Response to referee #1:

The manuscript is well organized and should be accepted after a minor revision, especially related to the method section.

We greatly appreciate the referee's valuable comments and helpful suggestions. We have addressed all comments as below and revised the manuscript accordingly.

Here below the specific comments:

- **Line 15, page 1: "boundary layer (BL)", better "planetary boundary layer (PBL)"**

We have changed "boundary layer (BL)" to "planetary boundary layer (PBL)" through the entire manuscript as the referee suggested.

- **Line 17, page 1: "aerodynamic diameter (PM2.5) within the BL was around 3 μg m^{-3}"; please add the confidence interval at 95%.**

To give a better description, we have revised the manuscript as below.

Page 1, Line 16: During the campaign, mass concentrations of particulate matters smaller than 2.5 μm in aerodynamic diameter ($PM_{2.5}$) within the PBL ranged from 0.5 to 12.0 μg m^{-3}, with an average and standard deviation of 3.4±2.3 μg m^{-3} …

- **Line 22 page 1: "One mode was exclusively coarse particles up to roughly 15 μm and peaked around 5 μm", please detail the cound mean diameter and the geometric mean standard deviation of each mode.**

The description of this coarse mode as well as the other two modes in the abstract intended to summarize the characteristics of average PMSDs for the PBL, the RL and the FT, followed by estimating contributions of possible sources to each mode from PMF analysis, not the exact fitted modes in the PMSD for each layer.

- **Line 80, page 4: It should be nice to add (in supplementary) a picture of these religious burning events.**

Pictures of religious burnings in temples as mentioned in section 2.1 on page 4 were not available, since it is not allowed to take photos in temples.

- **Line 100-103: Using different calibration standards (ammonium sulfate and polystyrene latex sphere) would affect the detected size distribution. Indeed the two standards are characterized by different refractive indexes thus generating different responses of the optical particle counter to the ambient aerosol. Please add a discussion on this methodology point considering the expected size distribution distortion.**

We agree with the referee that the two calibration standards differ in the refractive index, which could lead to different optical responses. We intended to report ammonium sulfate-based PNSDs, as the refractive index of ambient dry particles is closer to that of ammonium sulfate (Shingler et al., 2016) rather than polystyrene latex sphere (PSL), though PSL is widely used in the calibration of OPCs. However, without available methods to classify super-micron ammonium sulfate particles, a calibration curve combined from experimental responses of ammonium sulfate particles at submicron sizes and PSL at super-micron sizes was adopted in the original manuscript. Referee #2 also raised concerns using two calibration materials. We noticed in Gao et al. (2016) that theoretical response curves for PSL and dioctyl sebacate (DOS) were highly oscillatory above the particle size of 600 nm, leading to unrealistic particle sizing. Similar oscillations were also found for ammonium sulfate particles based on our calculation. Therefore, the response curve for converting the optical signal to the particle size above 600 nm should be smoothed to be monotonically increasing. In the revised manuscript, we re-calibrated POPS data using a combined calibration curve from the experimental response of ammonium sulfate particles for diameters smaller than 600 nm and the smoothed theoretical response of ammonium sulfate particles for diameters larger than 600 nm. By using the new calibration curve, PNSDs were obtained assuming only the refractive index of ammonium sulfate. Qualitative conclusions based on results using the new dataset remained the same, though quantitatively there were small changes in numbers and plots. We have revised the manuscript accordingly. We have also revised the description regarding POPS calibration as below.

Page 6, Line 101: The POPS was calibrated by establishing a relationship between the scattering signal and the particle size before the campaign. Both polystyrene latex sphere (PSL) with known sizes and ammonium sulfate particles with sizes selected by a differential mobility analyzer were employed. Though the experimental responses of the two calibration materials generally agreed well with the simulated theoretical responses, both theoretical response curves were found highly oscillatory above the particle size of 600 nm (Gao et al., 2016). Considering

that the refractive index of ammonium sulfate is closer to ambient dry aerosols than PSL (Shingler et al., 2016), a combined calibration curve from the experimental response of ammonium sulfate particles for diameters smaller than 600 nm and the smoothed theoretical response of ammonium sulfate particles for diameters larger than 600 nm was used to obtain 42 logarithmically equal size bins over the size range of 0.124~2.55 μm.

