Answer to comment of Reviewer 1

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We thank the comments of the Reviewer 1 (#C8895), which definitively help to improve the manuscript. We specially thank the comments in which proposed us to determined new parameters. It really encouraged us to do new calculations for performing a more detailed study. Below we give a detailed reply to each question.

REFEREE: p.24128 l. 2 “free troposphere” .Is there actually proof for this claim? Other than being on a mountain, has any research been done on the subject? Based on the results it would seem that all one observes is fed by the PBL; is it then justified to speak of the free troposphere? Are any data available that could serve as a metric to quantify the PBL influence?

AUTHOR: We fully agree with the referee comment. In fact the title of our previous publication about nano-particles in Izaña made reference to the upward transport from the boundary layer (Atmospheric nanoparticle observations in the low free troposphere during upward orographic flows at Izaña Mountain Observatory; *http://www.atmos-chem-phys.net/9/6319/2009/acp-9-6319-2009.pdf*). Moreover, the results of this study demonstrate that events are cause by transport from boundary layer. In section 3.1 we clearly interpreted the daily evolution of primary reactive gases and particle number concentrations in terms of transport from the boundary layer. This has been checked in the whole text of the revised manuscript. Moreover, the title will be reworded.

REFEREE: l. 25 “The growth of nucleated molecules”. This seems an odd statement and should be reconsidered. Molecules typically don’t grow in nucleation (even if some liquid-phase chemistry might occur at some point).

AUTHOR: We agree with the referee, this is important from a conceptual point of view. We have reworded using the term terminology of the cited paper (Kulmala, 2003): “The growth of nucleated clusters is an important source of atmospheric aerosols".

REFEREE: p. 24129. l. 2f “stable clusters (>2nm size)”. If memory serves correctly, the so-called cluster band measured f.ex. by AIS and NAIS resides BELOW 2nm, not above.

AUTHOR: Yes, we agree. This was a typing error. Thanks!!

REFEREE: l. 7 “For the first phase, or nucleation, the most studied mechanisms are […]”. This list seems somewhat outdated. It is true that these processes have been studied a lot, but mostly to the effect that they cannot explain atmospheric nucleation. What about more recent models (kinetic nucleation, cluster activation)?

AUTHOR: We agree. This part of the introduction have been reworded making reference to more recent studies.

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- Metzger, A., Verheggen, B., Dommen, J., Duplissy, J., Prevot, A. S. H., Weingartner, E., Riipinen, I., Kulmala, M., Spracklen, D. V., Carslaw, K. S., and Baltensperger, U.: Evidence for the role of organics in aerosol particle formation under atmospheric conditions, Proc. Natl. Acad. Sci. USA, 107, 6646–6651, \doi{10.1073/pnas.0911330107}, 2010.
- Kulmala, M., Kontkanen, J., Junninen, H., Lehtipalo, K., Manninen, H. E., Nieminen, T., Petäjä, T., Sipilä, M., Schobesberger, S., Rantala, P., Franchin, A., Jokinen, T., Järvinen, E., Äijälä, M., Kangasluoma, J., Hakala, J., Aalto, P. P., Paasonen, P., Mikkilä, J., Vanhanen, J., Aalto, J., Hakola, H., Makkonen, U., Ruuskanen, T., Mauldin, R. L., Duplissy, J., Vehkamäki, H., Bäck, J., Kortelainen, A., Riipinen, I., Kurtén, T., Johnston, M. V., Smith, J. N., Ehn, M., Mentel, T. F., Lehtinen, K. E. J., Laaksonen, A., Kerminen, V.-M., and Worsnop, D. R.: Direct observations of atmospheric aerosol nucleation, Science, 339, 943–946, doi:10.1126/science.1227385, 2013.

REFEREE: l. 20 “thermodynamic processes (e.g. T and RH)” . Are T and RH processes? Some would simply call them conditions.

AUTHOR: We agree. It was reworded as: "thermodynamic variables (e.g. T and RH)"

REFEREE: p. 24132. l. 24ff “Dust concentrations were measured by combining two techniques: […]”. But is this information actually used to any meaningful extent? See later comments.

