<u>Response to Reviewer 1</u> on "Investigation of New Particle Formation mechanisms and aerosol processes at the Marambio Station, Antarctic Peninsula" by Quéléver et al. submitted to ACP Discussions

"The authors present a very significant work observing the prevalence of New Particle Formation (NPF) events (~ 40%) for the later 35 days of austral summer in 2018 (15 January-25 February) at the Marambio research station (64°15'S–56°38'W), located North of the Polar region of Antarctic Peninsula, which has minimal anthropogenic influences. This manuscript characterizes NPF events, formation pathways leading to NPF, and growth parameters from reported observations of both neutral (iodic acid (IA), sulfuric acid (SA) and methane sulfonic acid (MSA)) and charged (anion clusters of dimethylamine (DMA)-bisulphate (2SA) as well as numerous ammonium-(bi)sulfate clusters) aerosol precursor molecules with their, number concentrations and size distributions. The role of abundant biogenic precursors (MSA) to aerosol formation via ocean-ice-land interactions in NPF events enhanced by optimal meteorology in austral summer for the northern Antarctic Marambio site is highlighted as well. Various insights were confirmed with past observations and some novel insights (qualitative, if not quantitative yet) were made regarding NPF pathways in Antarctic region: SA-MSA found to be more significant contributor in NPF event days compared to IA (low concentrations in this location and austral summer study period), and SA-DMA clusters being 1000x more efficient than SA-NH_3 clusters in creating peak nucleation events. However, DMA or amine concentrations are not quantified in this study to assess the exact contribution of DMA or NH_3 or amine concentrations on nucleation. Ion-induced nucleation driven by the ternary system SA-ammonia(-water) is consistent with previous observations reported in literature, but not as significant as that driven by neutral clusters. Caveats like, instrumental artifacts (not sufficiently explaining compositional factors contributing to ion-induced nucleation) and possibility that IA may contribute to NPF in other season(s). These new insights on formation of atmospheric aerosol particles are a crucial step to improve modelling capabilities in predicting the future climate in Earth system/Global-scale climate models. However, as authors suggest mechanisms need to be characterized in more detail as observed in this Antarctic polar region study. Overall, the discussions and conclusions are logically summarized, with effective visualization to present and explain the observational data in this manuscript. I would suggest the following minor and technical comments to be addressed before accepting this manuscript"

We thank the reviewer for the positive feedback and the input that are significantly improving the quality of the manuscript. We assess here the comments points by points and address the changes in the manuscript as tracked changes on Starting with highlighted comments as RC1 (as RC2 and RC3 is addresses the second and third reviewers' comment).

Minor and technical comments (C):

- **C1: Lines 15-18, Page 1:** *"Antarctic primary particles, mainly originating from sea spray or blowing snow, only weakly contribute to total particle number concentration (Lachlan-Cope et al., 2020). Secondary formation of aerosol particles, on the other hand, is believed to be the principal contributor to cloud condensation nuclei (CCN) (Jokinen et al., 2018; Kerminen et al., 2018)."* **Can the authors also quantify the above stated contributions?**

  The modelling study presented by (Merikanto et al., 2009) reports ground level total particle concentration originating from nucleation ranging from 75 % to 100 % in the region of the Antarctic peninsula, which are considerable fractions comprising both NPF mechanism both within and above the boundary layer. At the scale of the Antarctic continent, the total contribution of secondary particle to the total particle number is

estimated to ~ 98 %. More recently, (Tatzelt et al., 2021) reported only small contribution of sea spray aerosols to the CCN concentration and suggested important contributions of potential secondary formed particle around the Antarctic continent. In the previously mentioned modelling study, (Merikanto et al., 2009) suggests a contribution of secondary-sourced particles to CCN of up 87 %, for CCN at 1% supersaturations for the Antarctic continent. This number falls to 69 % considering 0.2 % supersaturation levels.

Despite the mentioned numbers, the source contribution of aerosol particle in Antarctica is highly variable and depends on seasonality and regional landscape (e.g., coastal regions vs continental plateau). Due to the sparsity of in situ measurements in the southern most continent, specific quantification of these contribution is challenging.

A typical example of this is the austral summer seasonal pulse of the particle number concentrations with total concentration increased by a factor of 20 and up to 2 orders of magnitudes (Shaw, 1979; Lachlan-Cope et al., 2020). This seasonal increase of particles was found in connection with increased emissions and/or vertical transport in the warm season and can be explained by new particle formation processes (Lachlan-Cope et al., 2020; Kerminen et al., 2018). Additionally, in situ measurement reports elevated CCN concentration by 11% as a comparison between the high season and the typical Antarctic background aerosol population (Kim et al., 2017).

