510 phase clouds are invigorated by the presence of aerosols from vegetation fires (Andreae et al., 511 2004; Lin et al., 2006; Rosenfeld et al., 2008; Altaratz et al., 2010; Koren et al., 2012; Storer and 512 Heever, 2013). Rosenfeld et al. (2008) have proposed a conceptual model based on the effect of 513 aerosols on deep convective cells, one that is mainly associated to the microphysical effect. 514 According to the authors, due to the high concentration of aerosols in polluted environments, 515 raindrop nucleation would be slower than in unpolluted areas. This process delays the initiation 516 of precipitation, leading the droplets and aerosols to ascend into the atmosphere and reach the 517 frozen layer. In addition, these droplets and aerosols could act as ice nuclei and release latent 518 heat, which could increase the updrafts and invigorate the cloud dynamics (Lin at al. 2006;

Rosenfeld et al. 2008). However, even with this evidence, the cloud invigoration process by

520 aerosols still needs to be better understood (Altaratz et al., 2014)."

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5. Follow the new text in the manuscript related to this item:

- "One of the most important issues regarding the analysis of aerosol-cloud interactions is the
- 524 determination of the predominant effect, radiative or microphysical. In warm rain suppression
- 525 conditions, both effects seem to act together. However, the quantification of their respective
- 526 contributions remains an important issue. Warm rain suppression evidence was firstly
- documented by Rosenfeld (1999), and similar results have subsequently been presented in the
- 528 literature, e.g., Koren et al., (2004) and Qian et al., (2009). The physical mechanism suggested
- 529 for warm rain suppression is related to the fact that BBA has the potential to act as cloud
- condensation nuclei. Thus, in polluted environments, a large number of small cloud droplets could
- form (Rosenfeld, 1999; Ramanathan et al., 2001; Andreae et al., 2004; Qian et al., 2009), which
- compromises the coalescence process (Borys at al., 2000; Hudson and Yum, 2001; McFarquhar
- and Heymsfield, 2001; Yum and Hudson, 2002; Borys et al., 2003; Hudson and Myshra, 2007;
- Kaufman et al., 2005). These droplets do not reach the required size to precipitate and can rapidly
- evaporate due to the semi-direct effect (Artaxo et al., 2006). However, as previously mentioned,
- the atmospheric level at which the BBA is located could lead to different results. Even a boundary
- layer cooling is likely to be observed. This could explain some of the controversy in the literature.
- Kaufman et al. (2005) and Brioude et al. (2009), for example, obtained results that differed from
- those demonstrated in other warm rain suppression publications. In these studies, the authors
- observed an increase in stratiform cloud formation in atmospheres with an elevated presence of
- 541 aerosols."

New References related to cloud microphysics

- 543 Hudson, J. G., and S. S. Yum (2001), Maritime-continental drizzle contrasts in small
- 544 cumuli, J. Atmos. Sci., 58, 915–926.
- 545 McFarguhar, G. M., and A. J. Heymsfield (2001), Parameterizations of INDOEX
- 546 microphysical measurements and calculations of cloud susceptibility: Applications for
- 547 *climate studies, J. Geophys. Res., 106, 28,675–28,698.*
- 548 Yum, S. S., and J. G. Hudson (2002), Maritime/continental microphysical contrasts in
- 549 *stratus, Tellus, Ser. A and Ser. B, 54B, 61–73.*
- Hudson, J. G., and S. Mishra (2007), Relationships between CCN and cloud microphysics
- 551 variations in clean maritime air, Geophys. Res. Lett., 34, L16804,
- 552 doi:10.1029/2007GL030044.

- Borys, R. D., D. H. Lowenthal, and D. L. Mitchell (2000), The relationships among cloud
- 554 microphysics, chemistry and precipitation rate in cold mountain clouds, Atmos.
- 555 Environ., 34, 2593-2602.
- Borys, R. D., D. H. Lowenthal, S. A. Cohn, and W. O. J. Brown (2003), Mountaintop and
- radar measurements of anthropogenic aerosol effects on snow growth and snowfall
- rate, Geophys. Res. Lett., 30(10), 1538, doi:10.1029/2002GL016855.