AUTHOR: APS and filter data were used to evaluate the role of dust in the formation and growth rates. Correlations between these variables were represented to observe if dust may influence on FR and GR. Following your suggestions, in the revised version of the manuscript, we use APS data to calculate condensation sink. “Dust concentrations were measured by combining two techniques: […]” in section 2.2.2 was reworded as: "Because Izaña is within the Saharan Air Layer in summer, the measurement program includes (1) size distributions with an Aerodynamic Particle Sizer (APS, TSI™) for the range 0.7 to 20 μm aerodynamic diameter (~ 0.7 to 12 μm geometric diameter for dust) and (2) dust concentrations with the methods described by Rodríguez et al., 2011".

REFEREE: p. 24133 **.** l. 22 “Plots with “5 min time resolution 3-D – dN/dlog” data” **.** All these plots (figs. 3, 4, 5) should be presented with a logarithmic colour scale (as everybody else does). The current presentation basically hides all the background aerosol and is thus quite misleading**.** Not even Antarctica is as clean as these figures suggest. As a side note, what does dN/dlog mean? (also, lose the quotation marks here and in many other places.)

AUTHOR: We agree with the comments. The logarithmic colour scale of the plots has been changed. This, dN/dlog, is a typing error; we reworded it as dN/dlogDp (number size distribution).

REFEREE: p. 24134. l. 12f “Kulmala et al., 2004” **.** Kulmala et al. have published a comprehensive cookbook on particle formation measurements and analysis just last year it seems odd to refer to old (and obsolete) sources without citing very good reasons. In any case, the chosen approach seems rather coarse and the analysis too shallow. Where is the loss correction (coagulation, growth)? Why not determine J10 and then extrapolate J2? What about

vapour concentrations? With smps+aps, the condensation sink could be determined. What about using a sulphuric acid proxy to compare to the vapour estimates? Why not use the data to its full potential? (see kulmala et al. 2012)

AUTHOR: Thanks for this comments. It really encouraged us to do most of these determinations. In the revised version of the manuscript, data are used to all their potential.
(a) With the new paper of Kulmala (2012, 'Measurement of the nucleation of atmospheric aerosol particles, Nat. Protoc., 7, 1651–1667, 2012'), the 'cookbook', we followed most of the recommendations. (b) Following the recommendation of Kulmala et al (2012) corrections for the Formation Rate were introduced (Condensation and Coagulation sink were calculated). (c) We decided do not extrapolate to J2. That particle size is out of the range we usually measure. We do not have data on data range and experience to be sure about the accuracy. (d) We agree with your comment about using data to its full potential. Condensation sink was calculated for event and not event days (using SMPS and APS). Condensation sink calculated for the size range of the SMPS was compared with the CS calculated with the size range covered with both SMPS+APS for studying how coarse (Saharan dust) particles may contribute to vapours condensation. Condensable vapours were estimated and its source rate were determined. An approximation to acid sulphuric proxy was carried out. New calculations (discussion and figures) were inserted in the revised version of the manuscript.

REFEREE: p. 24136**.** l. 18ff "Previous studies at Mauna Loa and Izaña concluded that NPF is favoured in the boundary layer to the free troposphere transition region" . And that's why the talk of free troposphere is misleading (see comment somewhere above).

AUTHOR: As already said above we fully agree. Along the manuscript we have perform small change in order to let clear that the NPF is linked to the arrival of precursors from the boundary layer due to orographic upward flows.

REFEREE: p. 24137. l. 16f "In these months, 50 to 75 events per month were observed during the 4 study years (Fig. 6b)". Useless repetition and somewhat confusing.

AUTHOR: We agree with this comment. We changed both the caption of Fig 6 and the sentence. The caption of Fig 6 was reworded as: "Fig. 6. Number of days per month (%) in which each event (Ia, Ib, II, III, Undefined, No Event and Bad Data) was observed (A, and C-E). Accumulated number of days (for the period 2008–2012) in which an event (Ia, Ib and II) was observed in each month (B)".

