

The authors present an interesting study on the resulting emission and exponential decay of bioaerosols after heavy rainfall during case studies in Australia. Although the concept and results are quite interesting and are worthy to be cyclod in the peer-reviewed literature, the manuscript suffers from several issues that need to be addressed before it is suitable for publication. In general, the authors do not provide enough details on their methods or explanation of the events and conclusions presented. Overall, the paper was difficult to follow because it severely lacked discussion on the methods and results, the structure and order of the sections interrupted the overall flow, and the figures did not represent the conclusions made in the text. This research has great potential to be published, however, major modifications are necessary in order to accept into ACP. In order to be acceptable for publication, the authors should address the comments below.

General comments:

1. The concept of “after and before” or “after-before” is somewhat nebulous. After reading deeper into the paper, it is evident the authors are referring to the IN concentrations *after* a heavy event *as compared to before*. Perhaps the wording throughout should be changed to simply “after” heavy precipitation or key day and detail that these concentrations are scaled to concentrations before the heavy precipitation or key day.
2. Throughout the manuscript, ice nuclei, ice nucleation, and IN are used interchangeably. Please define ice nuclei as “IN” and use throughout. Also, can use IN-active instead of ice nucleation active. This is generally consistent with what is in the literature.
3. Please be consistent with terminology: are the authors suggesting microorganisms and bioparticles are the same thing? If so, eliminate the use of bioparticles and use microorganisms. Include the definition of airborne microorganisms as bioaerosols in the context of this paper.
4. There seems to be an ongoing issue with ambiguity throughout the manuscript. For instance, on p 22507 lines 5-8: “The importance of these different factors is most likely specific to different sites and seasons, therefore it is important to identify regions and periods of time where rainfall occurs in positive or negative feedback cycles in order to elucidate the details of the underlying processes.” What factors, the aforementioned scenarios? What sorts of sites and seasons would determine the importance of these factors? What are the positive or negative feedback loops? Do the authors mean the enhancement or suppression of precipitation? What underlying processes?

If the authors add more detailed background in the introduction and revise the manuscript to not use vague definitions such as “the process” or “the factors” (as examples, in introduction), “the experiment” (p 25510, line 5), “these were” (p 25508, line 6), or “it” (p 25508, lines 18 and 22) it would be much clearer what the authors are attempting to state. The authors should avoid using words such as “these” alone as on p 25508, line 6 and use the actual word, such as “key days” or “these key days” in this case.

5. There is no mention of statistical significance or error calculations. All data presented should undergo both of these calculations and error bars should be shown in the figures, particularly for Figs 1-5, 8, and 9.

6. I can see how the various sections are related, however as written they seem disconnected. The results jump from IN case studies to long-term rainfall statistics to anthropogenic influences on rainfall. Further, there is a separate focuses on spatial variability. The sections should be better linked together to follow the overarching context of emission of bioaerosols after heavy rainfall, and the resulting impacts of bioaerosols on IN concentrations and subsequent rainfall.
7. Overall, the link between IN and rainfall trends to bioaerosols is weak. Because actual measurements of bioaerosols were not conducted, the authors should take extra care and time discussing this potential link. This would be a speculation, which is not emphasized in in the paper; the way it is written, it sounds like bioaerosols were in some way measured. Please elaborate on this speculation and what clues lead one to believe the IN were likely biological in origin.

Specific Comments:

Abstract:

