Answer to the anonymous Reviewer #1

Comments

The manuscript describes measurements of aerosols in tropical regions, on the island of Puerto Rico. The authors had the interesting idea to shed light on the aerosol composition in San Juan, the capital city of Puerto Rico, and try to identify the role of biological aerosols as CCN. The authors used various measurement techniques (WIBS, CCN counter, CPC, Burkard trap) to characterize the aerosols with temporal resolution over a period of 8 days. The main findings of the paper can be summarized as follows:

1. Most aerosol particles in the region are emitted from human emission sources, e.g. cooking and traffic. However, the authors were able to distinguish those from the bioaerosol fraction which was much smaller in number concentration (using fluorescence and fungal morphology data).
2. The daily trend is different for the measurement methods, e.g., different aerosols categories and sizes follow different trends in time (due to primary and secondary aerosol formation mechanisms).
3. High relative humidity triggers bioaerosol emissions, likely fungal spore production.
4. Most fungal spores present in the area are from the phylum Basidiomycota or Ascomycota. Those contribute to the fraction of fluorescent particles detected with WIBS and follow a trend which peaks in the early morning hours.
5. There is no evidence for a correlation between the CCN number concentration and the number of fluorescent bioaerosol. This suggest that bioaerosols emitted from tropical urban areas do not significantly contribute to cloud condensation in tropical clouds.

Although this manuscript provides interesting data and fills a lack of missing information from aerosols and bioaerosols from tropical cities, I would suggest major revisions (mainly in the introduction and in the discussion) before it is considered to be published:

First, I am wondering about the motivation behind the effort to link CCN concentrations to bioaerosol measurements. It is well known, that bioaerosols are generally low in number concentration around the globe (Hoose et al., 2010) and that most aerosol particles can act as CCN when a certain supersaturation is present. The authors make a statement about giant CCN in the introduction (line 78) but do not explain why bioaerosols from the city could be an important source of CCN. I encourage the authors to extent the introduction with a more detailed statement of bioaerosols as CCN.

*Response (refer to the revised manuscript Page No.3 Line No.82 & 91): Agreed that bioaerosols contribute a small fraction (50 Tg $yr^{-1}$) of the total natural global emissions (~2900-13000 Tg $yr^{-1}$) (Hoose et al., 2010; Stocker et al., 2013). However, their mass and number concentrations are site specific and greatly vary depending upon the location and climatic condition (references therein Zhang et al., 2021). In terrestrial ecosystems, bioaerosols constitute a major fraction of total aerosol load. As far as urban and rural atmosphere are concern, bioaerosols of size greater than ~1 µm may account for around 30% (references therein Fröhlich-Nowoisky et al., 2016).*

*There is evidence where bioaerosol may constitute a significant fraction (5-50%) in the urban air (Jaenicke, 2005). The reference to giant CCN has now been expanded upon to explain that giant CCN play a special role in precipitation development because they can form larger droplets that more easily collide and coalesce to form raindrops. Hence, although small in number concentration they make up for in their size and capacity to contribute to early precipitation development.*

*Note: Most of the bioaerosols detected in the city of San Juan originated in the El Yunque rain forest and not in the city itself.*

Second, the paper provides important data about bioaerosols in cities, measured close to emission sources. Tropical island, such as Puerto Rico are remote areas where a large fraction of aerosols comes from local emissions. Thus, it is very important to gain data about local, urban, aerosol compositions in order to make conclusions about human health, environmental and meteorological influence. Although, the authors make a few statements in the manuscript about the importance of their measurements, the paper would strongly benefit if the statements were more emphasized, and the knowledge gap would be identified more clearly in the introduction.

*Response (refer to the revised manuscript Page No.4 Line No.100): The reviewer makes an important point and more background information has been added as well as why our measurement site in a location that is fairly representative of a typical Puerto Rican urban. We do disagree somewhat with the reviewer's comment about a large fraction of the aerosols coming from local emissions. Previous papers (Allan et al., 2008; Raga et al., 2016; Torres Delgado, 2021) have shown that because of the persistent flow of air coming from over the ocean, the largest fraction of aerosols are of marine origin.*

