We would like to thank the reviewers for their valuable input. Due to the overlap of some of the reviewer comments, we combined the responses into one document. Reviewer comments are in black, while our responses are provided in blue italics.

Anonymous Reviewer #1

This is a reasonable important advancement in the field although this is minimized by the fact that it forms the third in a set of three papers. The references Ault et al. (2011) already described the 2009 data while Creamean et al. (2013) described the 2011 data. Thus the only new data in this paper is the 2010 data set and the fact the authors attempt to use these three sets to draw interannual statements on aerosol sources and effect on precipitation. These are generalized as 'dust and biological particles in the first year, biomass burning and pollution in year two, and long range transport in the 3rd (note as an aside that 'long range transport from distant sources', the phrase used in the manuscript, is redundant and the last three words can be eliminated in all cases).

My major concern here is the level of repeat of previous work (see this paragraph and next regarding the figures). As a whole, this paper seems to just meet the minimum requirements for publication in ACP. There is a bit of new data here - the addition of one season and some slightly expanded correlations that result, but really the authors appear to have published essentially all this data and conclusions before. I sincerely hope that there are not going to be yearly publications from this study; I, personally, think the minimal new data presented here just barely passes for justification of a new paper. Unless something new is really learned in a future year I hope the authors will step away from such minimal incremental additions to the literature.

Regarding figures, I went back and re-read the highly referenced Ault et al. (2011) and Creamean et al. (2013) – much material is cited instead of appearing here. What is rather surprising is that many figures here are essentially repeats of what was published previously. Figure 1 in Ault et al. is Figure 1 here (Ault et al. actually contains more data) – please eliminate and use reference as is done in the text ; the representative spectra shown in Figure 2 appear identical – but contain less information than Fig. S2 of Creamean et al. (2013) ; Figure 3 here appears to repeat the data from Figure 5 of Ault et al. ; Figure 5 here appears to repeat the data from Figure 1 of Creamean et al. I will ask the editor to decide how to handle this but the authors should remove these repeated data figures and instead reference the material.

"From distant sources" was removed.

The reviewer does bring up a valid argument in that the results shown from 2009 and 2011 are similar to those shown by Ault et al. (2011) and Creamean et al. (2013). In order to avoid replication, the original Figures 2, 3, and 5 were removed since, as the reviewer points out, a version of these are shown in Creamean et al. (2013, 2014a) and Ault et al. (2011). We modified Figure 1 to just show the map, with the regions of the Sierra Nevada and Central Valley, and SPD highlighted, in addition to source contributions. We intend to keep a map to enable the reader to visualize the topography and different source regions surrounding the site—since this is vital to understanding the residue sources—without having to dig into another reference. We also substantially revised Figure 4 (now Figure 3) to include all the samples from 2010. We revised the text to reflect the removal of the previous figures. For instance, we now state that mass spectra

(was Figure 2) are shown in these previous papers. When discussing results from 2009 or 2011, we reference Ault et al. (2011) or Creamean et al. (2013), respectively instead of showing figures including those data. We have also expanded the discussion on 2010 results (see lines 311-335), providing more examples that support our conclusions and including ambient aerosol measurements from 2010 (lines 210-222 and 279-298). Overall, we switched the focus to 2010, and subsequently discuss all samples from all three years in comparison to each other.

1. The technique is interesting. In essence, ice-phase precipitation and collected. The trapped particles are then analyzed and the authors draw some correlations. What isn't discussed here – and only minimally in the references to the past work – is what fraction of the analyzed particles were actually responsible for ice nucleation? What fraction are scavenged from the particle phase? What fraction are scavenged as droplets or other ice was collected during precipitation? As it reads the authors seem to suggest that all particles were ice nucleating agents but this can't be true. A reasonable calculation for the different meteorological conditions is warranted and should be included in a revised manuscript. Without it I find it hard to believe the correlations are statistically relevant. I'm thinking here of past references from the DeMott group who indicate from ice chamber measurements that only something like 1 in 10^5 or 6 particles act as ice nuclei. If each precipitation element were to scavenge say 10 or 100 particles would the authors draw a reasonable correlation that had anything to do with the formation of precipitation or only the background aerosol?