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- **6.** Here is the new explanation:
- "In addition to the influence of BBA on warm rain, ice-phase cloud development is
- 562 affected in polluted environments. In fact, laboratory measurements indicate a high
- 563 capacity for ice nucleation by BBA (Petters et al., 2009). In recent years, some studies
- have suggested that ice-phase clouds are invigorated by the presence of aerosols from
- vegetation fires (Andreae et al., 2004; Lin et al., 2006; Rosenfeld et al., 2008; Altaratz
- 566 et al., 2010; Koren et al., 2012; Storer and Heever, 2013). Rosenfeld et al. (2008) have
- 567 proposed a conceptual model based on the effect of aerosols on deep convective cells,
- one that is mainly associated to the microphysical effect."

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- **7.** The description was modified as follows:
- 571 "Rosenfeld et al. (2008) have proposed a conceptual model based on the effect of
- aerosols on deep convective cells, one that is mainly associated to the microphysical
- 573 effect. According to the authors, due to the high concentration of aerosols in polluted
- 574 environments, raindrop nucleation would be slower than in unpolluted areas. This
- 575 process delays the initiation of precipitation, leading the droplets and aerosols to
- ascend into the atmosphere and reach the frozen layer. In addition, these droplets and
- 577 aerosols could act as ice nuclei and release latent heat, which could increase the
- 578 updrafts and invigorate the cloud dynamics (Lin at al. 2006; Rosenfeld et al. 2008)."

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8. The sentence was removed to make this point clear.

- **9.** We have change the sentence in the manuscript for a better understanding as
- 583 follows:
- "As Manaus radiosondes were released by an operational station, only soundings at
- 585 00:00 and 12:00 UTC, 08:00 and 20:00 local time, were available. The most
- 586 appropriate time for a sounding in this study is approximately noon, when convection
- starts to develop in the area. However, the CAPE dataset was evaluated and shown to

- have substantially useful information and ability to capture daily instability features
 even though the recordings were not made at the most appropriate times. The 08:00
 local time soundings also presented a considerable population of high CAPE values
 (greater than 2600 J/Kg). In addition, more than 70 % of the days that had CAPE
 values higher than 2600 J/Kg at 00:00 UTC presented high CAPE values the next
 morning (as measured at 12:00 UTC)."
- **10.** The sentence was changed as follows:

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- "Therefore, due to its potential for ice nucleation, the presence of BC would favor the invigoration of pre-existing ice-phase clouds, including deep convection."
- 598 **11.** The sentence was rewritten when we recalled the importance of this effect, as follows:
- "As discussed previously, BC also has the capacity to absorb radiation in the visible portion of the electromagnetic spectrum (Ramanathan et al., 2001; Storelymo, 2012; Tiwari et al., 2013; Ahmed et al., 2014). This characteristic could warm the layer where BC is present (Myhre, 2009; Mahowald, 2011; Jones et al., 2011; Wake, 2012; Wang, 2013), which could stabilize the atmosphere and reduce the formation of cumulus clouds. In turn, this characteristic of BC could also affect cloud properties and precipitation indirectly as discussed above."

12. The rainfall and ice indexes were better described in the text as follows:

608 "The three indices described above indicate the spatial distribution of the rain properties in the study domain. The higher their value, the greater the part of the study 609 610 area covered by the physical property analyzed. The RF and IRF indices express how the rain and the intense rain are distributed in the domain according to the respective 611 thresholds of 20 and 45 dBZ, which approximately correspond to rain rates of 1 mmh-612 613 1 and 30 mmh-1, respectively, according to Marshall and Palmer (1948). IF indicates the percentage of the area covered by ice clouds. As previously described, the value of 614 1 mm was chosen from the VPR method to determine IF. Note that the procedure was 615 tested with higher VIC values. However, the number of samples was observed to 616 decrease drastically, leading us to choose the value of 1 mm. Petersen et al. (2005) 617 showed that this value corresponds to the beginning of a relationship between vertical 618 ice content and lightning flash density." 619

- **13.** In most of the sentences the word upslope was substituted by "elevated regions"
- or "elevated regions". Just the following sentence it was kept:
- 623 "This suggests that in the absence of a large scale circulation that could support the
- 624 precipitation, the upslope triggering plays an important role in the formation of rain
- 625 *cells.*"

- **14.** The last sentence ("Thus, this area should receive special attention under forest
- 628 protection policies.") was removed from the manuscript as we agree that it was out
- 629 of the context.

