Responses to reviewers of ACPD 14, 25503-25532, 2014 (Bigg, Soubeyrand and Morris)

We thank the reviewers for their thorough work and their exhaustive comments. In light of the > 90 comments provided by the first reviewer and the overlap in comments among some of the reviewers, we have grouped the comments according to the section of the manuscript that they cover. The reviewers' original remarks are presented in *italics* and our response is in normal script.

Title: *Reviewer 3 suggested turning the title into a question.* We changed the title to: Persistent after-effects of heavy rain on concentrations of ice nuclei and rainfall suggest a biological cause.

Abstract: *Reviewer 2 suggested addition of "atmospheric" to IN*. Adopted.

Reviewer 1's comment 8 suggested introductory material should be in the Abstract. This is required by the journal, so it has been included.

Comment 9 didn't realise that the four areas referred to were two for IN and two areas for rain. The changed wording might make this clearer.

Comment 10 is not pertinent– differences, not ratios, were calculated. Therefore, days without rain have no ill-effects on the calculation.

Comment 11: Where did the 92 years come from? The new wording might make this clearer. There were mistakes in the text: in number of sites and the minimum length of record. These have been rectified.

Comment 12: How is there an increase in rain on the day following heavy rain? By describing the calculation more fully, difficulty in understanding our description of the events should have been overcome.

Comment 13: Bacterial populations not measured …speculation. This is now stated as a hypothesis. Reviewer2 made a similar comment.

Comment 14: What if there was no rainfall before a key day? It wouldn't matter because differences were measured, not ratios. A more detailed description of the calculations has been made to attempt clarification.

Comment 15: *Differences in cumulative totals is vague*. As mentioned above, the improved description of the phenomenon should eliminate difficulties in understanding.

Comment 16: *Unclear how power stations related to bioaerosols*. The discussion in section 6.1 should make this clear. It would not be appropriate to give the reasons in the Abstract.

Comment 17: Not applicable to revised Abstract.

Introduction:

***Reviewer 2**, p.6 line 19. Add "supersaturation" after 0.3%.* Done

p.5, line 14: "*There are no known phenomena by which mineral or inert CCN, GCCN or IN could be enhanced in this way" seems a dangerous shortcut*. The statement has been modified.

p.6 line 23: What does "much more influence" refer to? "than changes in CCN" has been added.

p.6 lines 23,24. How could changes in IN and GCCN have more influence if 4 orders of magnitude fewer than CCN? If it is accepted that IN and GCCN have a significant influence on rainfall, their **proportional** changes as a result of rain would be very much larger than those of CCN.

p.4 line 25. There is no mention of Rosenfeld's paper on negative feedback on rainfall of desert dust. This has now been added, noting that dust storms of the scale of those in the Sahara are so rare in the regions considered in this manuscript that they can have little effect on overall rainfall.

***Reviewer 1: Comment 18.** Define Bowen ratio*. This has been added, but it is unlikely to help non-meteorological readers.

***Comment 19:** It would be better to introduce global scale analysis before short-term effects.* We think the sequence of events as presented in the original version of the manuscript is better.

Comment 20: *Suggested restructure of p.5 l.22-23 and define "stimulation"*. The sentence has been re-worded.

Comment 21: *Add information on aerosolization*. This is now linked to the later sentence about *P.syringae*.

Comment 22: p.6 l.8. "Spectacular" is qualitative. Add comparisons. Details have been added.

Comment 23: *Could meteorological influences be responsible for the 20 day increased rainfall following rain shown by Soubeyrand et al?* There is some serial correlation in rainfall sequences that can be illustrated by lagged correlation of lengthy sequences. It is usually very slight beyond 2 days and we know of no evidence that it could last for 20 days. Exceptions were found for sites with highly seasonal rainfall where in some cases more heavy falls of rain occurred before the period of most frequent rain. This leads to apparent increases in after-effects that are of meteorological origin. The method described in the manuscript for removing this effect was also criticised and has been re-written.