Indeed, there are relatively low concentrations of IN in the atmosphere, as delineated by the DeMott group. However, the residues we measure in the precipitation are also very low in concentration. For instance, the numbers in Table 1 are typically less than 500 residues, which are analyzed in the time period of about a day or more. The precipitation samples are quite dilute, demonstrating the low concentrations of particles in the solutions. If aerosols were scavenged during descent, we would expect a much higher residue analysis rate, similar to past ambient ATOFMS studies (at SPD, we analyzed thousands of aerosols a day).This is shown in Ault et al. (2011).

Scavenging calculations are dependent on the type and size of aerosol and typically require modeling efforts (e.g., Chate et al., 2011; Croft et al., 2009; Flossmann et al., 1985, 1987, 1988, 1989; Henzing et al., 2006), thus these calculations would be extensive and outside the scope of this manuscript. Although we cannot quantitatively estimate the amount of scavenged versus nucleated residues, we can qualitatively estimate the contribution of what was nucleated versus scavenged. We added a new figure and table (now Figure 2 and Table 2, respectively) which compares the precipitation residue composition to the ambient aerosol measured using ATOFMS concurrent with sample collection time periods. We now discuss how similar types of particles (i.e., dust residues versus ambient dust) were poorly correlated thus were more likely nucleated versus scavenged. This is now discussed on lines 279-298, and reiterated on lines 353-356.

Chate, D. M., Murugavel, P., Ali, K., Tiwari, S., and Beig, G.: Below-cloud rain scavenging of atmospheric aerosols for aerosol deposition models, Atmos Res, 99, 528-536, DOI 10.1016/j.atmosres.2010.12.010, 2011.

Croft, B., Lohmann, U., Martin, R. V., Stier, P., Wurzler, S., Feichter, J., Posselt, R., and Ferrachat, S.: Aerosol size-dependent below-cloud scavenging by rain and snow in the ECHAM5-HAM, Atmos Chem Phys, 9, 4653-4675, 2009.

Flossmann, A. I., Hall, W. D., and Pruppacher, H. R.: A Theoretical-Study of the Wet Removal of Atmospheric Pollutants .1. The Redistribution of Aerosol-Particles Captured through Nucleation and Impaction Scavenging by Growing Cloud Drops, J Atmos Sci, 42, 583-606, Doi 10.1175/1520-0469(1985)042<0583:Atsotw>2.0.Co;2, 1985.

Flossmann, A. I., Hall, W. D., and Pruppacher, H. R.: A Theoretical-Study of the Wet Removal of Atmospheric Pollutants .1. The Redistribution of Aerosol-Particles Captured through Nucleation and Impaction Scavenging by Growing Cloud Drops, J Atmos Sci, 42, 583-606, Doi 10.1175/1520-0469(1985)042<0583:Atsotw>2.0.Co;2, 1985.

Flossmann, A. I., and Pruppacher, H. R.: A Theoretical-Study of the Wet Removal of Atmospheric Pollutants .3. The Uptake, Redistribution, and Deposition of (Nh4)2so4 Particles by a Convective Cloud Using a Two-Dimensional Cloud Dynamics Model, J Atmos Sci, 45, 1857-1871, Doi 10.1175/1520-0469(1988)045<1857:Atsotw>2.0.Co;2, 1988.

Flossmann, A. I., and Pruppacher, H. R.: A Theoretical-Study of the Wet Removal of Atmospheric Pollutants .1. The Redistribution of Aerosol-Particles Captured through Nucleation and Impaction Scavenging by Growing Cloud Drops, And .2. The Uptake and Redistribution of (Nh4)2so4 Particles and So2 Gas Simultaneously Scavenged by Growing Cloud Drops - Reply, J Atmos Sci, 46, 1870-1871, Doi 10.1175/1520-0469(1989)046<1870:R>2.0.Co;2, 1989.