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- **15.** So, we have decided to exclude this sentence from the manuscript, as the main
- 632 results are not related to comparisons between wet and dry seasons. The
- 633 specification of that is throughout the text.
- **16.** We understand that this could be explicit in the manuscript. So, the sentence was
- 635 modified as follows:
- 636 "Beyond this value, the RF slightly increased. Although it was not statistically
- 637 significant, this characteristic led us to attempt a filtering out of the possible aerosol
- 638 influence on precipitation from an atmospheric feature that could modulate this effect.

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- **17.** The following original text is in agreement with that.
- "The wet scavenging process, an important component responsible for the removal
- of aerosol in the atmosphere, could also contribute to the results observed, mainly
- for lower BC concentrations, at which the RF and IF reach their higher values.
- **18.** The sentence was corrected, as follow:
- 645 "As the RF and IF decrease for elevated BC concentrations, the dominant effect could
- 646 be explained by the wet deposition theory and/or the rain suppression theory. In
- 647 addition, the radiative effect acts to increase the population of stable atmosphere cases
- and results in less rainfall for the situation of high aerosol loading.

- **19.** The consideration of the time-scales difference was added to the manuscript as
- 651 follows.

"Thus, the concept of this test was based on eliminating from our statistics all samples for which precipitation was observed over the EUCAARI site. This filtering excluded the samples in which precipitation could have cleaned the atmosphere, throughout the scavenging process. After this, comparisons between the RF and BC concentrations were performed. No significant differences were observed when comparing the results utilizing this criterion with those in which no consideration was given to a potential aerosol wet scavenging effect. This characteristic indicates that the local scavenging effect seems to be of a second order on the BC-rain interaction. However, this test does not take into account the reduction of BC sources outside the measurement site due to rainfall. Therefore, it is not possible to separate the scavenging effect from other physical effects associating reduction of rainfall with the increase in BC concentration, even if the scavenging effect seems to be of a second order, locally. In addition, Gryspeerdt et al. (2015) commented that the time difference between the measurements of rain properties and aerosol could lead to misinterpretations in the aerosol-rainfall relationship, as the timescale of wet scavenging is narrow. In this study, the temporal sampling frequency difference between the EUCAARI and radar measurements is five minutes in the worst case scenario. This sampling time difference, though minor, could have allowed the scavenging effect to remain even with the use of the described test."

New reference

- 672 Gruspeerdt, E., Stier, P., White, B. A., Kipling, Z. Wet scavenging limits the detection
- of aerosol-cloud-precipitation interactions. Atmos. Chem. Phys. Discuss., 15, 6851–
- 674 6886, 2015. doi:10.5194/acpd-15-6851-2015

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- **20.** The text was modified and the link between RF and IF analysis was improved as
- 677 follows:
- 678 "The RF increased in cases where higher concentrations of BC were observed. The
- 679 precipitation appeared to spread over the region when the atmosphere was favorable
- 680 to the development of convection associated with BBA. The distribution of RF for three
- 681 categories of BC (Fig. 4d) shows that the greater the particulate material
- 682 concentration, the more elongated the tail of the RF distributions. As commented in
- 683 Section 2, RF is an indication of the distribution of rain in the study domain. Therefore,
- an increase in values for this variable for polluted atmospheres does not necessarily
- 685 mean that more intense convection is occurring. However, this behavior was observed
- 686 when the atmosphere presented conditions for intense convection. Consequently, this
- 687 result could be an indication that convection is invigorated by higher BBA
- concentrations (Lin et al., 2006; Graf, 2004; Rosenfeld et al., 2008; Altaratz et al., 2010;