Comment 24. The authors should provide details of the properties of sites selected for analysis. The requested details of positions, record length and elevations of sites given in the added Tables 1, 2 and 3 already involve a large use of journal space. With more than 160 rainfall sites this would be a huge task, even if the information were available. Large changes in the environment will have occurred at most of them during the 97 to 155 years of records and would be impossible to document. The selection of rainfall sites is discussed in a revised section 2.2. For IN sites, selection was simply based on the information needed from the particular series of measurement. The motivation for each is described in the revised section 2.1.

Comment 25 p.6 line 17: *Elaborate on combined effects on rainfall of CCN, IN and GCCN.* There is a very large amount of previous literature dealing with these effects. The paper by Möhler et al. was quoted as a relatively recent contribution describing them.

Comment 26, p.6 l.24: "*This section would be more suitable earlier in the Introduction*". We feel that the content of this section would not be well-enough appreciated if it were moved up earlier.

Comment 27: "*A lot of mention of feedback loops. Could be more distinctly presented*". *In comment 4:* "*what is meant by positive and negative feedback*". Feedback loops are well-described phenomena in ecology and environmental sciences, in biochemistry, in modelling and in economics. In the Earth sciences the most well-known feedback loop is the CLAW hypothesis that describes the feedback loop between the oceans and the Earth's climate as proposed by Robert Charlson, James Lovelock, Meinrat Andreae and Stephen Warren in the late 1980's. We have added some extra explanation (lines 2-6 of the revised manuscript), but overall we feel that most readers will already understand this concept.

Comment 4: Use of vague terms, e.g. quoting p7 lines 5-8. What factors? The previous sentences having described several possible influences on rain, "those" obviously refers to them. Repetition of the subject of a previous sentence is tedious. In the other examples of vague references, the same comment applies. Such abbreviations are very commonly used in English.

Comments on Methods, section 2.1 and 2.2.

Reviewer 1: Comment 28: *Restructuring necessary. Sources and details of data should come first. Full details of sites and maps should be provided*. *The authors should clarify why the specific IN sites were chosen. Details of measurement methods should be given. Comments 35-41 ask for much more detail of IN measurements. Comment 32 suggests the data appear to have been "cherry picked". Comments 46 and 51 show that the distinction between IN measurements (section 3) and rainfall (section 4) was overlooked.*

Reviewer 2: *Information on raw data missing. Ranges of IN concentrations, rainfall frequency and intensity, means, quartiles or any statistical parameter that could summarize the whole data set*.

The restructuring is a good suggestion and has been followed. Information on the raw data has been provided in the form of Tables 1-3. Figure 2a gives a map of the widespread IN sites of group 1, Table 1. The co-ordinates of the remainder are in Table 1 and can be located with sufficient accuracy on Figure 2a. Figures 6 and 7 showed the locations of all rainfall sites used. Reasons for site selections have now been included. It is apparent from some of the comments of reviewer 1 that the paper did not make a sufficiently clear distinction between IN and rainfall data. We have provided additional information to reduce this uncertainty. Measurement methods have been given for IN. On the other hand, indicating methods of rainfall collection are unnecessary because a simple standard rain gauge was used at most sites. The statistical information requested by Reviewer 2 for 36 IN sites and 166 rainfall sites would create an immense and confusing file. In the case of rainfall records extending as much as 155 years there have been changes with time that would have to be considered in detail. Such information would be valuable in attempting to find reasons why after-effects of heavy rain varied within a much smaller selected number of special sites and could be the subject of future investigations.

Comments on "data manipulation", now called "basic method for calculating after-effects of heavy rain."

Reviewer 1 comment 34. The CD method is very unclear.

Rev.2: I had some difficulties to follow the calculations that have been done and giving generic equations would probably help. Showing an example would help.

Rev.3: The description of the method could be improved. I couldn't understand it without reading Soubeyrand. A mathematical formulation and/or graph would probably help.

Reviewer 1: Comment 28: It is unclear why and how the authors chose to calculate CD and no error analysis is presented.

Noting that Reviewer 1 confused rainfall intensity with rainfall rate (mm/hr), we have changed intensity to quantity in the Soubeyrand et al. paper. Because of the adverse comments on "feedback", instead of F we have used CD_Q and CD_F where $_F$ now means frequency (=occasions). We didn't use $_Q$ because it is too easily confused with Q

Rev.1: Comment 86: *Are key days included in figures 6 and 7*? A statement was added in the following section that days with rain above the chosen threshold occurring in the 20 days before or after a key day are included and why this was done.