Henzing, J. S., Olivie, D. J. L., and van Velthoven, P. F. J.: A parameterization of size resolved below cloud scavenging of aerosols by rain, Atmos Chem Phys, 6, 3363-3375, 2006.

2. Speaking of which, I'm somewhat surprised by the lack of references to the work of DeMott, especially field studies, in this paper. Indeed most of the non-self-references to ice nucleation here are laboratory studies. It would be good to include a solid section on past studies of ice nucleating particles regarding the above comment on abundance versus scavenged particles.

We added a few DeMott references about field measurements of IN in the introduction on lines 65-66. We also elaborate more on the Pratt et al. (2009) field measurements of IN, on which Dr. DeMott is co-corresponding author.

Unfortunately, we cannot discretely distinguish between the residues that were removed via the different "scavenging" processes, which according to Borys et al. (1988) refers to nucleation and cloud droplet/ice crystal scavenging of aerosol particles. According to Borys et al., "Nucleation scavenging appears to be the process most likely to result in chemical fractionation of the aerosol." Thus, nucleation is the most important process for aerosol removal in-cloud.

We included discussion on lines 279-298 involving comparison of residue with ambient data and that although we cannot determine with great certainty that most residues are nucleated versus scavenged we hypothesize that the majority of the residues were based on the evidence we present on these lines and on lines 353-356.

Borys, R., Hindman, E., and DeMott, P.: The chemical fractionation of atmospheric aerosol as a result of snow crystal formation and growth, J Atmos Chem, 7, 213-239, 10.1007/BF00130931, 1988.

3. I notice that in the text "dust/biological" and "dust and biological" are almost always grouped together and yet the figures separate these groups. Can the authors explain why these two are carried through as a group but then separated in the figures? Perhaps more specifically, can the authors offer a correlation of these two categories? Is the implication here that the two have the same source? That seems to be the implication but doesn't appear to have been actually proven.

They are discussed as a group to represent the residues that potentially served as IN as we clarify on lines 321-325, "Throughout this discussion, the dust and biological residues are combined to simulate the percentage of residue types that likely served as IN and because they are likely from a similar source (Creamean et al., 2013). However they are shown separately in the figures to demonstrate the relative contribution of each, which is particularly important for the biological residues as discussed in more detail below." We added the last part of this sentence in the revision for clarification on our reasoning for separating in the figures.

We show them separately in the figures because we discuss their individual dependence on cloud temperatures as provided on lines 357-368. We did revise the figures so that the colors for dust and biological residues are the same, just with different patterns to demonstrate their relationship but also their different contributions to each overall sample residue composition.

4. Something not described in any detail here is the lack of soluble species in the figures. I think I understand that this material – perhaps sulfate or sea salt or water soluble organics – would be 'lost' when the ice-phase precipitation is melted. That seems reasonable if one is looking at ice nucleating particles but what about for the CCN? Looking at figures 3, 4 and 5 during 'rain' events (that is, the CCN conclusions drawn in the paper) what is the expectation that the insoluble components represent any significant fraction of the droplet forming particles? Aren't these essentially all expected to be hygroscopic – sulfates and the like? This makes me wonder if any of these conclusions are at all reasonable or only an artifact of the method.

Unfortunately, we do not have measurements of soluble ions for all of the samples. We do however, have ion chromatography measurements of the 2009 samples as shown by Creamean et al. (2014a). Results presented there showed correlations between sodium, potassium, sulfate, chloride, nitrate, and phosphate and insoluble organic residues, thus signifying these insoluble organic residues were likely cores of the original particles from biomass burning and/or pollution. Thus, these particles did likely serve as CCN, where the original soluble species that were likely within the coating were indeed hygroscopic. This effect has been demonstrated by a number of studies (i.e., Broekhuizen et al., 2004, 2006 and Ervens et al., 2010). Even partially or slightly soluble organics have been shown to serve as CCN (i.e., Bilde and Svenningsson, 2004, Broekhuizen et al., 2004), particularly if the particles were wet versus dry (Henning et al., 2005).