8. The abstract should open with a few sentences of introductory material. For instance, describing the importance of bioaerosols as IN on precipitation and why this is important. Or introduce the concept of the bioprecipitation feedback loop, i.e., microorganisms are emitted into the atmosphere in response to heavy rainfall, these airborne microorganisms or “bioaerosols” then can induce further precipitation by serving as IN.
9. The authors state that four areas of Australia are examined, but only two are presented in the manuscript (south-west and south-east). Please revise.
10. It is unclear where “> 92 years” is from; 92 years of data are not presented in the manuscript. From what I can see, specific case studies are presented from 22 Dec 1956 (Fig 1), and 18 months to 3 years (unclear which months but from 1987 – 1990, Fig 2 and 3). The dates are not provided in the figures for the  $F_Q$  or  $F_v$  so it is very unclear where the 92 years of rainfall records originates from. How did the authors calculate 92 years? Perhaps the sites do have a total of 92 years of rainfall records, but all 92 years are not presented thus should not be presented as such in the abstract.
11. Along these lines, if there are 92 years of rainfall records, how long are the records for IN concentrations? It would be helpful for the authors to provide the actual number of years of both rainfall and IN records used in their analysis.
12. p 25504, line 6: How is there an increase in rain on the day following heavy rain? Would this not be included in the storm with the heavy rain? It would be more clear if the authors discussed the impulsive increase then subsequent exponential decrease in IN concentrations separately from the effects of the emissions of IN on rainfall after the key days.
13. p 25504, line 8: “The similarity of ice nucleus concentrations, bacterial populations, bioaerosols...” Bacterial populations and bioaerosols were not measured, thus this should not be a main conclusion but rather a speculation as to what the IN were.

14. How can a cumulative difference be calculated for rainfall amount? Was there substantial rainfall before the key days? What if there was not any rainfall before the key day, then the after value of “x” would be divided by zero...
15. p 25504, lines 12-14: “Comparison of cumulative totals pre-1960 and post-1960 showed differences bearing apparent relations to upwind coal-fired power stations, growth of metropolitan areas and increased areas of cultivation of wheat.” This is vague. What kind of differences? Need to clarify in the abstract.
16. Also, coal-fired power stations and metropolitan areas are not typically associated with sources of bioaerosols. It is unclear in the abstract how this related to the bioaerosol theme. The authors should briefly describe the results from the pre-1690 to post-1960 analysis in the abstract, and how there was a higher frequency of rainfall prior to these infrastructural developments, i.e., when the land was a larger source of bioaerosols. This would elucidate why these sources are mentioned in the abstract.
17. p 25504, line 17: “...interactions between micro-organisms, bioaerosols, and rainfall...” First, IN concentrations should be mentioned here, since that is one of the measurements presented. Second, aren’t microorganisms a type of bioaerosol when airborne? The authors should clarify that they mean microorganisms on plants (i.e., not their airborne form as bioaerosols).

Introduction:

18. p 25504, line 23: Please define Bowen ratio.
19. The beginning of the introduction would flow better if the authors started with discussing the global scale analysis, then with the smaller scale short-term positive feedbacks. This provides a better segue from the more general points to the more specific.
20. p 25505, lines 22-23: “Growth of vegetation that harbors ice nucleation active microorganisms such as *P. syringae* is stimulated by rain and would provide an increased habitat prolonging the initial stimulation.” This sentence needs to be restructured to something like, “Growth of vegetation that harbors ice nucleation active microorganisms such as *P. syringae* is stimulated by rain, thus would provide an increased habitat prolonging the initial stimulation of X.” The authors also need to define the stimulation of what (bacteria?) and what is meant by stimulation (release? multiplication?).
21. It would be helpful if the authors added a sentence in the introduction on the aerosolization process for microorganisms after rain, is it due to the drying mechanism, growth and release from leaf surface turbulence, etc.?
22. p 25506, line 8: “Spectacular but fluctuating increase” is very qualitative, what is the value for spectacular? It would also be helpful to provide standard atmospheric IN concentrations for comparison, to show that the value the authors are referring to is actually spectacular.
23. p 25506, lines 10-15: How did the authors eliminate influences from meteorological conditions on the increased rainfall after a heavy rainfall? Surely the atmospheric conditions had something to do with the increased rainfall, and not the IN emission alone. Could the authors

please evaluate how meteorological changes were accounted for in influencing rainfall frequency and quantity?