We want to emphasize that the aforementioned values might not reflect the aerosol population of the Peninsula that has likely a significant influence from a marine environment. Still, we added the following sentences in the manuscript: *"Modelling studies have estimated that primary particles would only contribute to ~2 % of the total particle count that the ground level in Antarctica (Merikanto et al., 2009). Secondary formation of aerosol particles, on the other hand, is believed to be the principal contributor to cloud condensation nuclei (CCN), especially in the Antarctic peninsula were models showed contributions varying from 75% up to 100 % (Jokinen et al., 2018; Kerminen et al., 2018; Merikanto et al., 2009)."*

Finally, our work investigations did not concentrate on CCN but on secondary aerosol formation mechanistic. In our introduction, we aimed at making the point that the NPF in the region has relevance for the climate as well, thus it is important to fully understand secondary aerosol formation processes. Assessing contributions for secondary aerosol to CCN on a more local scale is of great interest and should be assessed in future dedicated work.

- **C2: Line 35, Page 1:** *"At Marambio, ABOA…"*: Please expand the abbreviation of ABOA on first use and also **highlight the location of the Marambio site more clearly in Figure 8 (using a separate legend) or through a separate figure showing the geographical location of the study site.**

  We made the mistake in the writing of the station's name, ABOA was replaced by Aboa as it is not an acronym.
  We added an additional set of figures to locate the study site in the method section as Figure 1 a), b) and c).

- **C3: Lines 39-42, Page 1:** Specify that this period corresponds to 'austral summer' (i.e., summer in the southern hemisphere) here or wherever relevant in the Introduction section: *"At the Antarctic coast, oceanic DMS concentrations are the highest during December to*

January (i.e., austral summer) *with concentration that could exceed 15 nM within the upper 10 m layer of the ocean around the Peninsula compared to a yearly average of ~5 nM (Lana et al., 2011).”*
Changes were made through the introduction.

- **C4: Lines 42-44, Page 1:** Please add suitable citations here: *“DMS has two well-known oxidation products formed from gas-phase reaction with OH radicals: sulfuric acid (H_2SO_4, SA, formed via sulfur dioxide, SO_2) and methane sulfonic acid (CH_3SO_3H, MSA), which can then initiate particle formation and subsequent particle growth.”*
(Barnes et al., 2006; Mardyukov and Schreiner, 2018) were added as suitable citations

- **C5: Lines 51-53, Page 1:** *“At the Antarctic Peninsula, the Weddell Seaside – that undergoes consistent and recurrent phytoplankton bloom episodes every early spring - is a potential reservoir for iodic acid emissions, especially due to slower ice retreat during the summer and colder sea surface temperature than the Southern Ocean on the north and west-side of the peninsula.”* a) **Suggestion to authors to highlight the geographic location of, Weddell Sea and Marambio site (used frequently in this manuscript text) in Figure 8** or in a separate introductory figure, to familiarize the wider atmospheric community readers with the context of this study at the start of the manuscript. (Same comment as on **Line 35, Page 1**) b) **Please add suitable citation(s) that establishes this early spring phytoplankton bloom as a significant source of iodic acid (IA) neutral clusters critical to NPF?**
As answered in Comment 2, we added a set of figures to ease to locate the study site.
(Von Berg et al., 2020; Atkinson et al., 2012; Sipilä et al., 2016) were added as suitable citations.

- **C6: Line 17, Page 5:** Is it supposed to be $N_{Dp'}$ instead of N_{Dp} here? *“N_{Dp} is the number concentration of the pre-existing particles.”*
The correction was made; we thank the reviewer for noticing the typo.

- **C7: Line 49, Page 5:** edit to “area”: *“(especially in the Weddell Sea aera)*”
The correction was made.

- **C8: Lines 1-17, Page 8: Can the authors elucidate more on different possible reasons/hypothesis on why, formation rates for 1.5 nm and 3 nm particles** ($J_{1.5}$ and J_3) **observed in this study are much higher than those observed in past literature**? Also from **Lines 295-301, Page 18 [(7) in Discussion]** is convection (upward/downward draft) a possible explanation? And what further steps say via modelling or analysis can validate that?

Our data shows significantly higher nucleation rate for small particles than referenced in the literature. Although, measurement of the smallest particle in the range of 1-3 nm requires relatively recent measurement techniques - especially using the nCNC (PSM + CPC), it is possible than these values could be impacted by downward transport from the upper tropospheric layer as Merikanto et al. (2009) shows significant contribution of upper tropospheric nucleated particle to the total particle number in antarctica. This is why, we still mentioned it in Point (7) of the discussion. Still, the contribution from boundary layer nucleated particles to the total aerosol number is also seen particularly high in the area from the southern-most sector of South America and at the Antarctic peninsula as well.

Given the fact that we observed NPF already from very small sizes of the nCNC, it is also reasonable to conjecture rather local formation within the boundary layer as well. In this marine-influenced site, it is very likely that there would be much more source of condensing vapours (as those measured and showed in our studies, but potentially other vapours as well) and lower sinks than at the other Antarctic sites (used to compare our observed nucleation rates).

