Response to Referee #2

We sincerely thank the editor and all reviewers for your time and the constructive comments on our manuscript. We have carefully considered all the comments and suggestions. Below are our point-by-point detailing how we have addressed each issue in the revised manuscript. In the following, paragraphs in **black** are reviewers' comments; paragraphs in **blue** are point-to-point responses; paragraphs in **red** are revisions in the manuscript.

General Comments:

The manuscript investigates whether a large-aperture scintillometer (LAS) can be used to estimate aerosol mass fluxes above water surfaces. Measurements above an urban lake and a coastal bay are compared with an extinction-based eddy-covariance (EC) approach and filter-based dry-deposition estimates. The subject is relevant to Atmospheric Measurement Techniques, and spatially averaged aerosol-transport measurements using scintillometry could be valuable.

However, the manuscript does not yet provide sufficient methodological information or evidence to demonstrate that the reported quantities are quantitatively reliable aerosol mass fluxes. The main concerns involve the response of the visibility sensor, the absence of spectral flux diagnostics, the sign and physical meaning of the LAS-derived quantity, the applicability of the free-convection formulation, hygroscopic effects on the extinction-to-mass conversion, filter treatment, and the interpretation of modelled dry deposition as an independent validation method.

I therefore cannot recommend publication in its present form. Major revision and substantial reanalysis are required. In particular, the LAS–EC validation can be retained only if the authors demonstrate that the visibility sensor provided independent extinction measurements with sufficient temporal response to resolve turbulent covariance.

Before responding point by point, we wish to explain a fundamental restructuring of the manuscript prompted by Reviewer 2's concern about the PWD50. Although the PWD50 output was logged at 1 Hz, the standard instrument has an independent measurement interval of 15 s; therefore, the 1 s records cannot be treated as independent fast-response extinction observations. Our reanalysis of the four campaigns, including vertical-wind–extinction co-spectra and cumulative ogives, did not show a stable and reproducible turbulent covariance that could support a conventional aerosol eddy-covariance flux. We consequently removed the entire LAS–EC comparison and the associated argument that agreement with EC demonstrated the feasibility of LAS. The PWD50 is now used only as a low-time-resolution visibility/extinction reference for the hourly R_{MN} calibration and for reproducible fog screening. The revised manuscript instead extends the LAS framework from terrestrial to open-water-dominated environments from the theoretical and measurement perspectives: it evaluates the separation of the real and imaginary refractive-index contributions in the LAS spectrum, introduces campaign-specific continuous R_{MN} (RH) relationships, and retrieves aerosol mass-flux magnitudes. Accordingly, the revised claims are restricted to path-integrated aerosol mass-exchange magnitudes and their temporal variability under the stated operating conditions; independent EC validation is no longer claimed.

Specific Comments:

Comment 1: Flux cospectra and cumulative ogives should be calculated for all accepted averaging intervals and all four campaigns. Velocity spectra alone cannot demonstrate that the covariance between vertical wind and aerosol extinction represents a resolved turbulent flux. Cospectra are required to identify the frequencies contributing to the flux and diagnose high-frequency attenuation, noise, time-lag errors, aliasing and phase shifts. Ogives should show whether the covariance converges within the 30 min averaging period or remains affected by low-frequency non-stationarity and mesoscale variability. Campaign-level median cospectra and ogives with interquartile ranges could be presented in the manuscript, with individual diagnostics supplied in the supplement or data repository. Equivalent LAS log-intensity spectra are also needed to support the separation of the high- and low-frequency components used to derive the real and imaginary refractive-index structure parameters.

The manuscript must distinguish between logging frequency, output interval, temporal resolution and response time (check, e.g., Buzourius et al., 2003). Although PWD50 data were reportedly recorded at 1 Hz, the standard instrument specification gives a measurement interval of 15 s. Polling the output every second may therefore only reproduce internally averaged values rather than provide independent 1 Hz extinction measurements. This corresponds to a maximum independent Nyquist frequency of approximately 0.033 Hz, even before additional response attenuation is considered. A longer averaging period cannot recover covariance already removed by the instrument response.