*Puerto Rico is characterized by tropical climate, urban land cover and use, moist soils, unique topography, and dense vegetation. These factors, associated with the easterly trade winds from the East, could influence the concentration of airborne particles, for examples, organic particles, viruses, bacteria, fungi, pollen, etc. (Velázquez-Lozada et al. 2006). Nevertheless, meteorological variables (high humidity and wind speed) are also the important factors, influencing the airborne particle population in the tropics, including rainy seasons. There are various sources of particulate matter degrading the air quality of Puerto Rico, i.e., from industrial activities, anthropogenic inputs, African temperatures dust storms and volcanic eruptions. The urban areas of Puerto Rico are considered developed with industrial growth, most of which is related to pharmaceutical and power generation plant. The power generation plants are responsible for releasing millions of pounds of air pollutant annually (Torro-Heredia et al., 2020). Data show that a large number of organic compounds (e.g., n-alkanes, esters, phthalates, siloxanes, and other) including plasticizer released into the atmosphere which could pose major health threat in this area (Torro-Heredia et al., 2020).*

Third, the authors conclude that the influence of bioaerosols as CCN to be from minor importance. I was wondering if the paper would proliferate when highlighting the health effects which bioaerosol can have in more detail. Are those concentrations of fungal spores comparable to other

cities? How is the concentration in rural and remote areas compared to this study (e.g. compare with El Yunque rainforest data if that is possible)? In addition to that it is well known that bioaerosols, especially fungal spores, are among the most active ice nucleating particles (see e.g. Kunert et al., 2019). Is it possible that those particles do not influence the budget of CCN drastically, but still have an influence on the INP budget? I would encourage the authors to include the role of bioaerosols as INPs in the discussion and to shortly state health effects. This can help transferring the results of the paper into a puzzle piece from the bigger picture of bioaerosols.

*Response (refer to the revised manuscript Page No.27 Line No.590): It is evident that almost all of the fungal spores are released in the El Yunque rain forest (Lewis et al., 2019). Two of the air sampling sites, Pico Del Este (PDE) and Cabezas de San Juan (CSJ) are very similar and very low in the fungal spores (less 5,000 spores/$m^3$). At another sampling site in El Verde (located to the west within El Yunque National Forest), the concentrations increase to 72,000 spores/$m^3$ and are found to have a decreasing gradient of fungal spores towards the Metro Area. For the rest of Puerto Rico, the Central Mountain Range is the other source of fungal spores. Bioaerosols, especially fungal spores, are very good ice nucleating particles (INP); however, except for periods with tropical storms, cloud tops rarely grow higher than the freezing level. As per the reviewer suggestion, we have added the statement (Line No. 599) on the health aspects of bioaerosols*

In summary, the paper provides an interesting insight to a pilot study of aerosol research in a tropical city. I hope that my feedback helps the authors to revise a manuscript with a clearer storyline. Furthermore, the manuscript would benefit from spelling and grammar checks. In terms of further field campaigns, I would encourage the authors to focus not only on CCN, but also on ice nucleation and/or health effects driven by urban bioaerosols.

Specific comments:

19 ... change to are capable of …

*Response: Correction made in the revised manuscript (refer to the revised manuscript Page No.1 Line No.19)*

20 … What's the difference between plant spores and pollen?

*Response: Pollen is essential for sexual reproduction of flowering plants and plants that produce cones. Spores are microscopic propagative bodies, with single nucleus, whose primary function is plant dispersal and reproduction. Spores are produced by "lower" plants which include mosses, liverworts, clubmosses, horsetails and ferns.*

27 … change to a population of 2,448,000 people …

*Response: Correction made in the revised manuscript (refer to the Page No.1 Line No.29)*

68 … change to where vehicular and industrial emissions …

*Response: Correction made in the revised manuscript (refer to the Page No.3 Line No.70)*

69 … The reader might be confused reading about local aerosols and then African dust in the next sentence. Maybe it is important to highlight the long-range transport before explaining African dust as CCN.

*Response: A statement on long range transported aerosol is added in the revised manuscript (refer to the Page No.3 Line No.71)*

*"Apart from this, clouds and rainwater in this region are influenced by long-range transported natural aerosols."*

96 … What meteorological factors? Please elucidate more clearly.

*Response: The meteorological factors (relative humidity and wind speed) were clarified in the revised manuscript (refer to the Page No.5 Line No.124)*

101 … This is an important statement why this time was chosen.

*Response: Thanks to the reviewer for acknowledging the statement*

134 . I was wondering how representative the location of the university is for the city center. Can you make a statement about the areas and the distance to the city center of San Juan in the text?

*Response: A statement is added in the revised manuscript as per reviewer's suggestion (refer to the Page No.6 Line No.164).*

*Moreover, the measurement sites are located at the center of the San Juan city (199 $km^2$), a clear representative of typical urban atmosphere.*

136 … How much rainfall, can you state a yearly average in brackets?