Further, insoluble organics alone have been shown to serve as CCN, and modeling studies have found better agreement between predicted and observed CCN when assuming insoluble organics, such as discussed by Broekhuizen et al. (2006), Ervens et al. (2010), and Wang et al. (2008).

Thus, our organic residues, although insoluble, could have potentially been cores with soluble coatings or partially soluble organic particles that remained while in solution. We added discussion regarding these possibilities and included the following references on lines 413-429.

Bilde, M., and Svenningsson, B.: CCN activation of slightly soluble organics: the importance of small amounts of inorganic salt and particle phase, Tellus Series B-Chemical and Physical Meteorology, 56, 128-134, DOI 10.1111/j.1600-0889.2004.00090.x, 2004.

Broekhuizen, K., Kumar, P. P., and Abbatt, J. P. D.: Partially soluble organics as cloud condensation nuclei: Role of trace soluble and surface active species, Geophys Res Lett, 31, Artn L01107 Doi 10.1029/2003gl018203, 2004.

Broekhuizen, K., Chang, R. Y. W., Leaitch, W. R., Li, S. M., and Abbatt, J. P. D.: Closure between measured and modeled cloud condensation nuclei (CCN) using size-resolved aerosol compositions in downtown Toronto, Atmos Chem Phys, 6, 2513-2524, 2006.

Ervens, B., Cubison, M. J., Andrews, E., Feingold, G., Ogren, J. A., Jimenez, J. L., Quinn, P. K., Bates, T. S., Wang, J., Zhang, Q., Coe, H., Flynn, M., and Allan, J. D.: CCN predictions using simplified assumptions of organic aerosol composition and mixing state: a synthesis from six different locations, Atmos Chem Phys, 10, 4795-4807, DOI 10.5194/acp-10-4795-2010, 2010.

Henning, S., Rosenorn, T., D'Anna, B., Gola, A. A., Svenningsson, B., and Bilde, M.: Cloud droplet activation and surface tension of mixtures of slightly soluble organics and inorganic salt, Atmos Chem Phys, 5, 575-582, 2005.

Wang, J., Lee, Y. N., Daum, P. H., Jayne, J., and Alexander, M. L.: Effects of aerosol organics on cloud condensation nucleus (CCN) concentration and first indirect aerosol effect, Atmos Chem Phys, 8, 6325-6339, 2008.

Anonymous Reviewer #2

1. The study is only about the impact of insoluble particles on precipitation, by serving as either CCN or IN, or both. Nevertheless, the title of the manuscript doesn't specify that. The title looks like a comprehensive discussion of all aerosol sources. But the soluble aerosol species is not the purpose of this study and would be a totally different story. It won't undermine the significance of the study by specifying that. Similarly, all the discussions about the aerosol sources need to be defined more accurately as insoluble aerosol sources since the soluble part is not measured and discussed in this manuscript.

The reviewer brings up a valid point. Accordingly, we changed the title to, "Impact of interannual variations in sources of insoluble aerosol species on orographic precipitation over California's central Sierra Nevada". We also defined more clearly throughout the text that we are focused on sources of the insoluble components of aerosols.

2. Random coincidence could play a large role in correlation when the sample size is small. The reliability of aerosolization method needs to be better supported. The authors admitted the composition change during the aerosolization process. Then the authors will need to prove that the change is minor or the change has a small effect on the way aerosol influences cloud seeding.