- Koren et al., 2012) when the atmosphere is highly unstable (i.e., presenting high CAPE
- 690 values).
- 691 An analysis evaluating the presence of ice in the precipitating cells within polluted
- 692 atmospheres has the potential to give support to the previously described evidence.
- Therefore, to understand whether the amount of cloud ice is influenced by the presence
- of high BBA concentrations, the IF index was calculated based on Eq. 3. This procedure
- 695 was important to account for the fact that ice formation could be influenced by BC
- 696 (Demott et al., 1999; Cozic et al., 2008; Kireeva et al., 2009). In addition to the
- 697 convective case hypothesis, we verified whether the precipitation suppression observed
- 698 for the less unstable case could link to the presence of stratiform clouds. The mean IF
- values decreases substantially in proportion to the increase in BC concentration for
- less unstable atmospheres (Fig. 5a). This result could be an indication that stratiform
- 701 clouds are negatively influenced by BBA. In more unstable cases, the result indicates
- that the convection invigoration hypothesis (Rosenfeld et al., 2008) based on the
- 703 presence of aerosols is likely to be true (Fig. 5b), in agreement with the RF-BC
- 704 relationship previously indicated."
- 706 **21.** No modification
- 707 **22** This sentence was moved to the section 4.2 (Discussion Possible Physical
- 708 Mechanisms) as we could not measure the droplet distributionas follows:
- 709 "Probably due to the high instability (high updraft), the large number of small droplets
- 710 inside clouds (formed by the microphysical effect) ascend very fast, thereby reducing
- 711 the extent of evaporation. Rosenfeld et al. (2008) commented that an unstable
- 712 atmosphere could carry the small droplets to higher atmospheric levels, invigorating
- 713 convection and increasing the amount of ice."
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- 715 **23.** We did not modify the text with regards to the first part of this question, as
- 716 follows:
- "However, this test does not take into account the reduction of BC sources outside
- 718 the measurement site due to rainfall. Therefore, it is not possible to separate the
- scavenging effect from other physical effects associating reduction of rainfall with
- the increase in BC concentration, even if the scavenging effect seems to be of a
- 721 second order, locally.
- A new sentence clarifying the cases with RF smaller than 2% was added:

"Although impossible to quantify, the wet scavenging process also seems to be of second order. It would act in the opposite direction through the fact that RF and IF are higher for polluted atmospheres (Figs. 4b and 5b). Notably, however, it is possible that for small IF and RF values, the effect of the wet scavenging process is still present in the statistical analyses. In these cases, the applied wet scavenging test may not have been able to identify this process."

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- 730 **24.** This sentence was modified as follows.
- "During the dry season, most of the precipitation is associated with elevated regions,
- and in the more unstable cases, it appears that BBA helps to increase ice nuclei
- 733 *formation and precipitation.*"
- 734 **25.** The new explanation in the manuscript:
- 735 "During the dry season, in a more unstable condition, our results indicate an invigoration of the precipitation with increasing BC concentration. Considering that 736 high CAPE values are associated with stronger updrafts, the aerosol effect on the 737 738 rainfall and on the severity of the convective processes could depend on the intensity 739 of the vertical motions. In addition, it is important to note that during the dry season, 740 only elevated regions trigger convection, which could support the intensification of the updrafts in more unstable atmospheres. The radiative effect, which also acts to 741 742 stabilize the atmosphere, is probably of a second order. Even with high levels of BBA, the atmosphere is highly unstable, and thermodynamics on this time scale seem to 743 dominate over the radiative process. Additionally, an increase in the droplet 744 745 evaporation process, which could be generated by the radiative effect, does not seem 746 to be the predominant mechanism. Probably due to the high instability (high updraft), the large number of small droplets inside clouds (formed by the microphysical effect) 747 748 ascend very fast, thereby reducing the extent of evaporation. Rosenfeld et al. (2008) commented that an unstable atmosphere could carry the small droplets to higher 749 atmospheric levels, invigorating convection and increasing the amount of ice. 750 Moreover, BC particles could be carried within the updrafts and act as ice nuclei. 751 752 During the dry season, most of the precipitation is associated with elevated regions, 753 and in the more unstable cases, it appears that BBA helps to increase ice nuclei 754 formation and precipitation. Although impossible to quantify, the wet scavenging 755 process also seems to be of second order. It would act in the opposite direction through the fact that RF and IF are higher for polluted atmospheres (Figs. 4b and 5b)." 756

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"In addition, it is important to note that Koren et al. (2012) observed that rain cells occurring under polluted conditions had their coverage area increased by approximately 20 %, which also lends support to the results presented in this research."

- 764 Reference to this question:
- 765 Koren, I., Altaratz, O., Remer, L. A., Feingold, G., Martin, V.: Aerosol-induced
- 766 intensification of rain from the tropics to the mid-latitudes, Nature Geoscience,
- 767 5,118-122, 2012.