*Response: The annual rainfall recorded in San Juan, Puerto Rico is 4.22±1.3 in. This value is added in the revised manuscript (refer to the Page No.6 Line No.167).*

202 … Are Tryptophane and NADPH the only molecules exited by fluorescent light in that region. Are there other molecules that could potentially also show fluorescence in that area. If yes, please explain in more detail.

*Response (refer to the Page No.8 Line No.243): There could be numerous molecules that can be excited by fluorescent light. For examples molecules such as proteins, large polymers, molecules having conjugated double bonds, heterocyclic aromatic compounds, particularly when nitrogenous substituents are present. Tryptophan is an amino acid that has the highest (~ 90%) fluorescence in the native protein. Nicotinamide Adenine Dinucleotide Phosphate (NAPDH) is one of the major contributors to the fluorescence signal when attached to the protein molecule and is produced widely in the metabolic cell. Other atmospheric relevant fluorophores are Riboflavin, Vitamin A, Vitamin D, Vitamin C, Vitamin B6 compounds, Cellulose, etc., which could be present in the FAPs but their emission and excitation matrix is yet to be conceptualized.*

221 … I have seen that the fluorescence signals alone can be misinterpreted in some cases (see e.g. Savage et al., 2017). Why are you sure that those are specific for bioaerosols in your case?

*Response: There could be a possible interference of the fluorescence signal but we have set a threshold to minimize fluorescence from non-bioaerosols, i.e. organic compounds that can fluoresce, but at an intensity that is typically much lower than bioaerosols. In addition, we observed a relatively strong correlation between the FAP signal and the fungal spores prevalent at the site. Also, the identified fungal spores have a systematic diel pattern, ubiquitous, and are observed to have a considerable population in the San Juan atmosphere based on this study and previously published literature.*

251 … I have seen different forms of writing units in the manuscript (e.g. L $min^{-1}$ or liters of air/min) please make sure that all the units are consistent in the manuscript.

*Response: Thanks to the reviewer for notifying the inconsistency. It is corrected in the revised manuscript(refer to the Page No.10 Line No.297).*

262 … change to … from a weather station that is located around 800 m away from …

*Response: It is corrected in the revised manuscript (refer to the Page No.10 Line No.308).*

264 … I am wondering how 100 m above ground level would correlate with your measurements at ground level? Do you assume the aerosol composition to be the same in those altitudes?

*Response: This will depend upon how well mixed the boundary layer is. In this region, the boundary layer depth is 300-500 m and due to the solar heating it becomes well mixed by late morning. Although usually light, the local winds also contribute to maintaining a mixed layer.*

*Note: Using the Burkard we capture the fungal spores and pollens on the roof of the $10^{th}$ floor building which is about 60m above the ground level*

281 … I was wondering if the number of CCN is always so low compared to CN. Is that also seen in other studies?

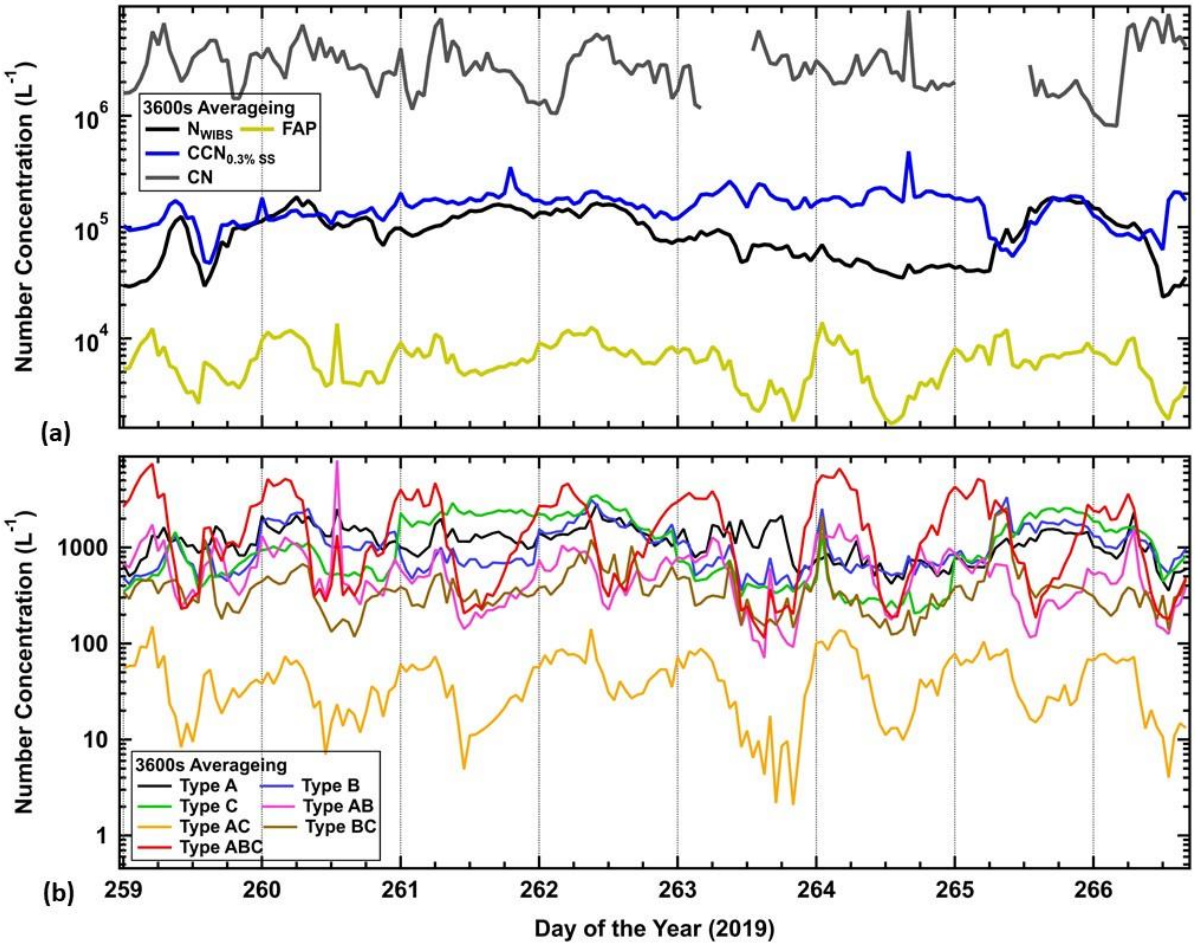
*Response: Yes, Allan et al. (2008) reported similarly low numbers of CCN compared to CN in Puerto Rico.*