This topic is the main focus of the Creamean et al. (2014a) publication, where control experiments are performed on standards to validate what is observed in the precipitation samples. We reference this and other material in order to prevent repetitiveness: "This aerosolization method can produce single soluble and insoluble particles, agglomerates of different particle types, and coatings of soluble species on insoluble residues. Thus, the composition is likely somewhat altered from how the particles would have existed in the atmosphere (Holecek et al., 2007). Even with the caveats associated with the aerosolization process as discussed in Creamean et al. (2013; 2014a), this method provides useful information on chemical differences in the aerosols seeding clouds."

We added more discussion on the caveats with regard to the biological residues, which are observed in the precipitation but not as ambient aerosols. On lines 368-381, we added: "It is important to note that the purely biological residues could be a result of the aerosolization process, thus might have originally been components of the dust particles. Although biological particles were not observed as ambient aerosol at the ground, they were observed as interstitial aerosol and in individual cloud particles during the 2011 in-cloud aircraft measurements (Creamean et al., 2013). However, when examining the 2011 measurements, the fact that: 1) a higher abundance of purely biological residues was observed in the precipitation samples compared to the interstitial aerosol or cloud particles and 2) a higher abundance of dust mixed with biological material was observed in the aircraft measurements compared to the precipitation collected on the ground, supports the fact that the majority of the biological residues are likely separated from the dust during the aerosolization process. Even considering this issue, the dust particles that were present in cloud still contained more biological material during time periods with warmer cloud temperatures, thus would have enabled the dust to serve as more efficient IN as delineated by Conen et al. (2011) and O'Sullivan et al. (2014)."

3. The pronounced effect of insoluble particles on precipitation over California's Central Sierra Nevada area, presuming the significance is supported by this study and other related studies, could also be important for some other regions. The paper could simply discuss the global significance of insoluble particles in terms of precipitation and the climate.

On lines 523-527, we added, "The relationships between insoluble precipitation residues and their potential climate impacts could translate to a global scale, i.e., apply to other orographic regions where such insoluble particles are found in and impact the formation of clouds and precipitation. Thus, understanding insoluble residue sources has implications on a global level, particularly when modeling their impacts on clouds."

Detailed suggestions:

1. In the abstract, there is one sentence: "The correlation between the source of aerosols within clouds and precipitation type and quantity will be further probed in models to understand the mechanisms by which local emissions vs. long-range transported dust and biological aerosols play roles in impacting regional precipitation processes". The model study is described neither in this manuscript nor in a companion paper of this manuscript. It might be an integral part of the funded project. But this sentence doesn't seem to belong to this paper, especially not the abstract.

This sentence was removed from the abstract.

2. The first paragraph of the introduction part tries to discuss the impact of CCN and IN on precipitation comprehensively. Again this paper discusses about the insoluble fraction of aerosol. It needs to be emphasized in the introduction as well.

The last two paragraphs of the introduction were revised to highlight that we are focusing on the insoluble components of the aerosols in the precipitation samples.

3. The last paragraph of the introduction tried to outline the purpose of the manuscript, but the order of the two goals seems to be reversible. It makes better sense to me to first identify the temporal variation of sources and then find the linkage between source variation and precipitation.

The order of these goals was switched.

4. In the second paragraph of the discussion part, there is one statement: "A modeling study of aircraft measurements from 2011 presented by Martin et al. (2014) show the presence of organic carbon residues at lower cloud levels during prefrontal storm conditions in the Sierra Nevada". It would be a poor citation to support the presence of a chemical species using a model study, especially in a paper discussed about measurement results. It would be great to infer the significance of a measurement study by a model study though.

We changed this sentence to, "A modeling study of aircraft measurements from 2011 presented by Martin et al. (2014) shows the presence of organic carbon residues at lower cloud levels during prefrontal storm conditions in the Sierra Nevada, demonstrating the significance of our observations and how they validate model results."