24. The authors should provide a discussion on the location of the sites selected for analysis. What sites are in more remote areas with vegetation? Which are in rural or urban areas? Do the trends in IN concentrations and rainfall differ at these locations? Perhaps the analysis should include a separation of sites into categories based on their surrounding terrain (i.e., vegetation versus rural and urban development).
25. p 25506, line 17: CCN, GCCN, and IN are introduced. It would be beneficial for the authors to elaborate on the effects these species alone and combined have on precipitation quantity in the introduction. For instance, a cloud containing a high concentration of CCN would inhibit precipitation, whereas a cloud containing some IN and GCCN would increase precipitation formation. In cases with IN and high concentrations of CCN, riming and thus precipitation formation efficiency is restricted. High concentrations of IN might not enhance precipitation (under non-Hallet-Mossop conditions) due to competition of the ice crystals for water vapor and reduced riming. There is no need to go into gory detail, but some examples could be provided.
26. p 25506 starting at line 24: The material in this paragraph would be more suitable earlier on in the introduction. In general the introduction could use some restructuring to enable it to flow better. Perhaps something along these lines: first discuss the bioprecipitation feedback cycle, then lead into under what conditions bioaerosols can serve as different cloud nuclei (CCN, IN, GCCN) and how these different classes of cloud nuclei affect precipitation formation in different scenarios (CCN alone, IN with CCN, IN with GCCN, etc.), then finish the introduction with the summary paragraph of the results presented in the manuscript (the current last paragraph in the introduction). Most of the material to structure the introduction in this manner is already presented, it just needs to be reordered with some additional discussion.
27. There is a lot of mention of “feedback loops” but nowhere in the introduction are these feedback loops clearly laid out. The subsequent mentioning of the feedback loops throughout the manuscript would be much clearer if the authors define in the introduction the feedback loops they are referring to. The authors do hint at the feedback loops, but it could be more distinctly presented.

#### Methods:

28. The methods section could also benefit from restructuring. Here is how I propose the authors restructure the methods section:
  - a. Site locations and dates: The authors should present the sites before anything else (as is, the sites are very briefly mentioned to originate from the Australian Bureau of Meteorology at the very end of the methods). What sites were chosen, all that are available through the ABM? What are the records of the data and which years are actually used (not all 92 years)? What years and sites have rainfall measurements? Of those, what years and sites have IN measurements used in the manuscript? A new figure should be added which includes a map showing the locations of all sites, with

colored/shaped markers for sites with rainfall measurements only and another colored/shaped marker for sites with colocated rainfall *and* IN measurements. Also, a visual of the topography/terrain overlap on the map would be helpful, with the urban and coal-fired plant areas clearly labeled. A map and initial section in the methods such as this one described would set the entire stage for where and when samples were collected. It might also be useful to provide some sort of table elucidating the date ranges (day/month/year) of data used for the authors' analysis. As is, there are specific case studies presented in the following sections. The authors should clarify why these specific case studies were chosen, and can provide this information in the methods.

- b. Thoroughly describe the instrumentation and methods used for the rainfall amounts and IN concentration measurements. Are each of these measurements hourly? Daily? As far as I can tell, they are mean daily values as shown in figures later on, but is the daily mean based on hourly measurements? If so, what is the standard deviation of each of these daily means (any figures presenting daily mean values should have error bars)? This should be specific up front. What data were missing?
- c. Then, finish with the data manipulation section. It is difficult to understand why the data is being manipulated when the data sources have not yet been presented. It is unclear as to why and how the authors chose to calculate CD and error analysis is not presented. It should also be noted that this is for IN concentrations only. Perhaps rename the subsection to "IN data manipulation" and create a separate subsection for "Rainfall data manipulation".

29. p 25508, line 1: Please briefly define the epoch method.

30. p 25508, line 4: What is the value for the threshold used? How was this threshold chosen, particularly if it varies for different days and sites? How many key days were there out of total days investigated? How many sites had key days? This section severely lacks statistical representation of the key days and thresholds used.

31. p 25508, line 7: How was the given number of days determined and why are they different for the case studies presented (in Fig 1 it is 30 days but it is 20 days for the remaining cases)?

32. The descriptions of sites, key days, dates, before values of rainfall and IN, number of days after, and rainfall thresholds are vague and thus the cases presented appear "cherry picked". It would help immensely if the authors defined and thoroughly discussed all these parameters and why specific cases were chosen. Perhaps it would be beneficial to include and first present a figure summarizing the rainfall and IN data over the entire time period of the records, then state that specific case studies will be presented and why, then elaborate on these case studies.

33. p 25508, lines 6-8: "These were transformed into time series of differences of those values on a given number of days after a key day to those on the same number of days before it." This sentence is quite vague and difficult to follow, please be specific with the method of choosing the number of after days.

