Response to the Comments of Keith Bigg (Reviewer 1)

We would like to thank Dr Bigg for his helpful comments and suggestions. We address the individual comments point by point below.

General comment *This manuscript provides a potentially valuable long series of IN data for an area from which there have been no previous IN measurements that I am aware of. The topic is suitable for ACP and the methods used appear to be satisfactory, although there is a suspicion that concentrations may be underestimated. The language and presentation are clear and the few changes that seem desirable will be listed at the end of this review. This is a good paper and I hope that some of the following comments might be helpful. Where the authors disagree with those comments, I would be happy to enter into a debate.*
Summarising, I recommend publication after the authors have made any changes that appear to be appropriate.

Specific comments

1. ***Abstract:*** *The paragraph that shows no relation between bulk chemical composition and IN concentrations is not very useful in the main text and certainly not in the Abstract. At a site where sea salt particles will be dominant by mass and IN active at temperatures relevant to cloud formation represent about one particle in a million, no other result would be credible.*

   We agree with Dr. Bigg that no direct correlation of the IN composition to the bulk aerosol composition is to be expected due to the rarity of IN in the total aerosol number concentration. The idea of the presented analysis is to use changes in bulk chemical composition of the aerosol as a tracer for different sources. This way, even if only one in a million particles acts as IN we expected to see a proportional increase in concentration with the increase of one of the "tracer materials" for different sources. The finding that this is not the case could indicate that none of these sources (marine, continental, combustion) is the strong supplier of IN active in this temperature range or that IN are distributed independently of their sources main compounds, or that additional conditions affecting the IN content in these sources have to be met. The result confirms that a simple attribution of an ice active particle fraction to a certain source is not feasible.
   The section was removed from the abstract. We decided to keep the part in the main text as we think the implications of comparing to bulk chemistry, support the findings in the section about the air mass origin.

2. ***Introduction:*** *p1., line 21. Is there any firm evidence for concentrations as low as 0.01m^{-3} except in polar regions? In addition to the listed factors influencing the formation of precipitation, updraft velocities and liquid water content could be added. Cloud extent suggests horizontal extent – cloud depth would be better.*

   We could not find published measurements of such low ice crystal concentrations. The reason for the lack of measurements could be the lower detection limit of ~0.1ℓ^{-1} typical for instruments deployed for the task (e.g. 2D-S, Farrington et al., 2016). The description of the visual appearance of ice clouds containing different concentrations of ice crystals is removed from the manuscript because it was not essential.
   Updraft velocities and liquid water content are added to the list of factors influencing properties of glaciating clouds and "cloud extent" has been replaced by "cloud depth".

3. *p2 line 1: What concentration of ice crystals is necessary for ground-based detection? The presence of concentrations of IN in clouds at -10C will usually be similar to that of the concentrations in the air feeding the cloud of all biological IN capable of nucleation at that temperature. The list of potential IN from those sources continues to grow and concentrations must often exceed 100m^{-3}.*

   The lower limit of ice crystal concentrations detectable by ground based Lidar measurements are on the order of ~0.5ℓ^{-1} (A. Ansmann, personal communication). In-situ detectors like the 2D-S are sensitive to concentrations larger than ~0.1ℓ^{-1}.

Sesartic et al. (2013) reviewed measurements of bacteria and fungal spore concentration in the atmosphere and modeled their impact on clouds. Concentrations can be more than $1000m^{-3}$, but most bioaerosol seem to remain near their source of origin. Their presence is strongly coupled to vegetation and their ability to act as IN sometimes needs to be triggered by stress factors. It could be speculated that bacteria and fungi living in a subtropical habitat rarely encounter stress factors that they could counteract by expressing ice nucleating properties.
Because the importance of biological IN is circumstantial in the present study, no changes to the text were made in this context.

4. *P2. line 5. Insert reference here: Hallett and Mossop, Nature 249, 26-28, 1974 who were first to investigate the effect experimentally and define the conditions necessary for it to operate. Line 15 Hallett and Mossop defined that condition in 1974!*

   We now refer to Hallett and Mossop (1974) and added to the manuscript: "Hallett and Mossop (1974) suggested that marine cumuli contain large ice crystal concentrations for dynamical reasons. They usually have higher cloud top temperatures, therefore the contact of ice and supercooled droplet occurs at temperatures favorable for splintering."