5. Conclusion part could be more concise and better organized. The last paragraph seems like a future study plan for a bigger project, not an indispensible part of this paper. The whole paragraph could be replaced by one sentence indicating the significance of this study in future modeling studies. The detailed discussion of how the result could be used in model studies seems to belong to the discussion session rather than the conclusion session.

We moved most of the modeling implications to the end of the discussion section on lines 480-488. We also restructured the last paragraph of the conclusions to emphasize the importance of our findings and added more discussion on the importance of these observations to the Sierra Nevada. We kept the sentences regarding these results serving as motivation for CalWater 2 to demonstrate their importance not only from a climate or modeling aspect, but also as a key factor for planning for future measurements. We did not want to disregard the fact that although our results do provide a noteworthy advancement, we need more work to better constrain aerosol-cloud-precipitation interactions.

6. Please check grammar inaccuracies in the manuscript. There are a few.

Grammar inaccuracies were checked and corrected.

Anonymous Reviewer #3

Comparing the conclusions drawn from the results to those of the accompanying and regularly cited studies of Ault et al., 2011 and Creamean et al., 2013, 2014a – the additional gain in

knowledge should be worked out more precisely. It should be differentiated what data was directly taken from previous work, what data was reanalyzed and exactly what data is new. This would help to better distinguish between new conclusions and conclusions already drawn from previous work.

We agree that this was an issue in our paper and revised substantially to emphasize the new findings while citing the old. Please see response to the first (unnumbered) comment of Reviewer #1 for the details of the changes.

One particular concern relates to the classification of the particle types into a) dust, b) mixed dust, c) bio, and d) biomass. To my knowledge the identification of biomass burning particles by single particle MS is pretty straight forward, whereas the identification of pure biological material and/or biological material on dust (= soil dust?) and/or pure dust is rather complicated. Therefore I wonder if the datasets of all three years were treated consistently concerning the classification process? This is important and might bias the interpretation since the difference in IN properties (activation temperature) and thus the impact on precipitation formation between pure dust and soil dust should be rather small compared to pure dust and pure biological particles. Did the authors grouped the particles in a similar manner, and were the mass spectra of the dust/biological types comparable? In the current study, the mixture of biological material with dust is combined to dust (p940, l22ff), and in a next step the categories dust and bio are combined to an IN-active fraction. Was this done in a similar way in the previous studies, and what is the fraction of the mixed dust/bio type in the dust class?

The datasets from all three years were treated consistently as the lead author carefully classified all residue types from all samples for this work. The lead author of the current work was also the lead author of the 2011 results presented by Creamean et al. (2013), thus the methods were carried out in the same manner as in that manuscript. Further, the lead author classified the residue types in the 2009 work with the lead author of Ault et al. (2011). This was done to ensure consistency in the classification methods for the previous works as well as the current compilation of all three sampling years.

For the mixed dust/biological type, almost all of the dust had some sort of biological signature (organic nitrogen and phosphorus; $^{26}CN^-$, $^{42}CNO^-$, $^{63}PO_2^-$, $^{79}PO_3^-$, $^{97}PO_4^-$). However, there was a calcium-rich dust that was predominantly present in 2011, which contributed 7-91% of the total dust residues in the samples. This type was not as dominant in 2009 or 2010 (<10% of total dust residues in each sample).

Figure 2 shows the representative mass spectra for each of the precipitation residue types. Are these average mass spectra after classification (i.e. after application a clustering scheme), or a these representative single particle mass spectra? How would a mixed dust/bio particle look like?

This figure was removed as similar figures showing these spectra are presented by Creamean et al. (2013, 2014a) and Ault et al. (2011).

Further remarks:

- P935, l7: The IN population is influenced by dust rather than the IN itself as the dust most likely represents the IN.

We changed "IN" to "IN populations" on line 87.

- P935, l10: I wonder if the Pratt et al., 2009 citation is the correct one in terms of showing that bio IN a more effective than dust IN. Isn0t it rather a case study of a mixed-phase cloud that included more bio residuals than other types? Can the authors draw the cited conclusion from this result?