With the lack of information about atmospheric mixing we can hardly make a statement toward one explanation or another and aimed to bring both aspect in the discussion section of our paper. In the manuscript, we considered trajectory altitudes, however it was not precise enough to discuss on the transports of aerosols or their precursors from the upper troposphere to the low boundary layer level. One solution could be to implement a regional chemical transport model adapted to a 3-dimensional system. As an example, the trajectory model for Aerosol Dynamics, gas and particle phase CHEMistry and radiative transfer ADCHEM (e.g., (Roldin et al., 2011)) could be an interesting approach to understand the characteristic, however it required a good understanding of the component of the model such as the chemical characterisation of both aerosol and gas-phase (which we aimed to reach in our study). Still, the origin of many compounds remains unknown and would be rather inaccurate to feed the model, also the big uncertainty on the real quantities of gas emissions, e.g., especially, ammonia would challenge the performance of the model and would likely false its interpretation. Also, this model (as other) was built including VOC emissions and oxidation production. In our case in lack of vegetation, we were unable to as VOC initial concentration or detect HOM reliably, probably due to the absence of targeted instrumentation, on one site, and likely a lack of organic emissions, on the other side. Once again, our study really stresses the need for ammonia and ammonia measurement as well as chemical characterisation of the aerosol phase with better time resolution to fully resolve NPF at this marine/ Antarctic site.

- **C9:** **Section 3.3.1, Page 10-11:** Briefly mention how 'zenith time' differs from noon (midday), at the first use of the term.
  With Zenith time we meant solar noon which for instance is different from 12:00 if located in high latitude. E.g., on 16 February 2018 at Marambio the solar noon was at 13:01.
  We modified the text as "*...peaking radiation time close to noon-time – coinciding with local zenith time, i.e., solar noon that is slightly later than midday - ...*"

- **C10:** **Lines 79-80, Page 12:** Please use consistent time format (12:00 or 24:00 hours, also ensure it throughout the manuscript) : *"...respectively around 05:30 – 06:30 and shortly after 13:00."*
  Here by 05:30 and 06:30 we mean 5.30 am and 6:30 am. We went thought the manuscript and insured a consistent time format.

- **C11:** **Line 185, Page 15 & Line 275, Page 17:** edit typo: *pinguin* to "penguin"
  The correction was made.

- **C12:** **Figure 7:** See if it can be revised with better resolution and labels (axis labels).
  We improved the quality of the figure and changed the labels

Additional references:

Atkinson, H. M., Huang, R. J., Chance, R., Roscoe, H. K., Hughes, C., Davison, B., Schönhardt, A., Mahajan, A. S., Saiz-Lopez, A., Hoffmann, T., and Liss, P. S.: Iodine

emissions from the sea ice of the Weddell Sea, Atmos. Chem. Phys., 12, 11229-11244, 10.5194/acp-12-11229-2012, 2012.

Barnes, I., Hjorth, J., and Mihalopoulos, N.: Dimethyl Sulfide and Dimethyl Sulfoxide and Their Oxidation in the Atmosphere, Chemical Reviews, 106, 940-975, 10.1021/cr020529+, 2006.

Kerminen, V.-M., Chen, X., Vakkari, V., Petäjä, T., Kulmala, M., and Bianchi, F.: Atmospheric new particle formation and growth: review of field observations, Environmental Research Letters, 13, 103003, 10.1088/1748-9326/aadf3c, 2018.

Kim, J., Yoon, Y. J., Gim, Y., Kang, H. J., Choi, J. H., Park, K. T., and Lee, B. Y.: Seasonal variations in physical characteristics of aerosol particles at the King Sejong Station, Antarctic Peninsula, Atmos. Chem. Phys., 17, 12985-12999, 10.5194/acp-17-12985-2017, 2017.

Lachlan-Cope, T., Beddows, D. C. S., Brough, N., Jones, A. E., Harrison, R. M., Lupi, A., Yoon, Y. J., Virkkula, A., and Dall'Osto, M.: On the annual variability of Antarctic aerosol size distributions at Halley Research Station, Atmos. Chem. Phys., 20, 4461-4476, 10.5194/acp-20-4461-2020, 2020.

Mardyukov, A. and Schreiner, P. R.: Atmospherically Relevant Radicals Derived from the Oxidation of Dimethyl Sulfide, Accounts of Chemical Research, 51, 475-483, 10.1021/acs.accounts.7b00536, 2018.

Merikanto, J., Spracklen, D. V., Mann, G. W., Pickering, S. J., and Carslaw, K. S.: Impact of nucleation on global CCN, Atmos. Chem. Phys., 9, 8601-8616, 10.5194/acp-9-8601-2009, 2009.