285 … This statement assumes that the WIBS and CPC would count the same number concentration for all aerosols above 500 nm. Can that be seen in your data or confirmed with literature?

*Response: Yes, this statement can be confirmed by previously published literature (Healy et al., 2012). However, the counting of the particles by the instruments may differ a little because of the different sampling flow rates used for WIBS (0.3 L $min^{-1}$) and CPC (1 L $min^{-1}$) in this study.*

Figure 2 … I have a hard time seeing the daily trend for CCN and CN. Would it be better observable if one would include vertical lines whenever a day ends in the diagram as a grid?

*Response: As per the suggestion, vertical line as a grid will be added to the figure 2 in the revised manuscript (refer to the Page No.13 Line No.346).*




318 … very interesting finding

*Response: Thanks*

Figure 4 … the letters (a) and (b) are poorly visible

*Response: The letters (a) and (b) were placed outside of the figure 4 in the revised submission (refer to the Page No.17 Line No.390).*

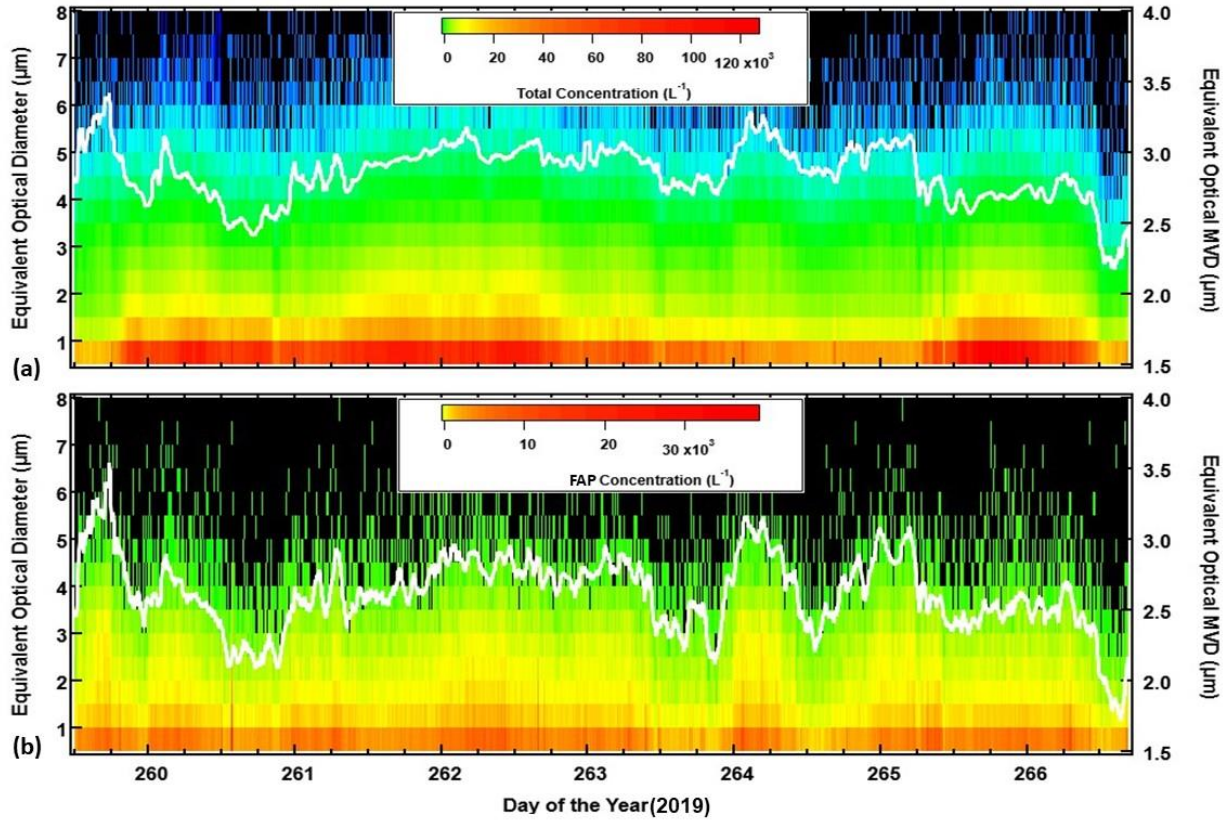
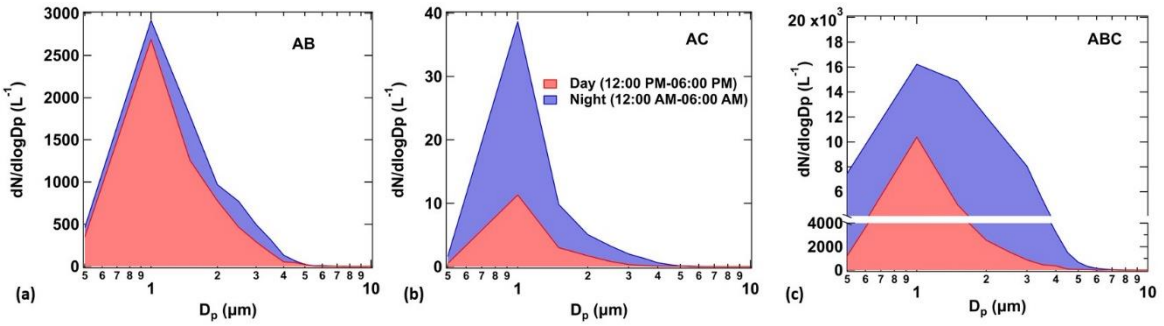



Figure 5 … I am confused by numbers on the y-axis in Figure5(c)

*Response: The y-axis in Figure 5 is corrected in the revised manuscript (refer to the Page No.19 Line No.415).*




409 … The findings about the relative humidity and the fungal spore production are interesting and one of the main findings in this work

*Response: Thanks*

444 … I have a hard time in understanding this paragraph and the conclusions of it. 1. Why is ABC fluorescence related to Basidio & Ascospores? Are those spores simply bigger than the other fungal spores and therefore the fluorescence signal extents the threshold for ABC? 2. Why is the concentration of the spores in WIBS so much higher than from the Burkart trap (Figure 7)? 3. Did you also count bacteria, bacteria agglomerates, and pollen with the Burkart trap samples? If no, why not? If yes, how did they influence the measurements of the fungal spores in the WIBS?