The authors should report the sensor's independent update interval, internal averaging, physical response function, synchronization with the sonic anemometer, time-lag procedure and frequency-response correction. They should also clarify whether any hardware or firmware modification was implemented and experimentally characterized. This is particularly important because Yuan et al. (2024) used a modified CS120A sensor and still reported response-time and high-frequency-noise limitations. If a standard PWD50 output was used, the resulting estimate should be described as a low-frequency covariance proxy rather than a conventional 1 Hz aerosol EC flux.

Response:

We sincerely thank the reviewer for identifying this critical issue. The comment prompted us to re-examine the temporal characteristics of the PWD50 and the validity of the original PWD50–sonic-anemometer covariance calculation. We agree with the reviewer's central concern. The PWD50 used in this study was a standard, unmodified instrument. Its output was polled and logged at 1 Hz, but the manufacturer-specified independent measurement interval is 15 s. No hardware or firmware modification was made. The earlier treatment of the repeated 1 s outputs as independent fast-response extinction measurements was therefore not defensible. A time-lag adjustment, longer averaging period, or frequency-response correction cannot reconstruct turbulent covariance that was already attenuated by the instrument response.

As requested, we calculated vertical-wind–extinction co-spectra and cumulative ogives for all 30 min windows that passed the basic spectral-data screening in the four campaigns. Figure R1 contains spectral windows for Siyuan Lake I, Siyuan Lake II, Sijiao Island I, and Sijiao Island II, respectively. For a reliably resolved eddy-covariance flux, the co-spectra should exhibit a reproducible frequency range contributing predominantly in one direction, while the corresponding ogives should approach a stable plateau toward the low-frequency end, indicating that the covariance has converged within the 30 min averaging period.

Neither characteristic was consistently observed. In Fig. R1, the individual ogives diverge markedly below approximately 0.01 Hz, frequently continue to change toward the lowest resolved frequencies, and terminate with both positive and negative values rather than converging toward a common plateau. The campaign-median terminal covariances were -7.3×10^{-9}, 9.8×10^{-9}, -9.4×10^{-9}, and 3.2×10^{-8} s^{-1}, respectively, and therefore remained close to zero compared with the approximately 10^{-6} s^{-1} dispersion of the individual windows. Thus, the near-zero campaign medians result primarily from cancellation between highly dispersed positive and negative low-frequency contributions, rather than from consistently convergent covariance estimates.

Figure R2 leads to the same conclusion in the frequency domain. After excluding covariance outliers and windows whose near-zero covariance would make normalization unstable, the normalized co-spectra were separated according to the sign of their final covariance. Both groups show broad contributions mainly between approximately 0.001 and 0.01 Hz, but their between-window dispersion is large and their group medians form approximately opposite-sign mirror patterns. Because the groups were defined by the sign of the final covariance, this mirror structure indicates that low-frequency fluctuations determine whether an individual 30 min covariance is positive or negative; it does not demonstrate a repeatable, same-direction turbulent-transfer band. The median co-spectral contribution becomes negligible at higher frequencies, consistent with the attenuation expected from the PWD50’s 15 s independent measurement interval.

Taken together, the co-spectra and ogives indicate that the calculated PWD50–vertical-wind covariance is dominated by sign-variable, low-frequency contributions and is not reproducibly convergent across averaging windows or campaigns. This finding does not imply that the actual aerosol exchange was zero.

Rather, it shows that the standard PWD50 configuration used here cannot resolve the magnitude or direction of turbulent aerosol transport with sufficient temporal fidelity to provide a conventional aerosol EC flux or an independent validation of the LAS retrieval.