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Shaw, G. E.: Considerations on the origin and properties of the Antarctic aerosol, Reviews of Geophysics, 17, 1983-1998, 10.1029/RG017i008p01983, 1979.

Sipilä, M., Sarnela, N., Jokinen, T., Henschel, H., Junninen, H., Kontkanen, J., Richters, S., Kangasluoma, J., Franchin, A., Peräkylä, O., Rissanen, M. P., Ehn, M., Vehkamäki, H., Kurten, T., Berndt, T., Petäjä, T., Worsnop, D., Ceburnis, D., Kerminen, V.-M., Kulmala, M., and O'Dowd, C.: Molecular-scale evidence of aerosol particle formation via sequential addition of HIO3, Nature, 537, 532-534, 10.1038/nature19314, 2016.

Tatzelt, C., Henning, S., Welti, A., Baccarini, A., Hartmann, M., Gysel-Beer, M., van Pinxteren, M., Modini, R. L., Schmale, J., and Stratmann, F.: Circum-Antarctic abundance and properties of CCN and INP, Atmos. Chem. Phys. Discuss., 2021, 1-35, 10.5194/acp-2021-700, 2021.

Von Berg, L., Prend, C. J., Campbell, E. C., Mazloff, M. R., Talley, L. D., and Gille, S. T.: Weddell Sea Phytoplankton Blooms Modulated by Sea Ice Variability and Polynya Formation, Geophysical Research Letters, 47, e2020GL087954, 10.1029/2020GL087954, 2020.

<u>Response to Reviewer 2</u> on "Investigation of New Particle Formation mechanisms and aerosol processes at the Marambio Station, Antarctic Peninsula" by Quéléver et al. submitted to ACP Discussions

"Since the second referee report is still missing, I will provide a review so that the open discussion can be closed. Quéléver et al. present an extensive study on new particle formation using data derived during a measurement campaign at Marambio station in 2018. The paper is generally well written and I only have suggestions for minor revisions ".

We thank the journal for reviewing our work in the lack of referee report by the closing date of the discussion period. The constructive comments improve the readability of the manuscript and its quality. We will assess the comments points by points and address the changes in the manuscript as tracked changes on the Word document with highlighted comments as RC2 (as RC1 and RC3 are addressing the first and third reviewers' comment).

General comments:

- " For the next submission it would be good if you could use the ACP manuscript style (line spacing, font size etc. See templates on the ACP webpage: https://www.atmosphericchemistry- and-physics.net/submission.html#manuscriptcomposition). The manuscript was as it was published as preprint quite difficult to read (e.g. too small font).
- Line numbering: On the first few pages you start numbering again with each page, but the second half of the manuscript the numbering continues until the end. Should be done one or the other way, but not mixed. Also here I would suggest to check the ACP guidelines which is the correct style."

We apologize for the poor readability of the manuscript in the pre-print form. The new version of the manuscript is using the ACP manuscript style, line spacing and numbering, font size and style.

Specific comments (SC):

- SC1: "Abstract: The abstract is quite long and my impression is that you go between L36 and L41 too much into detail. I am not sure if it is really necessary to list here all the numbers. I would suggest to rather do that in the summary and shorten these lines here in the abstract." In this version of the abstract, we tried to give as much relevant information as we could overviewing the result of our study. We added values for Js and Growth rates as these are key values that we would ourselves look out in other references to compare with our own result. Also, those number are relatively out of usual, and we judged important to mentioned it as early as in the abstract. However, we do agree that this ended up to a rather long abstract and revised it to be more concise. The new version of the abstract reads as:

"*Understanding chemical processes leading to the formation of atmospheric aerosol particles is crucial to improve our capabilities in predicting the future climate. However, those mechanisms are still inadequately characterized, especially in polar regions. In this study, we report observations of neutral and charged aerosol precursor molecules and chemical clusters composition (qualitatively and quantitatively), as well as air ions and aerosol particle number concentrations and size distributions from the Marambio research station (64□15'S - 56□38'W), located North of the Antarctic Peninsula. We conducted measurements during the austral summer, between 15 January and 25 February 2018. The scope of this study is to characterize New Particle Formation (NPF) event parameters and connect our observations of gas phase compounds with the formation of secondary aerosols to resolve the nucleation mechanisms at the molecular scale. NPF occurred on 40 % of measurement days. All NPF*

events were observed during days with high solar radiation, mostly with above freezing temperatures and, with low relative humidity. The averaged formation rate for 3 nm particles (J_3) was 0.686 cm^{-3} s^{-1} and the average particle growth rate ($GR_{3.8\text{-}12\ nm}$) was 4.2 nm h^{-1}. Analysis of neutral aerosol precursor molecules showed measurable concentrations of iodic acid (IA), sulfuric acid (SA) and methane sulfonic acid (MSA) throughout the entire measurement period with significant increase of MSA and SA concentrations during NPF events. We highlight SA as a key contributor to NPF processes, while IA and MSA would likely only contribute to particle growth. Mechanistically, anion clusters containing ammonia/ dimethylamine (DMA) and SA were identified, suggesting significant concentration ammonia and DMA as well. Those species are likely contributing to NPF events since SA alone is not sufficient to explain observed nucleation rates. Here, we provide evidence of the marine origin of the measured chemical precursors and discuss their potential contribution to the aerosol phase."