*Response (refer to the Page No.24 Line No.518): (1) Previous studies (Quintero et al., 2010; Rivera-Mariani et al., 2020) reported that the most common fungal genera detected were the Basidiospores and Ascospores in the San Juan atmosphere. Which is also confirmed in this study. The size of the Basidiospores and Ascospores (10-20 um) is usually larger than Aspergillus, Penicillium, and Cladosporium spores. They are release at the early morning hours due to dew point and increase in relative humidity at these hours. Furthermore, we observed a systematic diel pattern in the number concentrations of these fungal spores which is strongly correlated to diel pattern of FAPs detected in ABC channel of WIBS. (2) The difference in number concentrations because both the instruments work on different detection principle and different sample flow rate. Furthermore, the Burkart trap was not located in the same site (18°24'6.4"N, 66°03'6.5"W, 6 m a.m.s.l.) where the WIBS was operational but was installed at a different location (18°23'48" N, 66°4'30" W, 60 m a.m.s.l) in San Juan. (3) Due to their small size the bacteria are not usually counted with the Burkard. Only pollen and fungal spores are enumerated with this air sampler. Pollens are release during late morning and noon. They much lower in concentrations compared to the fungal spores.*

473 … I would be careful with this statement since you are assuming that the measurement techniques cover all particles in the area above 500 nm. Also, this would include that in terms of number concentration the study only looks at 2% of the particles closely. This further implies that bioaerosols are a very low fraction of the total aerosols, yet they are important for health and climate. Maybe the authors can state that more clearly and explain that coarse mode aerosol is typically smaller in number concentration, yet high in mass concentration.