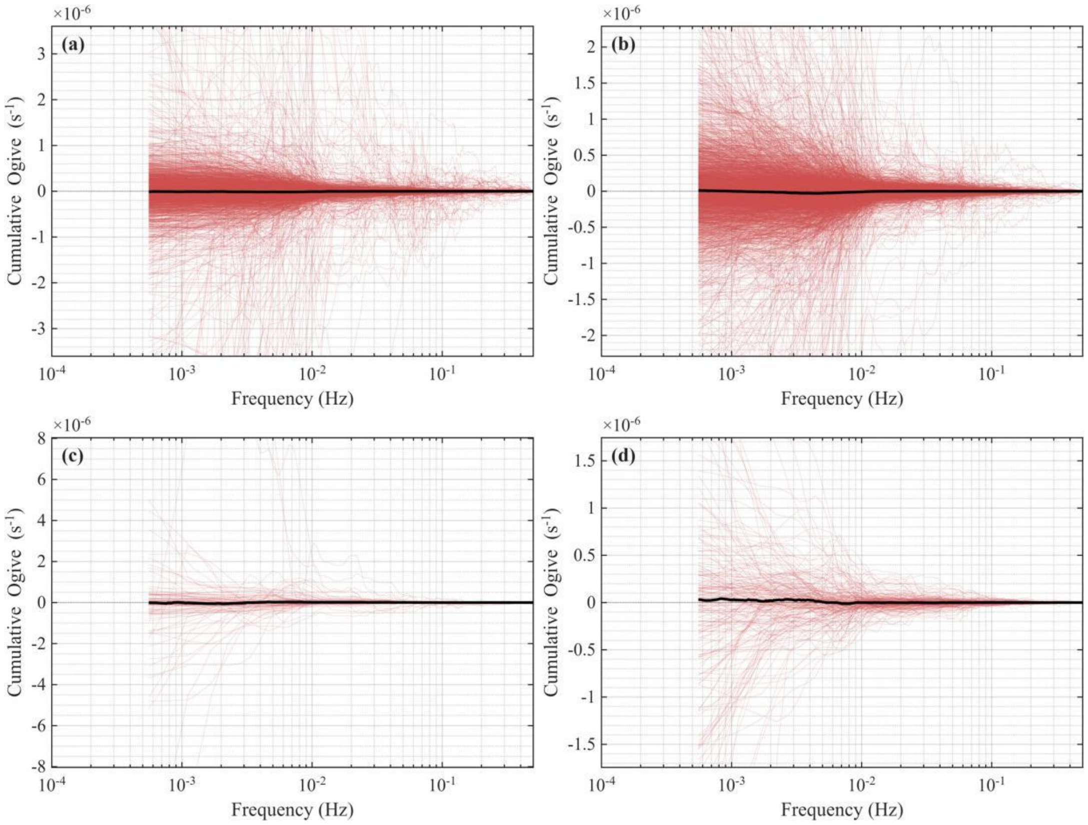

Figure R1. Cumulative vertical-wind–extinction ogives for all valid 30 min spectral windows during (a) Siyuan Lake I, (b) Siyuan Lake II, (c) Sijiao Island I, and (d) Sijiao Island II. Pale red curves denote individual windows and black curves denote campaign medians. The medians remain close to zero while the individual low-frequency contributions are widely dispersed in sign and magnitude.