- **SC2:** "P3, L21: "sufficiently away from the station……". Add here why. I assume it's to avoid that the measurements are affected by the station. Would be good to clearly state this."
  The clarification was made by adding "…to avoid interfering contamination signals in our measurements".

- **SC3**: "P5, L12: Add here "by collision and coalescence", so that it reads "are lost to pre-existing particles by collision and coalescence" to be more precise."
  Added to the manuscript

- SC4: "P5, L24: Also here I would suggest to add bit more information. How accurate is the approach by Kulmala et al. (2022)?"
  Indeed, this is an important point to think about, and it impacts all NPF studies comparing the J rate from events and non-event days. This is why, our co-authors designed a study in which the GR during non-event days is quantified (Kulmala et al. 2022 – under review). In order to quantify this, non-event days from 4 different locations representative of contrasting environments have been analysed (boreal x 2 , urban background, urban) – up to 10 years of data. The particle number size distributions of the non-event days during each location are averaged and then normalized by size, to enable observing a growing mode and thus calculate a GR. This study is currently under review. The results show in general, that there is not a huge difference between the GR acquired from event days and from non-event days for each of the locations, which makes us believe that using the same GR for events and non-event days in this study is plausible. Given that the data sets included in our current study from Marambio is rather short, we cannot apply the same approach as the aforementioned study. Another interesting observation is that the GR of particles in the size range 3 -7 nm, for example, is similar around the world regardless of the VOC sources (biogenic vs. anthropogenic), see for example Deng et al., 2020, which shows that the GR in Jungfraujoch and Beijing is similar within the uncertainty margins.

  For the purpose of this study, we compared the contribution of each of the terms to the total formation rate, for days with and without a GR measurement. We find, as shown in Fig. RC2-1 below, that regardless of the day, the GR term dominates the formation rate, and assuming the GR to be zero on event days (shaded green areas) will for sure, underestimate the particle formation rates in this size bin.