*Response: The suggestion made by the reviewer has been included in the revised manuscript (refer to the Page No.26 Line No.552).*

504 … This is a main message of the paper and should be extended with one or two more sentences of what particles matter for CCN.

*Response: The additional statement is added in the revised manuscript (refer to the Page No.27 Line No.585).*

*This suggests that if FAP are good CCN, they do not contribute significantly to the overall CCN population. It is important to note that contributions to the overall CCN populations depend on particle size, chemical composition, and number concentrations.*

Figure 8 … It is hard to see the trend in the graph with the current color scale. Would it be helpful to consider a logarithmic color scale or other colors?

*Response: The figure is modified with different color scale in the revised manuscript (refer to the Page No.29 Line No.626).*

603 … As far as I know Basidiomycetes and Ascomycetes are not fungal species but more of a phylum.

*Response: Reviewer is thanks for notifying this. The term Basidiomycetes and Ascomycetes changes to Basidiospores and Ascospores in the revised manuscript (refer to the Page No.31 Line No.698).*

References:

Allan, J. D., Baumgardner, D., Raga, G. B., Mayol-Bracero, O. L., Morales-García, F., García-García, F., Montero-Martínez, G., Borrmann, S., Schneider, J., Mertes, S., Walter, S., Gysel, M., Dusek, U., Frank, G. P. and Krämer, M.: Clouds and aerosols in Puerto Rico - A new evaluation, Atmos. Chem. Phys., 8, 1293–1309, doi:10.5194/acp-8-1293-2008, 2008.

Fröhlich-Nowoisky, J., and Coauthors, 2016: Bioaerosols in the Earth system: Climate, health, and ecosystem interactions. Atmos. Res., 182, 346–376, https://doi.org/10.1016/j.atmosres.2016.07.018.

Healy, D. A., D. J. O'Connor, and J. R. Sodeau, 2012: Measurement of the particle counting efficiency of the "Waveband Integrated Bioaerosol Sensor" model number 4 (WIBS-4). J. Aerosol Sci., 47. 94-99, https://doi.org/10.1016/j.jaerosci.2012.01.003.

Hoose, C., J. E. Kristjánsson, and S. M. Burrows. "How important is biological ice nucleation in clouds on a global scale?." *Environmental Research Letters* 5.2 (2010): 024009. *( refer to the revised manuscript Page No.35 Line No.810)*

Jaenicke, R., 2005: Abundance of cellular material and proteins in the atmosphere. Science (80-. )., 308, 73, https://doi.org/10.1126/science.1106335. *( refer to the revised manuscript Page No.35 Line No.817)*

Kunert, A. T., Pöhlker, M. L., Tang, K., Krevert, C. S., Wieder, C., Speth, K. R., ... & Fröhlich-Nowoisky, J. (2019). Macromolecular fungal ice nuclei in Fusarium: effects of physical and chemical processing. Biogeosciences, 16(23), 4647-4659. *( refer to the revised manuscript Page No.36 Line No.827)*

Lewis, L. M., and Coauthors, 2019: Characterizing environmental asthma triggers and healthcare use patterns in Puerto Rico. J. Asthma, ; 57(8): 886–897, https://doi.org/10.1080/02770903.2019.1612907. ( *refer to the revised manuscript Page No.36 Line No.831)*

Quintero, E., Rivera-Mariani, F. and Bolaños-Rosero, B.: Analysis of environmental factors and their effects on fungal spores in the atmosphere of a tropical urban area (San Juan, Puerto Rico), Aerobiologia (Bologna)., 26, 113–124, doi:10.1007/s10453-009-9148-0, 2010.

Raga, G. B., Baumgardner, D. and Mayol-Bracero, O. L.: History of aerosol-cloud interactions derived from observations in mountaintop clouds in Puerto Rico, Aerosol Air Qual. Res., 16, 674–688, doi:10.4209/aaqr.2015.05.0359, 2016.