34. p 25508: The CD method is very unclear. Is this for IN concentration, rainfall, or both? I am guessing it is only for IN? The paragraph on this page starts with IN, then shifts to rainfall.

What is the “before” average based on? The average of IN concentrations from the 20 days before each event at each site (specific to the event) or over all data? The authors conclude that IN concentrations initially increase, then exponentially decrease, but this is very difficult to see using the CD calculations. It is very difficult to discern the relationship of the IN concentrations after key days to the average IN concentration before the key day. Perhaps the authors should show a separate line in each of the figures for the mean daily IN concentrations 20 days after key days and another line for the mean daily IN concentrations 20 days before the key day.

35. Please provide units for any data introduced in the methods. Are all IN concentrations in  $\text{m}^{-3}$ ? What are the units for rainfall and rainfall rate ( $\text{mm}$  and  $\text{mm h}^{-1}$ , respectively)? Using “rainfall” alone is quite vague.
36. The rainfall maximum bias analysis is unclear. What is the purpose of this? It does not seem to be used anywhere in the results or discussion, besides Fig 5 which as I comment on below, should be separated into trend lines per season.
37. p 22509, line 1: More details should be provided on the IN concentration records. How far back do they go? At all sites? Any data interruptions/data not acquired from certain sites during certain dates? How were the samples stored? Any issues with artifacts? Were the samples analyzed at temperatures other than  $-20\text{C}$  or  $-15\text{C}$ ? Why were these temperatures chosen and why are they different (i.e., why not analyze at  $-15\text{C}$  for all sites)? How long after sample collection were the filters analyzed (X-X days after collection)? What sort of sampling instruments were used (i.e., a Hi-Vol, MOUDI, etc.)?
38. How can the IN concentrations be compared from the site with the cloud chamber to the sites with filter collection? The collection methods were different as were the operating temperatures ( $-15$  vs  $-20\text{C}$ ).
39. For the cloud chamber, how were air samples acquired at the site with the chamber? For the filter samples collected at other sites, was the filter simply placed in the cloud chamber then temperature and RH were controlled? What was the material of the filters and were clean filters run in order to establish a baseline of a blank sample?
40. p 25509, line 2: Four sequences of what? This sentence is vague.
41. p 25509, line 6: 300 L per what? 300 L of air pulled total or  $300 \text{ L day}^{-1}$ ?
42. Why were the samples collected at the site with the cloud chamber measured at  $-20\text{C}$ ? At this temperature, both mineral and soil dust can form ice, how did the authors account for this?
43. p 25509, line 7: Please clarify that this procedure was conducted in the cloud chamber.
44. p 25509, lines 13-15: What is meant by “multiple sites”? How many and which ones? There is no reference to the statement on these lines, has this statement been validated? There is no evidence presented that corroborates this statement.
45. For clarification, please distinguish the site with the cloud chamber on the map.

## Results:

46. According to the heading of section 3.1, the cloud chamber is not in the two regions discussed (south-west and south-east). Trying to follow which sites were used and why only two regions are discussed/shown in the figures is difficult. If the authors choose to discuss the spatial differences between the south-west and south-east, then there is no need to discuss the northern sites. However, there is some wavering between using south-west and west and south-east and east. Please be consistent with the regions of focus and eliminate discussion of those sites which are not used in the analysis.
47. p 25509, line 21: What day in “early November”? Also, what is meant by “only a few light falls of rain”? What is the threshold for light to heavy? Please provide some numerical representation of the threshold and what is meant by “a few”.
48. Section 3.1: Why was this particular example case chosen? Do all key days follow this? If not, then why not? Overall, the authors should specify why specific cases were chosen and if there were representative of other key days. Otherwise, it appears as if the cases were “cherry picked”. If the other key days do not follow the same trend as the one presented in this section, could the authors explain why this might be the case?
49. p 25510, line 5: “The experiment was described by Bigg and Miles (1964).” Please elaborate on what is meant by “the experiment” and what the experiment actually was.
50. p 25510, line 6: Here, the authors provide the height of the filter collection (1.5 m AGL), what was the sample collection height at the site with the cloud chamber?
51. Section 3.2: Why were these 24 sites chosen out of the 106 sites in south-eastern Australia as delineated in the abstract? Again, there is an issue with if the focus is on all of eastern or just south-eastern Australia.
52. p 25510, line 11: The threshold for rainfall for a key days is defined at  $\geq 25$  mm. Why do each of the following cases presented have different thresholds? Does each site have a different threshold or is the 25 mm for all 24 sites presented in Section 3.2? It seems as if the authors ambiguously chose certain thresholds without explanation.
53. p 25510, line 18: For better visual representation, please outline Victoria and label Tasmania on the map.
54. p 25510, lines 24-26: It would be helpful to provide the freezing temperature ranges of AgI and dry ice to demonstrate that these species did not interfere with the IN measurements, and why the authors can conclude that the IN concentrations were indeed biological in origin. However, according to Bigg, 2015, AgI can nucleate ice at -15C, so how did the authors account for this discrepancy? How do the authors know any of the IN were biological? Additional discussion provided on this topic is needed.
55. Section 3.4: Please provide some sort of visual representation (i.e., highlight the sites in the map since the reader may not know where Tasmania is) as to which sites were chosen. Also, provide discussion on why these 5 sites were chosen for this case study. Which dates were