REFEREE: p. 24140**.** l. 7ff “The low wind speed ratios observed during the formation and growth steps (0.6) indicate that these events occur under low synoptic wind speed conditions.”. That’s the observation. Where are interpretation and conclusion?

AUTHOR: Interpretation and conclusion. This low ratio indicates that formation and growth steps occur under low synoptic wind speed conditions. Low wind speeds hinder dilution and allow the vapours reach saturation conditions. This is described in 24141 L5-6.

When presenting data analysis, Table 4 was used just for describing the context in which the events occurs. Interpretations are performed (24141 L5-6), providing more details, when describing Fig 9.

REFEREE: p. 24141**.** l. 3 “organic compounds”. Care to speculate what these could be and what the sources are, considering the location of the research station? Has any research been done in that direction?

AUTHOR: Unfortunately research on this direction has not been performed in neither in Izaña or Tenerife. For this reason we simply reminded that, in general terms, organic compounds may contribute. This is true for Izaña (located at 2400 meters height) given that a forest of pines (*Pinus canariensis Chr, Sm. ex DC*) develops in the island between 600 and 2000 m.a.s.l. (see details in Rodriguez et al., 2009).

REFEREE: l. 5ff “This evidences how dilution of the gas phase precursors (by winds) makes it difficult to reach the saturation conditions necessary for the change of phase. Observe that FR >1.2 cm3s-1 are observed when wind speed is <= 3.5 ms-1.”. Let’s consider this statement for a moment. Are you observing nucleation in the FT, or are you indeed showing that nucleation in the FT doesn’t occur without PBL influence?

AUTHOR: We fully agree with this comment. As we said above, we have reworded some sentences of the manuscript to make clear that NPF events occur when air from the boundary layer reaches the low free troposphere.

REFEREE: p. 24142**.** l. 5 “This suggests that the availability of this precursor influence the length of the banana type events.”. It might suggest that, looking at averages over many hours. However, it most likely means only that an air mass change occurred half-way through the event.
It would be more interesting to investigate wind conditions during the short events. Also it would be interesting to know how the short events compare to the beginning of the long events, i.e. compare them only for the time the short events last.

AUTHOR: That is exactly what we mean, wind variability influence on SO_2 concentrations and consequently on the length of the events. That is demonstrated in the data plotted in Fig 10 and discussed in this section. This is corroborated in the analysis of many events (examples were not included in the manuscript for the sake of brevity).

REFEREE: l. 7ff “The availability of this precursor seems to be conditioned by wind speed, observe how wind speed decreases throughout the sequence of episodes no event, III (burst) and banana types II and I (Fig. 10c).”. Same as above: what does this say about the FT business?

AUTHOR: We fully agree with this comment. As we said above, we have reworded some sentences of the manuscript to let clear that NPF events occur when air from the boundary layer reaches the low free troposphere.

REFEREE: p. 24143**.** l. 11f “This indicates that the presence of dust may influence the year to year variability in the NPF frequency.” . Wouldn’t it be nice to quantify this somehow? You claim to have filter and aps data, yet none of it is used. Ok, there is fig. 9 (which really yields minimal information). But what about actual numbers? The condensational sink is xx% bigger, therefore we have yy% less nucleation.

AUTHOR: As said above, and following your proposal, we studied NPF by new parameters. New results shows that the condensation sink CS experience just an 8% increment when including the APS data. Moreover, the relationship between CS and GR does not change significantly when including the APS data in the CS. In contrast, the formation rates shows a significant negative correlation with dust concentrations (Fig. 9A3). This suggests that Saharan

dust may exert a more important role scavenging newly formed nano-particles than sulphuric acid vapours. These findings have been included in the revised version of the manuscript.

REFEREE: p. 24144. [the whole business with the two nucleation modes]. This is highly interesting, yet the analysis falls somewhat short of the possibilities. Why are you looking only at apr09 to aug10? How do those bananas look like? Do both nucl. modes appear at the same time? What about air mass movements during those days? Is it possible that two air masses converge around the measurement site? Is it possible that one nucleation mode originates from the PBL, while the second one springs into existence when PBL air enters the FT? an analysis of the timeline could give some hints.