5. *P2. Line 19-22. In Bigg's 1973 paper, 3 years of continuous measurements in the Southern Ocean, south Indian Ocean and south Pacific were summarised. The measurements were later used by Schnell and Vali (J. Atmos.Sci., 33(8), 1554-1564, 1976) to show that the measurements revealed a strong dependence on biological productivity. Their interpretation of biological IN rather than dust as the main factors in the measurements is much preferable to Bigg's.*
   This work brings up an important point in relation to your manuscript. The ocean measurements were made with membrane filters that are known to undercount the concentration of IN in a salt-laden atmosphere, yet concentrations in the biologically productive zones were considerably higher than those reported in your manuscript. Chlorophyll measurements in the vicinity of the Cape Verde is reported by Ramos et al. to indicate strong biological activity and significant biological IN should become airborne by bubble bursting. Am I right in assuming that your method only gives a "yes-no" answer for the presence of an active IN at temperature T? If so it needs to be pointed out that actual concentrations may be higher.

   We prefer to include both, the marine biological and the long-range transported desert dust IN interpretation. The evidence for marine biological IN being more important than other substances for ice formation at low supercooling, remains circumstantial. It could also be that biological IN adhere to desert dust and are transported with it (Conen et al., 2011).
   Biological particles exhibit a complex variety of dependencies of their activity as IN on environmental parameters. Research into the importance of organic marine aerosol, released from the sea surface microlayer by bubble bursting, has been conducted by Wilson et al. (2015). They reported a wide range of freezing temperatures, wherein samples from the Atlantic tend to freeze at lower temperatures than Arctic samples. The hypothesis would be, that marine bacteria in the subtropic ocean are less active IN than strains living at higher latitudes. See also comment 8 and 10.
   The measurement method does give a "yes-no" answer for each of the 103 subsamples. For the interpretation of the measurement, we use a non-stochastic view of the nucleation process. Under this premiss, the assumption that only one IN causes freezing in a freezing droplet, is justified in Vali (1971). A possible effect of long sampling times to deactivate IN by blocking active sites, is already mentioned in the introduction. We are not aware of other reasons why the method would under predict concentrations. No changes were made to the text.

6. *p.3 lines 3-4. A spectacular example of the changes in IN concentrations that can occur was published in J. Meteorol. 15, 561-562, 1958. This was later interpreted to be due to related to enhanced biological populations resulting from heavy rain, with a proportion becoming airborne. (Atmos. Chem. Phys., 15, 2313-2326, 2015).*

   The 20-30 day change in IN concentration following intense rainfall discussed in Bigg (1958); Bigg et al. (2015) is a spectacular example for the importance of biological IN under certain conditions, at places close to their origin. The fluctuations we refereed to in the manuscript are on shorter timescales. In Bigg (1961) you refereed to these events as "sudden onset storms". To include both, we changed the sentence to: "The concentration of potential IN measured

at a certain location and temperature can undergo large changes on timescales of days (Bigg, 1958) or hours (Bigg, 1961)." On Cape Verde rain events seem to be followed by prolonged low IN concentrations (at -8 °C). See Fig. 1 for a time series including precipitation events. Rain washes the IN out. This could indicate that biological IN active at this temperature are transported over long distances, eventually attached to larger particles like desert dust.

7. *p.3 line 11-12. Long sampling times aren't necessary. Membrane filters with pore sizes 0.45μm or larger can be sampled at >10l/min but sampling >300l leads to serious undercounts. For long-term measurements or simultaneous measurements at many sites, sampling at 300l/day avoided logistic difficulties but averaged out any short-term fluctuations.*

   The filter used in this study were collected with the intention to investigate the chemical composition of the aerosol at Cape Verde. We reused them to find out about the IN concentration. The sampling time of these filters was 1-3 days and before the time consuming IN-measurements, we estimated the concentration range for which the method would be sensitive to if we took subsamples of different sizes. The calculation showed that thanks to the long sampling time (large sampled air volume) we could use the smallest practical subsample size. This was important to minimize the background from the filter material of the subsample. Much shorter sampling times would not have allowed to use these filters for the measurement of IN concentrations. We agree that, when collecting filters with the purpose to measure IN concentrations, it would be better using the shortest possible sampling interval to reveal short-term fluctuations. We rephrased the sentence. "To measure the low concentration of IN, able to initiate immersion freezing above -20 °C, it is necessary to collect particles from a large enough air volume. The time required to collect this volume depends on the sample flow through the filter, which is influenced by its type (fiber or membrane), fiber or pore size, and the capacity of the pump. Long sampling times may be necessary."