chosen? Perhaps a table encompassing each of the dates used for this analysis is warranted. This comment applies to the previous two sections as well for the other case studies.

56. Section 3.5: I am left wondering why specific dates, sites, and thresholds were chosen and why not others. What happened on the other days? Were the set number of sites chosen based on the rainfall thresholds? The results for the IN measurements could perhaps be restructured to present case studies in the eastern (as already done in Section 3.2) and western regions of Australia, which would align nicely with the remaining theme of the paper where rainfall patterns are differentiated in each of these regions. Even though the authors state two of the case studies were in Victoria (i.e., eastern) and Tasmania (also eastern??), it would be better to clarify if these regions are eastern or western Australia. Following on this point, the results presented (except the one site with the cloud chamber in north-eastern Australia) do not encompass northern Australia, why is this? I would suggest presenting a case study from each region and using five domains: northeast, northwest, southeast, southwest, and maybe have Tasmania as its own area. If used, the borders of the domains should be clearly drawn out in a map, so that it is evident how many sites are within each domain.
57. Section 4.1: This section should be included in the methods and clearly differentiated from the IN CD calculations.
58. p 25512, lines 4-5: What are the units for this feedback factor? mm? Why is CD for rainfall discussed when only  $F_v$  and  $F_Q$  are shown? If CD for rainfall is referring to the difference in pre- and post-1960 rainfall, then this needs to be explicitly stated and differentiated from IN CD. This explanation and distinction could easily be laid out in the methods.
59. p 25512, lines 15-16: It seems like a comparison should be made between western and eastern Australia, not western and south-eastern. According to Fig 2a, there are sites in north-eastern Australia (although these are labeled as IN measurement sites, but I am assuming there were rainfall measurements at these sites as well). Why not use these in the analysis or, as delineated, in comment 53, do a comparison of the five different regions?
60. Section 4.3: The authors present trends among certain percentages of the sites, but what about the remaining sites? At only 22% and 60% of the sites discussed, that leaves quite a few sites that did not follow the logarithmic trend. The authors state, “At many of the remaining sites, the logarithmic trend terminated before 20 days.” The discussion section is lacking an explanation on why only specific sites followed the logarithmic trend.
61. p 25513, line 10: “rainfall feedback effect” is vague, as delineated in a previous comment, please discuss the mechanism for this feedback effect.
62. p 25513, line 24: The splitting of the rainfall data at 1960 is quite reasonable due to the developmental changes that occurred around that time. However, unless the authors have data from 1930 – 1990, the data sets were not “divided in half” at 1960. The date ranges should be provided and this sentence should be reworded.
63. p 25513, lines 8-10: The ratio of pre-1960  $F_{hv}$ /post-1960  $F_{hv}$  technically did not decrease itself. Do the authors mean the post-1960  $F_{hv}$  decreased from the pre-1960  $F_{hv}$ ? From Fig 6, it looks like the rainfall frequency decreased over time, which would make sense due to the



anthropogenic influences from the power station complex. Also, the terminology should be consistent (i.e., in the text, it is referred to as pre- and post- $F_{hv}$ , the figure caption has pre- and post-1960, and the figure itself has pre-1960  $F_v$  and post-1960  $F_v$ ).