AUTHOR: We agree with the referee that this is really a very interesting finding. The full comprehension and characterization of these events will require a deeper analysis that will probably be produced in a further study specific for the banana type two nucleation modes events. In the current study we simply aimed to introduce these events, to perform a basic description to show that there are NFP events in which two modes are present and that they should be characterised for properly estimating the formation and growth rates. We think that, even if more questions arise about these events, it may be of interest for other scientists to know about these episodes for studying if they are observed in other sites. Because the data analysis requires a long process, based on log normal fittings, we decided to illustrate the events with just a year period, there is no specific reason to select the period apr09 to aug10. In principle, we do not see any visual difference between 1 or two modes banana type events. The relationship between the two modes changes from one event to other, they do not necessary appears at the same time; in many cases both grow at different rates, whereas in other cases just one of the mode grow. It is possible that these events could be related with convergence of air masses at the mountain top measurements site (may be convergence at the mountain top due to upslope winds in the northern and southern slope of the Island). Because a proper analysis of the air masses dynamic will require high resolution modelling, it is out of the scope of this study, but will be considered for further analysis of these events.

REFEREE: p. 24159, fig 3 . Colour scale in log
AUTHOR: Done.

REFEREE: p. 24160, fig 4. Colour scale in log
AUTHOR: Done.

REFEREE: p. 24161, fig 5. A: color scale in log . E: the wind direction plot looks funny. And is rather not very instructive. Think of a better format.

AUTHOR: Fig5A was modified as suggested (log scale). About wind direction and speed plot (Fig5E), this is a conventional way to represent hourly wind direction, the idea is just to plot how it evolve along the day, other formats (e.g. winds rose) would require several subfigures that would enlarge the plot.

REFEREE: p. 24165, fig 9. This is really just scatter, isn't it? What is the information these figures are supposed to convey?

AUTHOR: We have revised this figure. Plots A, clearly show that FR and GR decrease with wind speed and this provides valuable information about the origin of the vapours (upslope winds rather than advections from in the free troposphere). Plots B, FR vs SO_2 and GR vs SO_2, were removed (given that the revised version includes a quantification of the contribution of sulphuric acid to the growth rate). Plot C shows that FR is negatively correlated with dust, whereas this is not observed in the GR. This latter result agrees with the above described new result that shows that the particle size range covered by the APS (which includes the range size of dust particles) have a low contribution to the condensation sink. This is described in the revised version of the manuscript.

REFEREE: p. 24166, fig 10. Terribly busy figure. Needs remake to actually provide any information. What's with the FR and GR? How does that make sense?

AUTHOR: In this figure mean values (per month) of a set of parameters (Nnuc, SO_2, wind speed, NO_x, temperature, RH, global radiation, UV-A and UV-B radiation) are to plot during the different type of events (Ia, Ib, II, III, no event) are compared with the monthly mean value. For the case Ia event, the values in the Fr and GR are included. The aim is to identify the context in which the events occur (described in the text). In our opinion findings are important (discussed in former section 3.4.2). Subplots of radiation were removed in the revised version. For simplicity, the same label is used for each event in all plots.

Summary

Major complaints:
- Too much reporting, too little interpretation.
- Good data set, not squeezed enough for information.
- Figures!
- English language (e.g. often growth where should be grow(s))
- Get rid of “”, only justified for the banana since it is an informal term. otherwise disturbing. E.g. p. 24129 l.4 clusters are “activated”. What do those “” mean? Aren’t clusters really activated? What do they do then?

The manuscript still needs some thinking and more in-depth analysis.

References
Kulmala et al. (2012):
http://www.nature.com/nprot/journal/v7/n9/full/nprot.2012.091.html

AUTHORS:

Thanks for the summary. All these criticisms have been taken into account when preparing the revised version of the manuscript. In order to avoid too much reporting and low interpretation, we have summarised many sections, and have focused just on interpret the key issues. Data analysis in the revised version, includes a number of new determinations that have allowed to assess several new issues, such as the contribution of sulphuric acid to growth rate, or the role of Saharan dust to growth rates. Writing also revised.