We thank both reviewers for their thorough reviews. We appreciate their constructive comments and suggestions and the manuscript has been revised accordingly. Our point-by-point responses to the comments are presented below. The comments are in **black**, responses are in **blue**, and revised manuscript are in **red** with specific changes marked by <u>underline</u>.

In this work, the setup and validation of a laser imaging nephelometer that can be used for aircraft deployment and measurement of polarization state of particles was introduced. In addition, this instrument was also applied in the FIREX-AQ campaign for measurement of smoke plume and was found to be able to measure very high scattering coefficients with high temporal resolution. This work expanded the application of LiNeph, which can play an important role in comprehensive measurement of aerosol optical properties. The manuscript fits well to the scope of AMT and I recommend it to be published after addressing the following comments listed below.

Thank you for positive review. Our responses to your comments are below.

Specific comment:

Although it is impossible to use polarized particles to calibrate PiNeph, it is helpful to check the measurement of -P11/P12 for non-polarized particle. Quantification of the uncertainty of P11/P12 measurement is very important for the measured polarization state of particles in field campaign.

We have modified to Figure 4 to show the P_{11} and $-P_{12}/P_{11}$ for PSL data. However, we have maintained the convention of presenting the differential scattering coefficients for each detector in the Supplementary Fig. S2-S5. We feel that this is a more straightforward way to evaluate the potential influence of a change in the scattering plane rotation, η , given each detector will have a different $\eta(\theta)$.

Line 228:

Figure 4 shows the good agreement between the measured and calculated P_{11} and DoLP. Mie theory is used to calculate the expected phase function and degree of linear polarization for two theoretical instrument geometries. Although the center axis of each camera is parallel or perpendicular to the polarization of the laser, the beams themselves are offset from the center axis by ~3.6 mm. This is roughly defined by the geometry of the series of four concentric apertures through which the lasers are introduced into the sample cell. Due to the nature of the wide angle lens, this can result in a scattering plane rotation angle, η , shown in Fig.1 and described in detail by Dolgos et al. (2014.) Figure 4 shows predictions of what the expected P₁₁ and DoLP would be if η is assumed to be zero (gold line) or if an estimated η were used (teal line). We do not observe improved agreement between the observed P₁₁ nor DoLP by using the estimated η , thus we treat our estimated η as an upper limit (see Supplementary Fig S2-S5.) For some scattering angles (e.g. near 30° and 135°), there is stray light from the instrument background which introduces additional noise as extraneous features.

Technical comments:

L138: It would be better to describe the cell volume and the exchange rate here.

We have added the sample cell volume to the text, but leave the exchange rate for the more detailed discussion later in the text.

Sample flow is pulled through the instrument sample cell by an external diaphragm pump and controlled by a mass flow controller (MCR-50, Alicat, Tuscon AZ, USA). For the FIREX-AQ mission aboard the NASA DC-8, a sample flowrate of 15 l min⁻¹ was used to maximize the sample exchange rate <u>in the ~3 l sample</u> <u>volume</u>, and thus improve the ability of the instrument to resolve spatial changes in aerosol concentration as the aircraft penetrated a smoke plume.

L142: Since you have measured the RH and temperature of instrument exhaust, how did they change during the measurement in the FIREX-AQ campaign?

We have included the following on line 211:

The aerosol sampled by the LiNeph was also dried to less than 20% RH using Nafion driers and passed through a cyclone with a calculated cut size of 1.5 μ m aerodynamic diameter. The aircraft cabin was temperature controlled, resulting in average sampling temperatures of ~26 °C, although temperatures could rise as high as 36 °C when sampling at low altitudes for an extended duration.

L181: The calibration requires not only compositions but also the size of aerosol, as you mentioned later.

We have modified the text as follows:

The calibration of the LiNeph requires the sampling of gases and aerosols of known <u>size and</u> composition.

L195: The cut-off size impactors before each instruments were different from each other. Why don't you use impactors with the same cut-off size?