64. p 25513, lines 21-22: This sentence should be either relocated to the very beginning of this section or in the methods. It was unclear up until this point how the contours in Figs 6 and 7 were developed; this method needs to be laid out earlier on before presenting the results from these figures. The method could also be elaborated on; interpolated how? Based on how many measurements?

#### Discussion:

65. The first two sentences of the discussion section are quite vague. Please provide more detail, i.e., X days instead of “weeks”, what is meant by fluctuated, what is referred to as “they” in “they have not previously been noted”. Please be clear and state that increases in IN concentrations (up to  $XX\text{ m}^{-3}$ , on average) were immediately observed within 1 day after heavy precipitation events ( $X\text{-}X\text{ mm day}^{-1}$ ), and exponentially decreased 20 days after the heavy rainfall.
66. The exponential decrease in IN concentrations is very difficult to visualize with the CD values; the figures all show increasing CD. Although this might account for a decrease with respect to pre-key day concentrations, it is not apparent. As delineated in a previous comment, it would be easier to distinguish the decrease if just daily mean IN concentrations were shown over the 20 day period following a key day. Along this, is the “before” value a mean over all 20 day or is there a mean per day and day 1 of the before mean is compared to day 1 of the after mean? Is this done separately for each site, each event or averaged? As evident, this before comparison causes some confusion which could be eliminated if daily mean IN concentrations for the 20 days before and after were separately shown in each of the figures.
67. p 25516, lines 7-8: How do biological particles multiply as a consequence of rainfall?
68. How did the authors account for emission of dust as IN after a heavy precipitation event? Surely after the ground dries, dust could potentially be lofted along with microorganisms, especially in regions in central Australia or other semi-arid regions.
69. p 25518, lines 1-2: Wouldn't the decrease in  $F_{hv}$  downwind be due to CCN and not GCCN?  $\text{SO}_2$  from the power stations would form smaller CCN. What is meant by GCCN ratios? This is the first time ratios of GCCN are mentioned.
70. Section 5.1: The role of biological GCCN is unclear in terms of decreases in precipitation quantity downwind of the coal-fired power stations. What do microorganisms have to do with power plants? Power plants are certainly not a source, perhaps the authors are attempting to say the development of the power plants induced decreased emissions of bioaerosols and increase CCN emissions, thus resulting in less precipitation events?
71. p 25518, lines 17-20: How are power stations a source of GCCN? They would presumably be a source of sulfate aerosols or CCN.

72. p 25518, lines 22-24: How are cities and power plants a source of microorganisms, in comparison to the pre-1960 undeveloped land? This concluding sentence is somewhat confusing. The downwind increase in what? Rainfall decreased downwind of these locations according to the figures and previous text, so what increase are the authors referring to?
73. Section 5.3 heading:  $F \dots Q$  or  $v$  or both? Please label properly.
74. Section 5.3: It would benefit this section to provide the location of Mt. Buffalo Chalet on the map figure. Further, the values of  $F_Q$  and  $F_v$  are discussed, however, these are not shown in any figures. Having a new figure (on top of the new Fig 1 map I suggest above) which includes an elevation map of Mt. Buffalo (elevation since various elevations are discussed in this section), and since one site was used for  $F_Q$  and  $F_v$ , as stated on p 25519 line 14, provide in this figure a plot containing four time series lines of pre-1960  $F_Q$ , post-1960  $F_Q$ , pre-1960  $F_v$ , and post-1960  $F_v$ . a figure such as this would make this section much stronger and enable visual representation of the discussion. As is, the results discussed in this section are not evident.
75. p 25520, line 1: What  $F$  value?  $Q$  or  $v$ ? During pre- or post-1960? Also, the results discussed in this paragraph are not evident in the figures ( $F$  is discussed, but the ratio of pre- to post-1960  $F_v$  is only shown. Perhaps another figure is needed to include just the  $F_Q$  and  $F_v$  for this part of the discussion, and not the ratio. Further, it is not clear where this result is coming from. As with this and other parts of the discussion, please reference to which figure number the authors want to guide to reader to while discussing results.
76. Overall, the discussion does not include a distinct comparison of the different areas of Australia. Why are the different areas brought up in the results if they are not going to be discussed? Please provide more discussion on the different in rainfall and IN concentrations in the 5 proposed domains as defined in comment 54.
77. The sections on the  $F_{hv}$  should be combined (5.1 and 5.2) since they have a common theme. These sections disrupt the flow of the paper and do not following the context of the paper. With that said, the results from these analyses can be used to emphasize that bioaerosols are important in terms of precipitation distribution. Pre-1960, before the power plant development and growth of Melbourne, one can assume the land was composed of grasslands of some sort and less people translates to less anthropogenic aerosols. The authors could use this to show that pre-1960 when bioaerosols were likely predominant compared to post-196, there was a higher frequency of rainfall. Thus, they could use this evidence to emphasize their speculation of bioaerosols serving as IN in these regions. As is, a large portion of the discussion is focused on the anthropogenic component when that is not within the context of the paper. The analysis and results are there, the discussion just needs to be revised to focus on how these results show bioaerosols are important within the bioprecipitation process.