4. Persistent effects of rainfall on atmospheric IN concentrations

Comments of reviewers: *Reviewers 1 and 2*: *The temperature of measurement was well below the threshold temperature for mineral dusts to become active IN. Rosenfeld's desert dust paper should be quoted.* The desert dust hypothesis is now included in section 2.1. *Reviewer 1's comments 46 and 51* failed to notice that the IN and rainfall sections are entirely separate issues. This has been made more obvious by giving the rainfall section a separate heading. *Comments 47-50* have been dealt with in the text.

5. Persistent increases in rainfall following days with rainfall above a certain threshold.

Reviewer 1 comments 4, 27, 58, 61 stressed the need to discuss feedback loops referred to in the text. The concept was added to the Introduction and is now also mentioned in the following section but further consideration of it deferred to the Discussion.

Reviewer 3 suggested the presence of an unknown mechanism that affects both IN and rain. One aim of this paper has been to show that heavy rainfall is one such mechanism. It is hard to think of another mechanisms, but a statement that it might exist (however unlikely) has been added. The feedback factors F_Q and F_v used in the manuscript have been changed to CD_Q and CD_F to conform with the revised equation 1.

Reviewer 1 wanted great detail about choice of rainfall sites and this has now been dealt with in section 2.2.

5.1 CD_F for sites listed in Tables 2 and 3.

Reviewer 1. Comment 59. Rainfall measurements at IN sites should have been included in the analysis. *Sites should be labelled western and eastern*. The IN sites used were not chosen for their quality or length of rainfall records, nor are the data sequences long enough for rainfall records to be of much use. It is obviously preferable to use the selection methods for rainfall sites discussed in section 2.2. The more specific southwestern and southeastern terms used here are not inaccurate and are preferable in our opinion.

Comment 60: Discussion needed on the failure of some sites to show a logarithmic trend in CD. This has been added.

Reviewer 2. There should be a delay between IN and rainfall if a feedback effect is involved. Should logically be a correlation between IN and rain. The evidence from Hirano et al (1996) and Huffman et al. (2013) is that the initial response of IN concentrations to rainfall is very much less than one day. The correlation between subsequent change in IN and rain is exemplified by their very similar logarithmic CD responses.

Plotting IN concentration against rainfall frequency. If we had 100 year records of IN measurements this would be very useful. Unfortunately we do not. In short records, meteorological influences dominate.

5.2 CD_{HF} for the southeastern group of sites listed in Table 2.

Reviewer 1: comment 62. Data not divided in half. Change made.

Comment 63. The ratio did not decrease itself. True. Changed the wording.

Comment 64. Calculation of contours, method should be developed earlier. We feel that this is best explained just before the results of the calculations are presented to help the reader follow.

Reviewer3, comment 4. Confusing discussion. Mistakes made in increased "ratios". A brief explanation of the inferences should precede the discussion. Mistakes were indeed made in discussing the changed ratios and have been rectified. An attempt has been made to improve the presentation and to give a preview of the relevant part of the discussion.

Comment 5. The aluminium smelter could not have had such a huge SO_2 emission. True. The error arose from confusion between the Alcoa power station at Anglesea and the Alcoa smelter at Portland. Statement withdrawn.

5.3 CD_{FH} for the southwestern group of sites listed in Table 3.

Reviewer 3: Errors in statement of downwind effects of Muja power station. Corrected**.**

*Rev.1: Comment 84: Provide the entire data range***.** This is now in Table 3.

How was the ratio averaged? The first suggestion made by the reviewer is obviously correct.

Comment 85: Why aren't figures 6 and 7 combined? **I**mportant detail would be lost in halving the size of the figures**.**

*Why are the contours cut off and why are two sites excluded***.** The reasons for this are now discussed in the text.

Comment 86: Do figs 6 and 7 include key days? A statement about this is now included in section 3.