8. *p3. Line 14. Reduction of the RH in the vicinity of a hygroscopic particle is a major factor. Allowing hygroscopic material from a $1m^3$ sample on a membrane filter to diffuse into an underlying wet filter, then drying the top filter and processing it, results in an IN count more than a factor of 2 higher than on a simultaneously sampled filter kept dry. (This work has not been published – use the information if you want to).*

   For the analysis of the filter samples we assume the IN are acting in the immersion freezing mode, residing in droplets. The method consists of immersing pieces of the filter in water drops and observe the freezing of those. The method is immune to water-vapour depletion effects and the water drops are large enough to prevent freezing point depression by the dissolved sea salt within the sensitivity of the temperature measurement. No changes were made to the text in this context.

9. *p.5, line2. At first I didn't understand this as all particles capable of forming IN at temperatures warmer than the test temperature will be activated. Does the answer lie in comment 5?*

   With decreasing temperatures the change in activity of single substances is steeper than the observed change in IN concentration. Consequently substances contribute only at high activity to the ambient IN spectrum (see response to comment 2 of Reviewer 2 and Appendix A).

10. *Figure 2. According to Ramos et al. chlorophyll is a maximum at the end of the year at Cape Verde. I don't see much evidence of a corresponding change in the -8C figure. As it is an important point in determining whether biological IN are effective at the site, running means of about 9 measurements at -8C shown on a diagram with a more extended scale might help. This procedure would be useful in reinforcing the surprising statement in lines 6 and 7 on p.8.*

    Concentration data at -8 °C for the years 2011, 2012 and 5 months of 2013 are shown in Fig. 1. The data show no clear signal corresponding to the maximum in chlorophyll, Ramos et al. found in Nov. and Feb. The increased IN-concentrations in Feb. 2011 and 2013 occurred several days after dust events. It has been reported that bacteria express ice nucleating properties as a response to environmental stresses. It can be speculated that during times of high biological productivity, bacteria have a good time as well and don't need to express ice nucleating properties.

    We didn't include Fig. 1 into the manuscript and no changes were made to the text.

11. *Bulk chemical composition. See comment 1.*

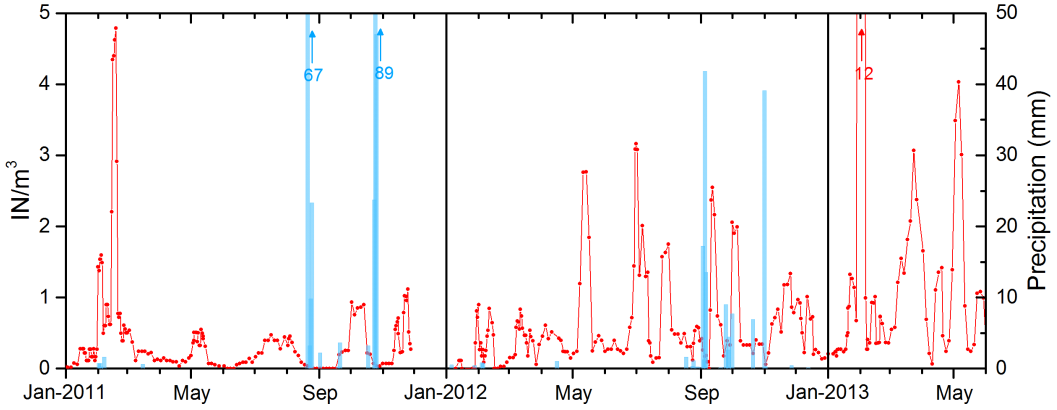

Figure 1. Time series of IN concentration at -8 °C is shown in red. Daily precipitation amount (right hand axis) is shown as blue bars.