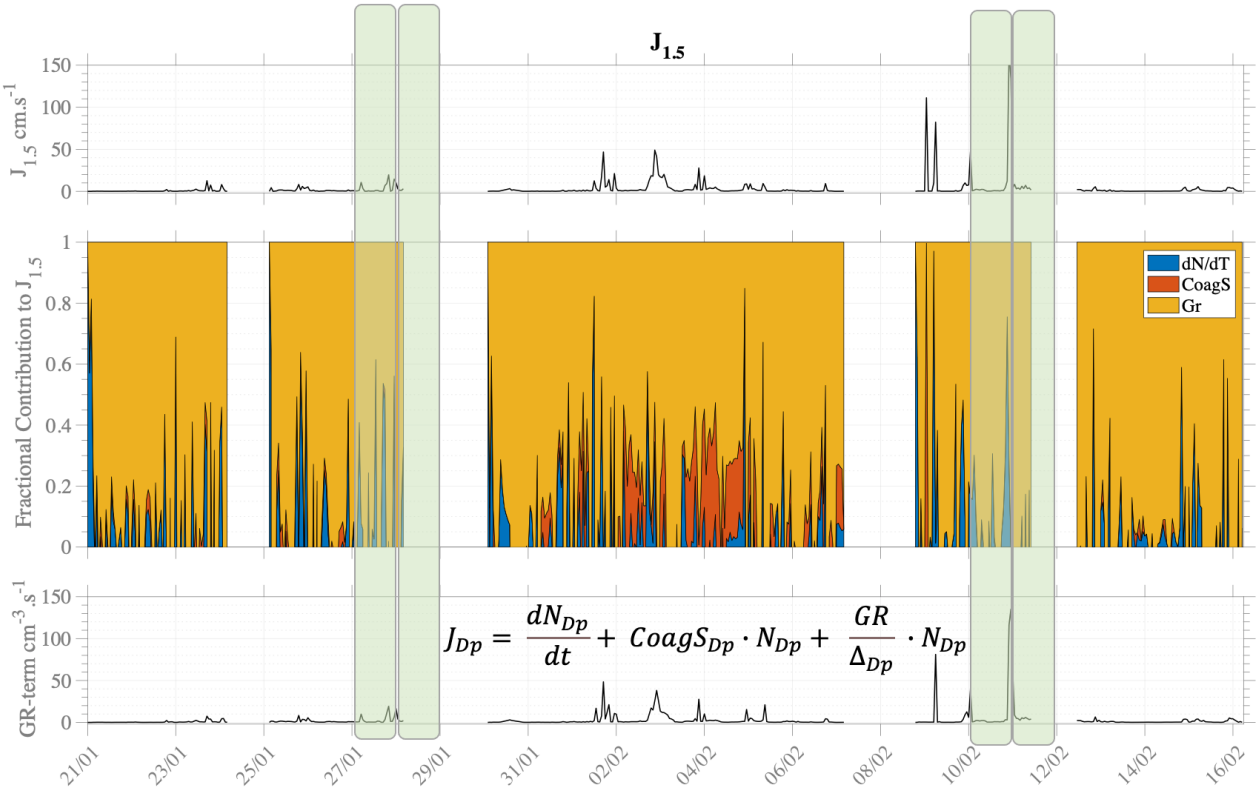

Figure RC2-1: Contribution to nucleation rate for 1.5 neutral particles. Top panel: estimated $J_{1.5}$ ($cm.s^{-1}$). Middle panel: individual contribution to $J_{1.5}$, i.e., dN_{Dp}/ dt (dN/dT), $CoagS_{Dp}$ ($CoagS_{Dp}$. N_{Dp}) and Gr (GR/Δ_{Dp} . N_{Dp}). Bottom panel estimated GR-term ($cm^{-3}.s^{-1}$). We highlighted un green shaded areas event days where GR could not be retrieved by measurements but with extrapolated GR.

- **SC5**: "P7, L28: Why these numbers? Why do you pick these size ranges? An explanation/justification is missing."
  A sentence was added for clarification as: *"These formation rates were calculated for the smallest possible particles but also for bigger sizes to ease the comparison with the literature."*

- **SC6:** "P8, L10-11: Sentence not clear. Please rephrase."
  The text was modified to: **"***Although the formation rates at 1.5 nm are higher than those reported previously, the rates for larger particles (i.e., J_3, J_5, J_{10}) are comparable with the previously reported studies. This discrepancy between formations rates of 1.5 nm particles and of bigger particles could be attributed to favorable nucleation conditions (i.e., sufficient nucleating vapors) but a lack of condensable vapors contributing to particle growth which, in all, reduces the probability of particle survival."*

- **SC7:** "P13, Fig. 6 and according text: This kind of analyses is new for me and I could not follow what actually the mass defect is and would you try to extract from it. Some more motivation/explanations are required."
  We added a clarification on use of such figure and its meaning in term of chemical significance. We modified the text as: "*By showing the divergence of the exact molecular mass of a detected ion in high resolution from its integer mass (i.e., defined as mass defect), a mass defect plot illustrates the mass defect of selected ions (in y-axis) over a studied mass range (in x-axis). Compared to a typical mass spectrum this has the advantage to show the most significant ion population at once, rather independently of signal intensity which is then scaled by the marker size, easing the identification of e.g., clustering mechanisms as further discussed. There, each*

point of the mass defect plot corresponds to a unique atomic composition. An example of such representation is shown in Figure 6, for the study case of 16 February." And added *"exact mass"* in the extra explanation from Fig. 6's legend.

- **SC8:** "P14, L116: I have difficulties to follow you. To what is the "high intensity" referring to?"
  We modified the text for clarity as: *"The numerous clusters containing both sulfuric acid and ammonia, as well as the high intensity of the respective cluster signal (depicted by the marker size in Figure 6) suggest a high concentration of such ion group in the gas-phase which also could indicate an ion induced nucleation driven by the ternary system SA-ammonia(-water)"*.

- **SC9:** "P14, L133: orange dots? To which figure are you referring to? Figure 6?"
  We added the reference to Figure 6.

- **SC10**: "P14, L140: Yellow and green dots? Same here. To which figure are you referring to?"
  We added the reference to Figure 6.

- **SC11:** "P14, L143-147: Sentence not clear. Please rephrase. Maybe it's better to split this sentence into two and check the grammar."
  We rephased the paragraph for clarity as: "However, these ions were only identified as small halogen clusters of low molecular weight and with only low signal intensity. Considering previous observations in the arctic and coastal environment (Sipilä et al, 2016) only shows iodine-related nucleation as successive addition of multiple iodic acid and water group, it is unlikely that IA promotes nucleation either through neutral or ion-induced mechanism at this Antarctic site. "

- **SC12:** "P15, L162: Add "aerosols" so that it reads "sea salt aerosols""?"
  We made the modification

Technical corrections:

TC1: P2, L4: Ipcc → IPCC
TC2: P2, L5: Add a reference?
TC3: P2, L28ff: To my knowledge this are not the only references on these aerosol compositions, thus I would suggest to add "e.g." before the references.
TC4: P3, L1-10: I would suggest to add here references to the respective chapters.
TC5: P5, L49: aera → area
TC6: P7, Fig 2 caption: here "a)" etc in bold face instead of "(a)" as normal text. Be consistent and check ACP guidelines for which way of writing should be used.
TC7: P7, L23: changed → changes
TC8: P7, L25: appear → appeared
TC9: P8, L33: particle → particles
TC10: P11, L39: concentration → concentrations
TC11: P11, L42: add "is" so that it reads "that IA is even anticorrelated with….."
TC12: P11, L46: concentration → concentrations
TC13: P11, L47: concentration → concentrations
TC14: P11, 3.3.2 Header: Study case of → case study on
TC15: P13, L104: noise to signal → signal to noise
TC16: P14, L124: rate → rates
TC17: P14, L134: Add "the" → have shown the possible roles
TC18: P15, L192: Add "the" → in the Supplementary
TC19: P16, L213: concentration → concentrations
TC20: P17, L221: emission → emissions
TC21: P17, L236: study case → case study
TC22: P17, L238: pathway proceeds → pathways proceeds
TC23: P17, L240: than negative → than the negative
TC24: P17, L241: should → could
TC25: P17, L246: in CLOUD-chamber CERN → in the CLOUD-chamber CERN
TC26: P17, L247: Add "that" and use plural: indicate that very high concentrations
TC27: P18, L299: in turns → in turn
TC28: P18, L299: source → sources
TC29: P18, L308: "by key well known"? "key" obsolete?

>> All technical corrections were assessed, and the modification were done in the manuscript and commented as "RC2 >> TC#".

Additional references:

Deng, C., Fu, Y., Dada, L., Yan, C., Cai, R., Yang, D., Zhou, Y., Yin, R., Lu, Y., Li, X., Qiao, X., Fan, X., Nie, W., Kontkanen, J., Kangasluoma, J., Chu, B., Ding, A., Kerminen, V.-M., Paasonen, P., Worsnop, D. R., Bianchi, F., Liu, Y., Zheng, J., Wang, L., Kulmala, M., and Jiang, J.: Seasonal Characteristics of New Particle Formation and Growth in Urban Beijing, Environmental Science & Technology, 54, 8547-8557, 10.1021/acs.est.0c00808, 2020.

Kulmala, M.; Junninen, H.; Dada, D.; Salma, I.; Weidinger, T.; Thén, W.; Vörösmarty, M.; Komsaare, K.; Stolzenburg, D.; Cai, R.; Yan, C.; Li, X.; Deng, C.; Jiang, J.; Petäjä, T.; Nieminen, T.; Kerminen, V-M.: Quiet new particle formation in the atmosphere, Frontiers, *under review*.

<u>Response to Reviewer 3</u> on "Investigation of New Particle Formation mechanisms and aerosol processes at the Marambio Station, Antarctic Peninsula" by Quéléver et al. submitted to ACP Discussions

"I congratulate the authors for the excellent paper, this is a ground breaking paper with lots of excellent measurements. I suggest publication on ACP following minor revisions:

1) The authors seem not to give importance to sympagic sea ice areas as a source of organic nitrogen, as discussed in Dall´Osto et al (2017), Dall´Osto et al (2019) and Rinaldi et al (2020) - given the fact it is a likely source of an important gaseous precursors, it may be worth to bring it into discussions

2) Following this, I am afraid the discussion on point 4 on page 17 is somehow hard to follow. The authors report 13 nucleation events: event 1 and 3 have high GR, event 6,9,10 have very high J. The authors then decide to reports an important case study for the last day (event 12 and 13). However, the discussion of the wind roses is rather weak. The whole interesting event of the case study event (high organic nitrogen) has the opposite wind roses of the claimed pinguin colonies. Also, event 6,7,8,9 have contrasting wind roses - so there must be some influence from the contrary sector (again coincidentally open pack sea ice regions around Marambio).

3)Following the points above, please consider to address other possible source regions. On this regards, you may want to consider to run concentration weitghted trajection for particle formation rate or to see where these particles may originate - the dataset is a brilliant one and worth analyzing a bit more in detail. Also, only one mass defect plot is presented (Figure 6). This is unusual, on previous papers (ie Antarctic measurements in Jokinen et al., 2018) mass defect api-tof measurements for each NPF events were presente - may be worth reporting them in supporting information

I congratulate once again for the brilliant dataset produced and the well presented paper, I hope these modifications can be considered before accepting the paper"

We thank Reviewer 3 for the positive review and feedbacks, and we appreciate the suggested inputs that bring more substance to the paper and significantly improve its quality. Here, we address every comment separately and make the changes in the manuscript as tracked changes on the Word document with highlighted comments as RC3 (as RC1 and RC2 is addressing the first and second reviewers' comments).

Comment 1: The authors seem not to give importance to sympagic sea ice areas as a source of organic nitrogen, as discussed in Dall´Osto et al (2017), Dall´Osto et al (2019) and Rinaldi et al (2020) - given the fact it is a likely source of an important gaseous precursors, it may be worth to bring it into discussions.