5.4 Magnitude of the after-effects of heavy rain.

Reviewer 1: Comment 87: Plot the two figures together in figure 8. Because of overlap of some of the curves it would be confusing to combine the figures. Therefore we preferred to not change the figures.

5.5 How does the choice of key day threshold influence D_Q and D_F?

Reviewer 1: I'm not clear on the point of this figure. Provide error bars. Figure 9 has now been omitted, and this section revised.

6. Discussion

Rev 2. (a) How did you build the cause-consequence relationship between IN and rainfall? (b) What is the delay between IN and rainfall frequency? (c) Would be interesting to find the correlation between IN concentrations and rainfall frequency. (a) The cause-consequence relationship has now been explained at the beginning of section 5. (b): as mentioned there, extra IN are available within a day to potentially influence the rain production. Delays are not therefore to be expected. (c): For this to be successful, measurements of IN concentrations would be required for periods on the order of 100 years because of the many possible influences on their concentration and on rain formation that are not connected with the direct link.

Rev 1. Comment 65: First two sentences of the Discussion are vague. This has been remedied.

Comment 66: The exponential decrease in IN is difficult to visualize when CDs increase. The failure of the reviewer to understand the nature of the calculations and their results has led us to write a new introduction to the discussion section.

Comment 67: How do biological particles multiply as a consequence of rainfall? Water is frequently a limiting factor for multiplication of biological organisms, most importantly as a substrate for the action of multiple enzymes that are fundamental to the biochemical processes involved in building cells. However, an explication of how water stimulates growth is clearly outside the aim of this paper and outside the scope of this journal. The references cited provide examples that illustrate that biological particles (i.e. the microorganisms themselves) do multiply as a consequence of rainfall.

Comment 68: How do the authors account for the emission of dust as IN after a heavy precipitation event? Surely after the ground dries dust could be lofted….? This speculative emission of dust would presumably be more important before the rain than after. The data about IN indicate that concentrations are greater after.

Comment 69: Wouldn't the decrease in F_{hv} (now called CD_{FH}) *downwind from power stations be due to CCN rather than GCCN*? D_F measures the difference in rain frequency after and before each rain event above a certain threshold. It, and its cumulative derivative CD_F, only respond to a physical

quantity that is altered by the occurrences of rain. Power station effluents do not do this (except during the period in which it is raining and shortly afterwards when scavenging reduces the number of such particles that remain airborne). The alternative hypothesis is that the influence on subsequent rain due to additional particles produced by rain becomes less important when there are many particles not influenced by rain that can initiate rain. The added CCN from power stations will tend to reduce the probability of rain but the very large particles that become coated with sulfate and are very numerous in power station effluents will tend to initiate rain. The balance between these two influences will alter with the distance from the source in ways that can only be determined by observations. It appears from Figures 6 and 7 that the balance is in favor of increasing the probability of rain.

Comment 70: The role of biological GCCN is unclear in terms of decrease downwind from power station. See the answer above. The revised text should clarify this.

Comment 71: How are power stations sources of GCCN? Sulfate derived from oxidation of sulfur dioxide in the emissions deposits on the large particles that power stations produce, rendering them very efficient GCCN because of size and hygroscopicity.

Comment 72: How are cities and power plants sources of microorganisms? Power plants are not important sources of microorganisms. Cities may be, if changes in habitat for microorganisms has resulted in a net increase in their populations. Humans emit a large number of microorganisms and buildings do as well. Maintenance of monuments is continuously plagued by microorganisms. Also, well-known example of cities as sources of microorganisms is the emission of bacteria – including Legionella – from towers of cooling and ventilation systems.

Comment 74: Location and elevation of Mt. Buffalo. This is now given in Table 3. Its location on the map of Figure 6 can be deduced from the co-ordinates.

Comment 76: Why are different areas brought in if not discussed? The revised text includes some discussion on areas chosen.

Comment 77: Sections 5.1 and 5.2 should be combined. Because we are dealing with two very different results and probably different physical causes, it is preferable to keep them separate.

Suggested revision of discussion to focus on biological rather than anthropogenic aerosols. Because both can exert some control on rainfall they have to be considered together.