We prefer to keep the section in the manuscript. See comment 1.

12. *Air mass origin. How many cases were involved in each of the 7 categories of figure 4? The sporadic rainy season from August-September would probably lead to much deeper atmospheric mixing at times and occasional scavenging of aerosol. How reliable are the 10-day trajectories to the site during that period?*

    The individual data points composing the box plots are shown on the left hand site of each box. This data points are color coded according to the quarter of the year in which they are observed. The number of individual data points is given in Tab. 1.

Table 1. Number of cases per air mass category. Categories are: 1. African coastal, 2. Atlantic marine, 3. North American, marine and coastal, 4. North American and marine, 5. dust and Europe, 6. dust (e.g. Saharan and Sahel region), 7. coastal and Europe.

Temperature	1.	2.	3.	4.	5.	6.	7.
-8 °C	74	11	19	40	47	55	30
-12 °C	111	12	28	54	76	93	44
-16 °C	109	13	28	51	73	92	41

How efficient wet deposition removes aerosol depends on rain intensity, raindrop size, aerosol size (Jung et al., 2013). The NAME model should be able to detect any deep atmospheric mixing (the UM met data should be able to track these events). Rainfall is a rare event on the Cape Verde island and often of very light intensity. The NAME model did the simulation for an inert tracer so as to detect the overall air mass transport, rather than act as a chemical transport model that tracks aerosol movement. An analysis separating the rainy season as suggested in comment 15 showed that IN concentrations do not generally change in this period.

13. *Frequency distribution, p.9 lines 7-9. Size distribution of particles produced by a common method frequently have a log-normal size distribution and this can be expected from a local source. An alternative to Ott's random dilution hypothesis might simply be preservation of the original distribution during transport.*

    If the size distribution is preserved, then we would not expect an effect on the concentration of IN over time. The change in concentration can have two causes. A change in the ice nucleation activity of the aerosol (e.g. due to size) or a change

in abundance. The change in activity with particle size can account for up to two orders of magnitude in concentration (cf. Fig.1 in the response to Reviewer 2). A connection of changes in the size distribution with abundance seems plausible e.g. lower wind speed, transporting less and smaller particles. However, if this would lead to a log-normal frequency distribution is unclear. It could be an additional factor besides random dilution that causes the large variation in IN-concentration. At this point, Ott's hypothesis seems the simplest explanation for the observed log-normal distribution. No changes were made to the text in this context.

14. *p.10, line 6. What is the minimum concentration of ice crystals needed for the lidar observations to detect them? It might be better to replace "to start" with "to be detected".*

    The sentence has been changed accordingly. The lower limit for Lidar to detect ice is in the range of ~$0.5\ell^{-1}$, see comment 3.

15. *Figure B1, p.14. It might be interesting to have a separate diagram for the "rainy" season of August and September and for the period of maximum productivity, October-December.*

    The extended Fig. B1 is shown below. No trend emerges from the rainy or biological active season. Fig. 2 is now included in Appendix B instead of former Fig. B1.