Although using the same cut-off size would have been better, we felt confident that for the inplume sampling the aerosol scattering would be dominated by submicron particles. This was confirmed by measurements by the LAS which did not have an impactor. We have included the following in the text:

However, in smoke plumes the difference between the total light scattering of PM1.5 vs PM2.5 is negligible due to the overwhelming abundance of submicron particles. This was confirmed using size distributions from the LAS, which showed few particles with diameters greater than $1 \mu m$ (Moore et al., 2021).

L305: How large in specific do you mean by "very large ash particles"? Because there is a PM1.5 impactor before PiNeph, it seems that "very large ash particles" should be smaller than 1.5um.

We have removed the phrase "very large ash particles" from the text.

One instance where this would not be the case is if rare but highly scattering particles, for example very large ash particles, transit through the sample cell but only transect the laser at a few angles.

L311: Which aerosol concentration do you refer to? Volume, number or mass?

It is more accurate to say this phenomenon would occur due to a rapidly changing β_{scat} , and that the e.g. rapidly increasing β_{scat} would occur due to an increasing aerosol number (or mass or volume) concentration. We have modified the text as follows:

Another potential source of error during ambient measurements is a <u>rapidly</u> changing β_{scat} , e.g. when <u>there is an increase in the aerosol number</u> concentration. If the sampled β_{scat} increases rapidly, the β_{scat} gradient within the sample cell will be observed in the scattering phase function. For example, if the β_{scat} at the instrument inlet increases during a measurement, there may be more scattering in the section of the sample cell corresponding to forward scattering angles than in the portion of the cell corresponding to backscattering.

L313: "in" should be "if".

This has been fixed, thank you.

L341: How did other imaging nephelometer perform in the comparison with AOP-derived scattering?

We have included the following:

If we take the AOP-derived scattering as a truth measurement, i.e. without its own error, we can say that the LiNeph is precise within <2%, although with a positive bias of ~30%, likely due to calibration error. This is consistent with the reported precision of similar techniques, i.e. the PI-Neph reports agreement with commercial integrating nephelometers to within 5% (Espinosa et al., 2017).

L363: It's not "cumulative".

We have corrected the text as follows:

Supplementary Fig. S11 shows the <u>average normalized</u> number-weighted size distributions for both transects as measured by the LAS with an applied ammonium sulfate calibration (Moore et al., 2021).

L372: It would be better to present the measured values of the asymmetry parameter in the main text besides in the figure.

We have modified the text as follows:

The asymmetry parameter, described below, increases from 0.568 to 0.620, showing an increase in forward scattering that is consistent with increasing particle size. However, it is important to note that changes in particle composition, hence refractive index, have also been observed as a consequence of photochemical aging in biomass burning aerosol.

L416: It's unclear that "accurately determine the asymmetry parameter within 3%".

We have modified the text as follows:

Line 455:

The phase function measurements (e.g. Fig. 7) allow for <u>precise</u> measurements of the asymmetry parameter with a relative standard deviation of less than 3%. <u>But, as discussed in Section 2.4, the</u> geometry of the LiNeph requires that the lasers be offset from the optical axis of the wide angle lenses, introducing a non-zero η . Based on measurements of PSLs (Fig. S2-S5), we can set an upper bound of this effect on the measured σ° . We used the same method to investigate the effect of η on the measured asymmetry parameter. Supplementary Figure S12 shows that for polydisperse lognormal

aerosol size distributions centered around 200 nm, with varying refractive indices, the effect of a nonzero n on g is a bias of less than 2%. For the largest modeled aerosol population, with a mode centered at 400 nm, the bias was 5%. This effect is small in part because the geometry of the instrument results in offsetting biases when $\sigma^{\circ}_{"Perp"}$ and $\sigma^{\circ}_{"Para"}$ are combined to calculate P₁₁. For the four P₁₁ measurements of PSL calculated from the data in Supplemental Fig. S2-S5, the average ratio of measured to Miecalculated g was 1.01 ± 0.09.

Line 482:

Finally, we showed that we can <u>precisely (less than 3% relative standard deviation) and</u> accurately (within 10% for the PSLs examined in this work) determine the asymmetry parameter.