References:

Gao, R. S., Telg, H., McLaughlin, R. J., Ciciora, S. J., Watts, L. A., Richardson, M. S., Schwarz, J. P., Perring, A. E., Thornberry, T. D., Rollins, A. W., Markovic, M. Z., Bates, T. S., Johnson, J. E., and Fahey, D. W.: A light-weight, high-sensitivity particle spectrometer for PM2.5 aerosol measurements, Aerosol Sci. Technol., 50, 88-99, doi: 10.1080/02786826.2015.1131809, 2016.

Shingler, T., Crosbie, E., Ortega, A., Shiraiwa, M., Zuend, A., Beyersdorf, A., Ziemba, L., Anderson, B., Thornhill, L., Perring, A. E., Schwarz, J. P., Campazano-Jost, P., Day, D. A., Jimenez, J. L., Hair, J. W., Mikoviny, T., Wisthaler, A., and Sorooshian, A.: Airborne characterization of subsaturated aerosol hygroscopicity and dry refractive index from the surface to 6.5 km during the SEAC4RS campaign, J. Geophys. Res. Atmos., 121, 4188-4210, doi:10.1002/2015JD024498, 2016.

- **Line 104: Please add the relative humidity ensured by the diffusion dryer.**

The sample air was dried by the diffusion dryer filled with silica gel. Previous tests showed that roughly 75% and 50% of the water vapor could be removed by the diffusion dryer in summer and winter, respectively. As a matter of fact, the relative humidity will be further lowered inside the instrument, due to the temperature difference between the ambient condition and instrument chambers. The combined effects could ensure the relative humidity inside the POPS to be well below 20% and 30% in summer and winter, respectively. We have added a description of the diffusion dryer in the manuscript as below.

Page 6, Line 113: Previous tests of the diffusion dryer showed that roughly 75% and 50% of the water vapor could be removed after the drying in summer and winter, respectively. As a matter of fact, the relative humidity will be further lowered inside the instrument, due to the temperature difference between the ambient condition and instrument chambers. The combined effects resulted in the relative humidity inside the POPS to be well below 20% and 30% in summer and winter, respectively.

- **Line 106-107: A direct conversion of PNSDs into PMSDs can bring to some errors as the density of the fine and coarse mode can be different. Please compute and add to the paper also the Volume Size Distribution analysis (PVSD) for scientific consistency. Consider that the POPS suffer of a truncation error and an ambient aerosol density application will bring to a mass concentration underestimation.**

We agree with the referee that the density of fine particles might differ from that of coarse particles. We also agree with the referee that converting PNSDs into PMSDs based on the assumption of a constant density over a range of particle sizes might bring some uncertainties in the calculated PMSDs. However, by assuming a fixed density of 1.7 g cm^{-3}, the analysis of PMSDs in this study could actually be taken to some extent as the analysis of particle volume size distributions (PVSDs), as the two distributions shared similar shapes and variations with a difference only determined by a fixed factor.

The upper limit size of the POPS in this study was 2.55 μm, equivalent to the aerodynamic diameter of 3.32 μm assuming a density of 1.7 g cm^{-3}. Therefore, the underestimation of $PM_{2.5}$ mass concentrations due to size truncation of the instrument was negligible. We agree with the referee that the actual density of ambient aerosols, which was not available in this study, could be different from the assumed value. However, results regarding PMSDs/PVSDs should not qualitatively be much affected by the assumption of a fixed density. We have thereby added a discussion on possible uncertainties in the manuscript as below.

Page 7, Line 121: Though the assumption of a fixed density for particles with different diameters as well as the difference between the actual density of ambient particles and the assumed one might introduce some uncertainties in the calculated PMSDs, results regarding PMSDs would qualitatively not be much affected.