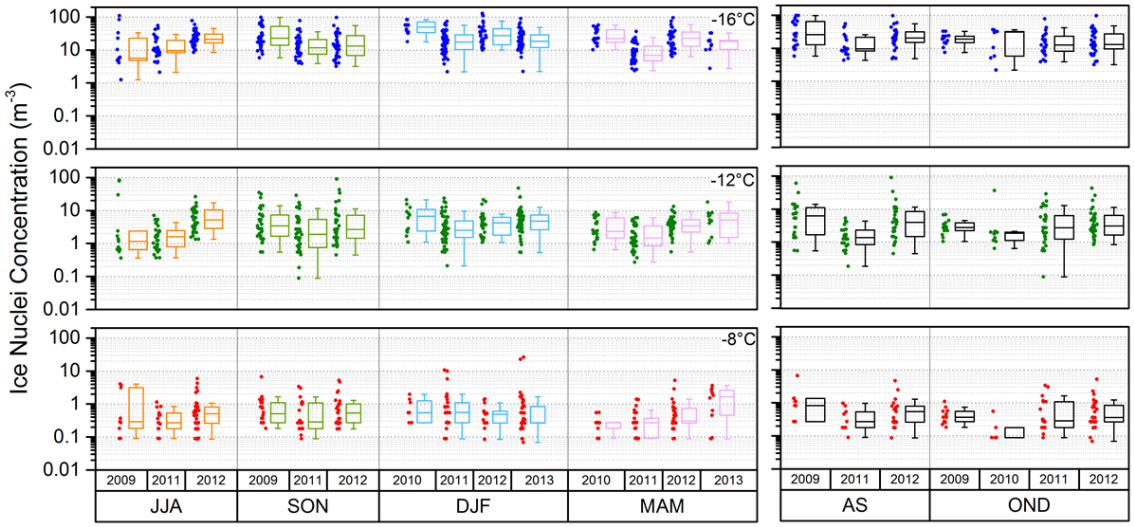

Figure 2. Fig. B1 extended by boxplots of the rainy season (Aug. and Sep.) and the season of high biological productivity (Oct.-Dec.)

Technical corrections

p.1 line 22 : change "cloud extend" to "cloud depth".
p.2 line 26. Change "consist to" to "consist of".
p.5 line 14 "exemplary shown". Change to "exemplified by". Change "year" to "years".
p.8. line 6. Change "effected" to "affected".
p.9 line 20. Change "to identify" to "identification of".
p.10. line 10. Change "to identify" to "in identifying". Line 19: change "must not" to "need not".

Done.

References

Bigg, E. K.: A long period fluctuation in freezing nucleus concentrations, J. Meteorol., 15, 561 – 562, 1958.

Bigg, E. K.: Natural Atmospheric ice nucle, Sci. Prog., 49, 458, 1961.

Bigg, E. K., Soubeyrand, S., and Morris, C. E.: Persistent after-effects of heavy rain on concentrations of ice nuclei and rainfall suggest a biological cause, Atmos. Chem. Phys., 15, 2313 – 2326, doi:10.5194/acp-15-2313-2015, 2015.

Conen, F., Morris, C. E., Leifeld, J., Yakutin, M. V., and Alewell, C.: Biological residues define the ice nucleation properties of soil dust, Atmos. Chem. Phys., 11, 9643 – 9648, doi:10.5194/acp-11-9643-2011, 2011.

Farrington, R. J., Connolly, P. J., Lloyd, G., Bower, K. N., Flynn, M. J., Gallagher, M. W., Field, P. R., Dearden, C., and Choularton, T. W.: Comparing model and measured ice crystal concentrations in orographic clouds during the INUPIAQ campaign, Atmos. Chem. Phys., 16, 4945–4966, doi:10.5194/acp-16-4945-2016, 2016.

Hallett, J. and Mossop, S.: Production of secondary ice particles during the riming process, Nature, 249, 26 – 28, 1974.

Jung, C., Lee, S., Bae, S., and Kim, Y.: Minimum Collection Efficiency Particle Diameter during Precipitation as a Function of Rain Intensity, Aerosol Air Qual. Res., 13, doi:10.4209/aaqr.2012.09.0255, 2013.

Sesartic, A., Lohmann, U., and Storelvmo, T.: Modelling the impact of fungal spore ice nuclei on clouds and precipitation, Environ. Res. Lett., 8, 4029, 2013.

Vali, G.: Quantitative Evaluation of Experimental Results on the Heterogeneous Freezing Nucleation of Supercooled Liquids, J. Atmos. Sci., 28, 402 – 409, doi:10.1175/1520-0469(1971)028<0402:QEOERA>2.0.CO;2, 1971.

Wilson, T., Ladino, L., Alpert, P., Breckels, M., Brooks, I., Browse, J., Burrows, S., Carslaw, K., Huffman, J., Judd, C., Kilthau, W., Mason, R., McFiggans, G., Miller, L., Najera, J., Polishchuk, E., Rae, S., Schiller, C., Meng, S., Vergara-Temprado, J., Whale, T., Wong, J., Wurl, O., Yakobi-Hancock, J., Abbatt, J., Aller, J., Bertram, A., Knopf, D., and Murray, B.: A marine biogenic source of atmospheric ice-nucleating particles, Nature, 525, 234 – 238, doi:10.1038/nature14986, 2015.