Rivera-Mariani, F. E., Almaguer, M., Aira, M. J. and Bolaños-Rosero, B.: Comparison of 751 atmospheric fungal spore concentrations between two main cities in the Caribbean basin, P. R. 752 Health Sci. J., 39(3), 235–242, 2020.

Savage, N. J., Krentz, C. E., Könemann, T., Han, T. T., Mainelis, G., Pöhlker, C., & Huffman, J. A. (2017). Systematic characterization and fluorescence threshold strategies for the wideband integrated bioaerosol sensor (WIBS) using size-resolved biological and interfering particles. Atmospheric Measurement Techniques, 10(11), 4279-4302.

Stocker, T.F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex, V., Midgley, P.M., 2013. IPCC, 2013: climate change 2013: the physical science basis. Contribution of working group I to the fifth assessment report of the intergovernmental panel on climate change. IPCC, 2013. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, p. 1535. Stommel, E.W., Fiel*( refer to the revised manuscript Page No.37 Line No.885)*

Torres-Delgado, E., Baumgardner, D., and Mayol-Bracero, O. L.: Measurement Report: Impact of African Aerosol Particles on Cloud Evolution in a Tropical Montane Cloud Forest in the Caribbean, Atmos. Chem. Phys. Discuss. [preprint], https://doi.org/10.5194/acp-2021- 88, in review, 2021.

Toro-Heredia, J., Jirau-Colón, H. and Jiménez-Vélez, B. D.: Linking PM2.5 organic constituents, relative toxicity and health effects in Puerto Rico, Environ. Challenges, 5, 100350, doi:10.1016/j.envc.2021.100350, 2021. *( refer to the revised manuscript Page No.38 Line No. 900)*

Velázquez-Lozada, A., Gonza´lez, J. E., and Winter, A.: Urban heat islands effect analysis for San Juan, Puerto Rico. Atmospheric Environment, 40, 1731–1741, 2006.*( refer to the revised manuscript Page No.38 Line No.913)*

Zhang, M., Khaled, A., Amato, P., Delort, A.-M., and Ervens, B.: Sensitivities to biological aerosol particle properties and ageing processes: potential implications for aerosol–cloud interactions and optical properties, Atmos. Chem. Phys., 21, 3699–3724, https://doi.org/10.5194/acp-21-3699-2021, 2021. *( refer to the revised manuscript Page No.39 Line No.918)*

Reply to Anonymous Referee # 2

Comments

This manuscript reports on results obtained from a pilot study connecting fluoresnce and CCN activity of urban aerosols in San Juan, Puerto Rico, which are interesting.

In the title "Measurement Report" could be removed

***Response:*** *The "Measurement Report" is included in the title as per the Journal instructions to categorize the different manuscript types. We will remove this from the title in the revised manuscript if the editor concurs.*

Sentence structure and grammar needs to be checked, for example, lines 19-20, 275-277, 300-301

***Response:***

*Sentence and grammar structure of the following sentences were changed in the revised manuscript*

*Lines 19-20: Many types of atmospheric aerosols cloud condensation nuclei (CCN) capable of activating as cloud droplets.*

*Changes to (page No.1; Line No.. 19): Many atmospheric aerosols are cloud condensation nuclei (CCN), capable of activating as cloud droplets when the relative humidity exceeds 100%.*

*Lines 275-277: The CN concentrations show systematic, daily trends that reflect the motorized vehicle traffic and nearby residential emissions, the latter mostly from cooking.*

*Changes (page No.11; Line No.. 329) to: The CN concentrations show systematic, daily trends that reflect the emissions from motorized vehicle traffic and nearby residential heating and cooking.*

*Lines 300-301: The CCN concentration shows an increasing trend throughout the day, reaching its maximum around 4 pm, five hours after the peak in the CN.*

*Changes (page No.14; Line No.. 356) to: The CCN measurements show a trend of increasing concentrations early in the morning but does not start its rapid increase until midday peak, then it begins increasing until reaching its peak around 4 pm, five hours after the CN peak. Prior to the afternoon peak, there are smaller peaks that occur at 2 am and 8 am.*

lines 161-162 - explain why absolute size of particles is not a factor in the current analysis

***Response*** *(page No.7; Line No.. 192)**:** The WIBS NEO, like all single particle, optical spectrometers, measures what is designated an "equivalent optical diameter (EOD)" that is defined as the size of a particle scattering the equivalent intensity of light as a spherical particle with known refractive index. Given that bioaerosols, dust and other types of environmental aerosols are not spherical, and that we don't know their refractive index, the geometric size can be estimate to, at best, ±20%, hence relative size is more relevant than absolute size.*

line 168-170 - needs to be rewritten, more clearly

***Response:***

*Line 168-170: A series of heaters are controlled to maintain a temperature gradient from cooler to warmer as the particles move down the chamber. The difference in diffusion rates between heat and water vapor creates a supersaturated environment at the centerline of the cylinder.*