- **Line 110-112, page 6: "Mass concentrations of particulate matters smaller than 1 μm and 2.5 μm in aerodynamic diameter (PM1 and PM2.5) were obtained under the assumption that the optically equivalent diameter could be considered equal to geometric diameter". Here there is a big mistake. Please consider that the PM1 and PM2.5 are defined in function of an aerodynamic diameter and of an efficiency collection curve in inertial impactors (or similar cut-off devices such as cyclones). The authors should add at least a comparison between their PM1 and PM2.5 estimation and the same data obtained using gravimetric samplers.**

We appreciate the referee's valuable comment. We considered the optically equivalent diameter equal to the geometric diameter and converted the geometric diameter to the aerodynamic diameter assuming the density to be

1.7 g cm^{-3}. However, no aerosol penetration curves were considered for the calculation of PM_1 and $PM_{2.5}$ mass concentrations in the original manuscript. We agree with the referee that the calculation of PM_1 and $PM_{2.5}$ mass concentrations should take into account aerosol penetration curves of impactors as they are defined. We recalculate PM_1 and $PM_{2.5}$ mass concentrations, adopting the penetration curve of SCC-2.229 cyclone at 16.7 lpm for calculating PM_1 mass concentrations (https://bgi.mesalabs.com/wp-content/uploads/sites/35/2015/02/scc_btr-2.229.pdf, accessed on 15/03/2022) and VSCC-2.946 cyclone at 16.7 lpm for calculating $PM_{2.5}$ mass concentrations (https://bgi.mesalabs.com/wp-content/uploads/sites/35/2015/02/vsccref6-2.946.pdf, accessed on 15/03/2022). According to comments of both referees, a new calibration curve was used to obtain particle sizes and corresponding number concentrations. The final results regarding PM_1 and $PM_{2.5}$ mass concentrations in the revised manuscript were based upon the newly derived PNSDs and the new calculation method involving penetration curves. Though we did not have simultaneous measurements from gravimetric samplers for comparisons, we compared PM_1 and $PM_{2.5}$ mass concentrations calculated from the previous method without penetration curves and the new method with penetration curves. Averagely, mass concentrations using the new method changed 1.1 ±0.8% for PM_1 and -0.2 ±1.6% for $PM_{2.5}$ relative to that using the previous method. Conclusions drawn from comparisons between PM_1 and $PM_{2.5}$ mass concentrations in this study and in other studies were not affected, when using the new method instead of the previous one. We have added more details on obtaining the aerodynamic diameters and a description of aerosol penetration curves in the manuscript as below.

Page 7, Line 125: …and the aerodynamic diameter could be converted from the geometric diameter with a density of 1.7 g cm^{-3}. The penetration curve of SCC-2.229 cyclone at 16.7 lpm (https://bgi.mesalabs.com/wp-content/uploads/sites/35/2015/02/scc_btr-2.229.pdf, accessed on 15/03/2022) was adopted for calculating PM_1 mass concentrations and the penetration curve of VSCC-2.946 cyclone at 16.7 lpm (https://bgi.mesalabs.com/wp-content/uploads/sites/35/2015/02/vsccref6-2.946.pdf, accessed on 15/03/2022) was adopted for calculating $PM_{2.5}$ mass concentrations.

■ **Line 113-114: Was the GRIMM 11-C equipped with a dryer. If yes was it deployed on the balloon?**

The GRIMM 11-C was also equipped with the homemade silica gel-filled diffusion drier as was POPS and attached to the tethered balloon for 24 launches. To better clarify the condition, we have revised the manuscript as below.

Page 7, Line 134: A portable aerosol spectrometer (Model 11-C, GRIMM Aerosol Technik GmbH & Co. KG), equipped with the homemade silica gel-filled diffusion drier, was also attached to the tethered balloon for 24 launches to concurrently measure the PNSD for dry particles within the size range of 0.25~32 μm.

- **Line 133-134: Are there any radiosoundings in Lahsa that should be used to unravel the situations in which the top of ML was above the top of the profiles?**

There were no radio soundings when flights of the tethered balloon were totally within the ML to unravel such situations.

- **Line 149: "strong species"; maybe better "strong aerosol types"? Species remind to chemical speciation which is not present in this work.**

We have revised the manuscript accordingly.

- **Line 163: better "ranged from" … "to"…**

We have revised the manuscript accordingly.