We eliminated the Pratt et al. (2009) reference as, the reviewer points out, it does not provide a direct result showing biological particles are more efficient IN than dust. We added the Murray et al. (2012) reference instead, as that manuscript has a great figure showing a compilation of biological versus dust IN. However, to provide examples of both laboratory and in situ measurements of biological IN, we changed the last sentence in this paragraph to, "Conen et al. (2011) demonstrated even biological fragments such as proteins can largely determine ice nucleation properties of soil dust in a laboratory setting, while Pratt et al. (2009) confirmed the importance of biological IN in orographic cloud ice formation using in situ aircraft measurements."

- P936, l24ff: The study investigates the impact of different aerosol sources on precipitation. In this context, I wonder if besides the precipitation samples in-situ ATOFMS measurements were made of the interstitial aerosol and the total aerosol during nonprecipitation periods – and if yes, how do those compare to the residual. This analysis might be beyond the scope of this paper, but it0s an interesting scientific question from the atmospheric perspective but might also help to understand some of the caveats associated with the aerosolization process (p939, l14ff). These caveats, by the way, should be repeated briefly as the analytical method and its potential errors are important for this paper.

We only have interstitial aerosol observations during the 2011 study, since that was the only time aircraft measurements took place. Those are discussed in detail in Creamean et al. (2013). We do have ambient aerosol measurements during precipitation periods for all three years. The ambient data for 2009 and 2011 are shown and/or discussed in Ault et al. (2011) and Creamean et al. (2013), respectively, therefore were not presented in this paper. In order to keep the focus on the new observations, i.e., the 2010 samples, we now present the 2010 ambient measurements in comparison to the precipitation residues in Figure 2 and Table 2 and discussion on ambient versus residue composition on lines 279-298.

Although comparing the detailed chemistry (i.e., mass spectra) of the ambient aerosols versus the residues is a great idea, it would indeed be outside the scope of this particular manuscript as it would add quite a bit of discussion unrelated to the main conclusions. Perhaps that analysis would be better suited for a follow up methods manuscript. Further, this was done to some extent for the 2009 samples as presented by Ault et al. (2011).

We do discuss the caveats briefly by stating, "This aerosolization method can produce single soluble and insoluble particles, agglomerates of different particle types, and coatings of soluble

species on insoluble residues. Thus, the composition is likely somewhat altered from how the particles would have existed in the atmosphere (Holecek et al., 2007). Even with the caveats associated with the aerosolization process as discussed in Creamean et al. (2013; 2014a), this method provides useful information on chemical differences in the aerosols seeding clouds." This topic is the main focus of the Creamean et al. (2014a) publication, where control experiments are performed on standards to validate what is observed in the precipitation samples, thus we did not explain in great detail here.

We added more discussion on the caveats with regard to the biological residues, which are observed in the precipitation but not as ambient aerosols. Please see the response to comment 2 of Reviewer #2.

- P940, l24: "possibly" is a vague word. Can the authors be more precise how likely the occurrence of soil dust is in comparison to the production of agglomerates during aerosolization?

We cannot quantify the exact contribution from either soil dust, dust mixing with marine air masses, or agglomeration to the mixed dust/biological particle type. However, we are confident the source is from the first two, thus we replaced "possibly" with "likely".

- P942, l20: "potentially" and "we demonstrate" are two rather conflicting expressions. How assured are the results? Do the authors demonstrate how…could potentially influence…, or do they demonstrate how they influenced…

"Potentially" was removed for clarity and to avoid conflict between the expressions.

- Section 3.2.1 Check figure numbering

Figure numbering was checked and ordered correctly.

- P946, l17ff: this sentence is a bit unclear. Did the authors conclude that the bio residues induce the formation of ice precipitation due to the corresponding correlation between bio residues and precip? Can this be done without any doubt taking the aerosolization process into consideration, the small number of sample residues analyzed and the typically low concentration of IN in the atmosphere. One might have to be more careful here.