Figures:

78. Fig 1: Why is 30 days shown when the remainder of the paper focuses on 20 days? Also, why is this in  $L^{-1}$  when the remainder of the figures are in  $m^{-3}$ ? Please be consistent with the units used, and if they need to be different then provide the reasoning. Please provide where these sites were in the caption and what “i” refers to. Provide error bars.

79. Fig 2a: Aerodome and Homestead sites are not discussed in the paper, please provide discussion on what these are and how they are different.
80. Fig 2b: Is this for all sites? Were there differences in Aerodome versus Homestead sites? How many key days > 25 mm were there (by the way, shouldn't this be  $\geq$  and not just >)? What is the error or standard deviation for  $15.3 \text{ m}^{-3}$ ? This value is confusing since it is CD and not just post-key day precipitation, CD may not be useful for this paper. As mentioned in previous comments, plot pre- and post-key day IN concentrations as two separate lines. Also, since one of the main focuses is the difference in the areas, plot one line per area. Provide error bars.
81. Fig 3: What are the  $R^2$  values for these? Please be consistent and provide correlation coefficients for all trend lines. What are the dates of, i.e., how many key days do these plots include? Provide error bars.
82. Fig 4: Provide units for  $F_v$ . What was the rainfall threshold used? Do the 22% and 60% only include sites that had key days or are these sites that have rainfall for 20 days following a key day? If so, why did the other sites not have key days? This plot is confusing because it shows that it rains every day after a key day. How is this possible? What time period does this cover (provide in caption)? Provide error bars.
83. Fig 5: Same comments as for Fig 4. It is not clear what the seasonal correction does. Perhaps it would be better to show a line for each season, which would warrant additional discussion on seasonal differences in rainfall frequency.
84. Fig 6: Please provide the entire date range. How was the ratio averaged? Was the pre-1960 and post-1960 average determined then ratioed, or were ratios determined first from all data points, then averaged? How many measurements does this include? 100? 1000?
85. Fig 7: Same comments as for Fig 6. Why aren't Figs 6 and 7 combined? Why are the contours cut off and why do they exclude two of the sites?
86. Do Figs 6 and 7 include key days or just rainfall preluding precipitation events? Seems as if they should not include the key days, since the focus is on the resulting effects of IN emitted after a key day on precip
87. Fig 8: Plot the frequencies for south-western and south-eastern sites together for direct comparison. Same goes for the quantity. This will provide a better visual representation for the spatial comparison discussion. Provide error bars.
88. Fig 9: I'm not clear on the point of the figure. What do the percentages mean? Provide error bars.

#### Technical Corrections:

89. Please be consistent with the dash or no dash in microorganisms/micro-organisms. It is used as both throughout the manuscript.

90. Same line and sentence as comment 18: “in IN concentrations” should be added to after, “...spectacular but fluctuating increase”, otherwise it is unclear as to what is spectacular and fluctuating.
91. p 25512, line 8: 20 what? Units?
92. p 25512, line 12: Define the “historical series” (i.e., 19XX-1960 = pre-1960 and 1960-19XX = post-1960).
93. p 25514, line 2: Define  $PM_{10}$ .