This is a very good point. Since we do not have direct measurement of nitrogen containing base species, especially nor of ammonia nor of dimethyl amine, it becomes tricky to properly assess their source. In our paper, we conjectured the penguin colony to be source of possible ammonia due to its vicinity from the measurement site. However, we cannot exclude other sources, and the sea ice could also likely explain a non-negligeable (if not the entire) fraction of nitrogen-containing species as observed from the gas-phase atmospheric composition. We now considered that aspect in the manuscript, in the result section and in the discussion section (cf. § 3.3.4 and § 4 (4)).

Comment 2: Following this, I am afraid the discussion on point 4 on page 17 is somehow hard to follow. The authors report 13 nucleation events: event 1 and 3 have high GR, event 6,9,10 have very high J. The authors then decide to report an important case study for the last day (event 12 and 13). However, the discussion of the wind roses is rather weak. The whole interesting event of the case study event (high organic nitrogen) has the opposite wind roses of the claimed pinguin colonies. Also, event

6,7,8,9 have contrasting wind roses - so there must be some influence from the contrary sector (again coincidentally open pack sea ice regions around Marambio).

We apologies the poor clarity of the discussion. We know the choosing event 12-13 as a case study is not optimum to characterize (all) NPF (occurring) at our site, especially considering the activity and elevated rates for other events. However, due to technical restrictions, we could only speculate on the possible aerosol activity to occur during the operation. Then, we had to manually stop the CI measurement and run the APi-TOF in negative or positive ion mode, inducing interruption of measurement and change on the instrument tunning. As shown in supplementary, we run the negative ion mode simultaneously to an event only 3 times during the whole campaign: on 6.2 (Event #8), on 12.2 (Event #11) and on 16.2 (Events #12-13, as our study case). We did not consider Event #8 worth of interest as (1) the growth - depicted from the shape of the size distribution was interrupted - on many occasions and (2) our ion mode measurement did not catch the start of the event and thus, would not be of use to resolve the initial nucleation formation pathway. On the other hand, Event #11 was suspected to originate from a polluted sector. In this case, we would focus our investigation on anthropogenic mechanisms rather than on the natural processes of the Antarctic peninsula. For these reasons, we decided to highlight Events #12-13 only. Joining comment 1 suggestions, we agree that the penguin-caused ammonia emission conjecture is only a possible cause and particularly in the case of Event #12-13, wind and trajectory analysis do not point toward this explanation. The activity of the sympagic environment could also very likely explain nitrogen-containing compounds although trajectory (specifically from Event #12) seems to originate from an ice-free sector (at least further away from the marginal ice zone). Still, we implemented the mentioned point (4) with this new input that actually drive the discussion to a better understanding of the role Antarctic marine ecosystem on secondary aerosol formation.

Comment 3: **Following the points above, please consider to address other possible source regions. On this regards, you may want to consider to run concentration weitghted trajection for particle formation rate or to see where these particles may originate** - the dataset is a brilliant one and worth analyzing a bit more in detail. Also, only one mass defect plot is presented (Figure 6). This is unusual, on previous papers (ie Antarctic measurements in Jokinen et al., 2018) mass defect api-tof measurements for each NPF events were present - may be worth reporting them in supporting information

In the new version of the manuscript, we now implemented sympagic water as a possible source for ammonia. We are not sure to understand what would be a '*concentration weighted trajection for particle formation rate'*, as suggested. However, we do agree that using a regional transport model, in future measurements, would be extremely useful to assess sources of aerosol and their precursors that would justify the observed high nucleation rate. Furthermore, that would help to determine if nucleation occurs locally, within the boundary layer, or in the upper troposphere, as conjectured in point (7) of the discussion. Concerning the measurement location and environment, the first step would be to get quantitative estimations of ammonia and amines, first, which we are critically missing with this dataset. Then, we should also consider additional source of ammonia such evaporation of ammonia from pre-existing (primary) particle (e.g., sea salt), also depending on particle acidity, which could be assessed in future measurement. With these considerations, additional measurement should be pursued in the future, in the same location, to properly assess all the source for the precursors that contribute to atmospheric nucleation in the regions.

The other point mentioning the one and only mass defect plot is due to the restriction in the operation of the instrument. While e.g., Jokinen et al. had two mass spectrometers, we had only one instrument to run the campaign. As mentioned in our earlier response, catching the ion composition at the start of a nucleation event requires a good prediction of the event to occurs which is not an exact science. Since we could not have simultaneous measurement of both neutral and ion composition, we had to sacrifice one measurement mode over the other in every moment. Event #12 was our best catch as wind came from favourable clean sector and the size distribution showed appearance of small clusters accompanied with continuous growth twice during the day. This, at first, allowed to describe the precursor molecules and then offered the opportunity to characterize the ion composition at the start of the second event.