Please see the last part of the response to comment 2 of Reviewer #2.

- Section 5: the conclusions contain to a large fraction already published results and a rather long outlook. Please emphasize the contribution of this study in more detail.

The conclusions were revised substantially. Please see the response to comment 5 from Reviewer #2.

Anonymous Reviewer #4

This paper describes the results of some nice work conducted over three years (Feb/March 2009, Jan/Feb/March 10, and Jan/Feb/March 2011) in CA to investigate the association of aerosols with precip. Forty two samples were collected at a single sampling site in CA, and a ATOFMS was used to characterize constituents in the precip samples. The authors found that dust and biological

residues were dominant when precip was in the ice phase, and local biomass burning and shallow clouds produced less precip (in general). This work is important for development of regional climate models for precipitation processes in California (and elsewhere in the world) and should be published. Portions of this dataset have been published already (particularly from the 09 and 11 collection years), but it is the opinion of this reviewer that the synthesis of the three years (which includes some new association/correlation analyses) adds some integrated value. The authors should however, be cautious of data duplication in figures across already published manuscripts.

We agree that this was an issue in our manuscript and revised substantially to emphasize the new findings while citing the old. Please see response to the first (unnumbered) comment of Reviewer #1 for the details of the changes.

Suggested points to be addressed in the revision: Table 1. It would be nice to know how many major precip events (above a certain threshold precip rate) were captured in each sample. In other words, for each sample ID, how many unique/discrete precip events were sampled? Were most of these more than one discrete event? For example, Figure 3 S7 and S10 appear to capture major discrete events, whereas S8 appears to capture a cumulative sample of multiple major events. Any trends that could be attributed to discrete events alone?

Storms consist of at least one sample as provided in Table 1 (storm numbers that correspond to sample numbers). Major precipitation events were classified as storms consisting of precipitation > 10 mm. Hourly precipitation rates do approach or equal zero during certain time periods of the storms (although not the majority of the hours), however, determining the number of hours with precipitation > 0 would not provide any valuable supporting information to our findings. Breaks in precipitation do not signify separate storms, but merely a break in precipitation from the same storm system.

Figure 1 could be removed. Providing GPS coordinates is sufficient for readers these days; a quick cut and paste into GoogleEarth and you are good to go. Picts of the trailers are great for .ppt presentations, but aren't needed here.

We modified Figure 1 to just show the map, with the regions of the Sierra Nevada and Central Valley, and SPD highlighted, in addition to source contributions. We intend to keep a map to enable the reader to visualize the topography and different source regions surrounding the site—since this is vital to understanding the residue sources—without having to dig into another reference.

The results often contain discussion points that would be more appropriate for the discussion section (e.g., p 10, L 24-27, p11, L4-13, etc.)

After considering this comment and moving some of the modeling discussion to the discussion section, we realized our discussion section was more of a broader implications section. We also wanted to keep the more discussion-oriented points in the results section as they explain some of the more specific results, and if moved would disrupt the flow. Thus, we changed to name of the "results" section to "results and discussion" and the "discussion" section to "broader implications".

How do you define a ‘shallow’ cloud? e.g., 3.2.2

A shallow cloud is one where the echo top heights are low, i.e., negative echo top height deviation. This is already defined on line 384.

P16, Line 12. Limited? How so? What future measurements should be taken?

We already define that this is a, “limited number of data points” thus more data points or measurements are needed to develop a longer-term record. However, we did add “additional” in front of “measurements” to provide more clarification.

P16, L16. How do you define ‘local’?

We now define “local” on lines 131-132 as from the Sierra Nevada and Central Valley.

What role might precip scavenging (e.g., collection of material by rainfall during descent to your collection beakers) play in the composition of the constituents in your samples? Perhaps this could be addressed in the discussion?

Please see the response to comments 1 and 2 from Reviewer #1.