*Changes to (page No.7; Line No.. 204): A series of heaters along the column are controlled to maintain a temperature gradient from cooler to warmer as the particles move down the column. Since water vapor from the wetted column diffuses to the particles faster than the heat, a supersaturated condition is maintained that is determined by the temperature gradient and flow rate.*

line 262 - met parameters are collected at about 800 m away from aerosol measurements, is it a concern? Will the distance between the two measurements infleunce the inferences/results, explain

***Response:*** *Although having meteorological sensors at the site would have been ideal, given the flat terrain and no local sources of heat or humidity, there is no reason to believe that the measurements reported 800 m from the research site would be significantly different from conditions at the site.*

lines 275-280 - comment on the reasons for differences with other results

***Response*** *(page No.11; Line No.. 322)**:** The differences in CN concentrations between the measurements at the university site and previous measurements are related to the geographical locations and average climatic conditions of the other regions discussed. The measurement site (Facundo Bueso) is an urban location influenced by emissions from vehicular traffic, vegetation, and other human activities such as heating and cooking. The CSJ is a remote coastal location where the atmosphere is relatively clean, influenced by marine aerosols or long-range transported aerosols. The PDE is a mountainous region that has a significant influence of aerosol from the nearby vegetation and from particles transported from marine boundary layer.*

line 318 - why ABC type is quite large and dominates the FAPs during a particular time, explain

***Response:*** *Our observation suggested that ABC types are the Basidiomycetes fungal spores which are more frequently released into the atmosphere. The intradiurnal variation is due to the humidity dependent release of the basidiospores (the most common fungal spores in the outdoor air of PR).*

line 328 - any specific reason for the diurnal variaion in FAP size distribution? Is it due to met parameters, explain

*Response: Yes, Met parameters are highly correlated with the number and size of FAP particles. This is clearly indicated in fig 6a and 6b, where nighttime increase of RH (>80%) under low wind speed appear to be related to the released of fungal spore (e.g., Basidiospores and Ascospores) and their growth due to condensation. Other studies at other locations have found similar correlations and the biological community is well aware of the link between spore release and high humidity.*

line 353 - explain why larger increases at EODs larger than 2 µm. Make a conclusion statement.

***Response*** *(page No.31; Line No.. 702)**:** Here larger shifts of EOD refer to ABC type FAP. These are most likely the Basidiospores whose size increases with the increase of RH in the nighttime. Based on the referee's suggestion, We have clarified this statement in the conclusion of the revised version.*

line 371 - explain the role of asphericity, and also its impact on FAP/non-FAP when it is low or high

*Response: The asphericity factor roughly estimates the shape of the particles. In theory, perfectly spherical particles exhibit an AF value of 0, whereas an AF value close to 100 indicates a fiber-like, columnar-shaped particle. We observed higher asphericity for non-FAP than the FAPs indicating the non-FAP particles were relatively non-spherical. The impact of asphericity on the FAP and non-FAP is not well known and requires further studies which are beyond the scope of this pilot study.*

line 388 - these inferences should be connected to the results discussed earlier, and the same or meteorolgy data also

***Response****: We did the required modifications suggested by reviewer in the revised manuscript (page No.20; Line No.. 448).*

*The increment in total particle number concentrations on DOY 260-262 and DOY 266 shown in Fig 4a are possibly attributed to air mass arrived over the Islands in the Southeast of Puerto Rico. It could be the reason why we observed higher values of asphericity of non-FAP on DOY 261 and 262.*

Figure 6 - how can FBAP Dmvd is high and low at the same RH? Explain. Similarly, FAP increases as RH increases in general, but this function is not clear as there is a spread at aroun 80% RH, explain

***Response*** *(page No.21; Line No.. 478): We apologize for the typographical error in Fig. 6b. The FBAP should be FAP which will be corrected in the revised version. The increase of FAP Dmvd and the number concentrations depend on the hygroscopicity of the particles. Among the different FAPs measured at the site, the ABC, AB, and AC types were observed to have systematic diel patterns and were believed to be more hygroscopic than others. Therefore, the Dmvd and FAP number concentrations were increasing when RH reaches 80% and above. The other spread of points at*

*>80% RH were possibly the less hygroscopic FAP types such as the A, B, C, and BC, not showing any increasing trend in Dmvd and the number concentrations.*