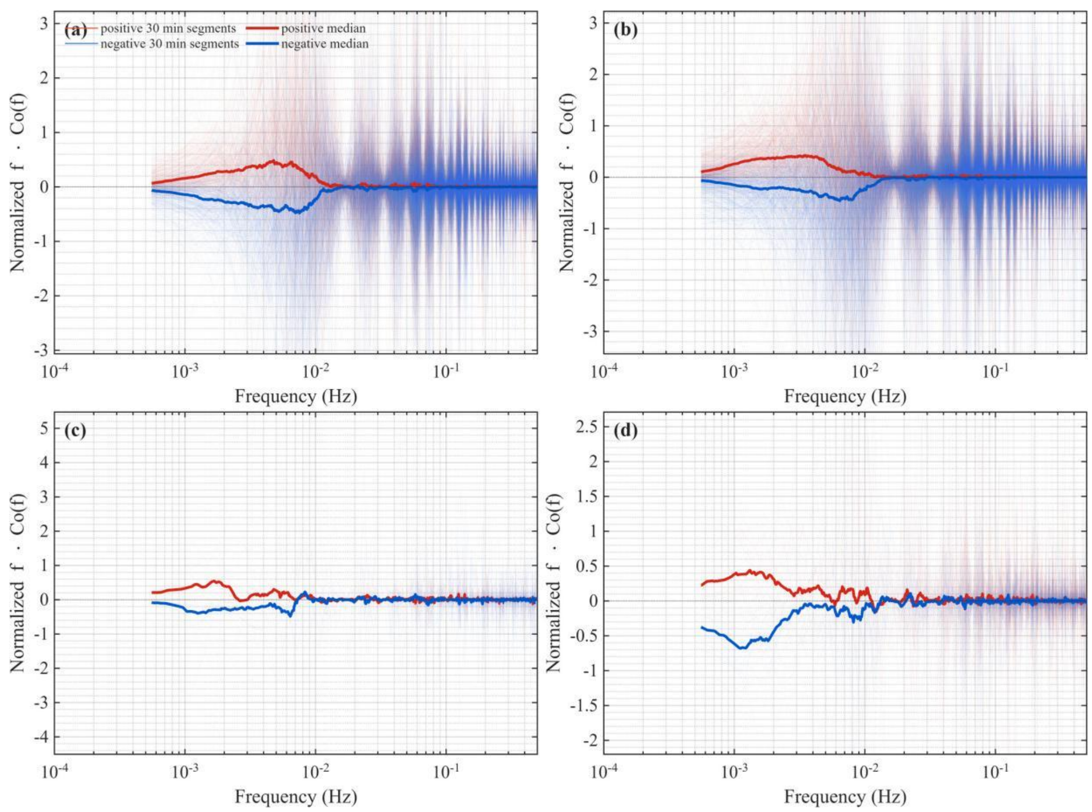

Figure R2. Normalized vertical-wind–extinction co-spectra for (a) Siyuan Lake I, (b) Siyuan Lake II, (c) Sijiao Island I, and (d) Sijiao Island II. Transparent red and blue curves show individual positive- and negative-covariance 30 min windows after exclusion of covariance outliers and near-zero normalizing covariances; thick red and blue curves are the corresponding group medians. Each curve is normalized by the absolute final covariance of its window.

This recognition led us to fundamentally restructure the manuscript. We removed all PWD50–sonic aerosol EC fluxes, LAS–EC correlations, temporally averaged comparisons, NRMSE values, ensemble EC/LAS cycles, and the associated claim that agreement with EC validated the feasibility of LAS over water. The PWD50 is now used only to provide visibility-derived extinction for the R_{MN} calibration and to identify fog-affected intervals.

We have also revised the manuscript to distinguish explicitly among the temporal concepts identified by the reviewer. For the PWD50, 1 Hz now refers only to the frequency at which the data logger polled and stored the instrument output, that is, a 1 s logging or recorded-output interval; it is not described as a 1 Hz independent measurement resolution. The independent measurement interval is 15 s, which corresponds to a maximum independent Nyquist frequency of approximately 0.033 Hz before any additional response attenuation is considered. To apply this distinction consistently across the measurement system, Sect. 2.1 and Table 1 now also report the relevant temporal characteristics of the other instruments. The LAS log-intensity signal was acquired at 500 Hz, and the CSAT3B wind components and sonic temperature were acquired at 20 Hz. The HMP155A and CS106 outputs were logged at 1 Hz; however, the specified RH response times of the HMP155A are 20 s for 63 % response and 60 s for 90 % response at 20 °C in still air, whereas the CS106 has a response time of 0.5 s following a pressure step. These acquisition rates, logging intervals, independent measurement intervals, and physical response times are now reported separately.

The equivalent LAS spectral evidence requested by the reviewer remains directly relevant to the revised retrieval and has therefore been added to the main manuscript. For each campaign, Fig. 2 presents the median measured 30 min log-intensity spectrum and its interquartile range together with the median theoretical imaginary and real refractive-index contributions. Section 3.2.1 now explains what can be concluded from these spectra: the imaginary contribution dominates at low frequencies, the real contribution dominates after the theoretical components intersect, and the crossover remains identifiable in all four campaigns. The campaign-level crossover is used only to illustrate the general spectral organization; the crossover applied in the retrieval is determined independently for every accepted 30 min interval.

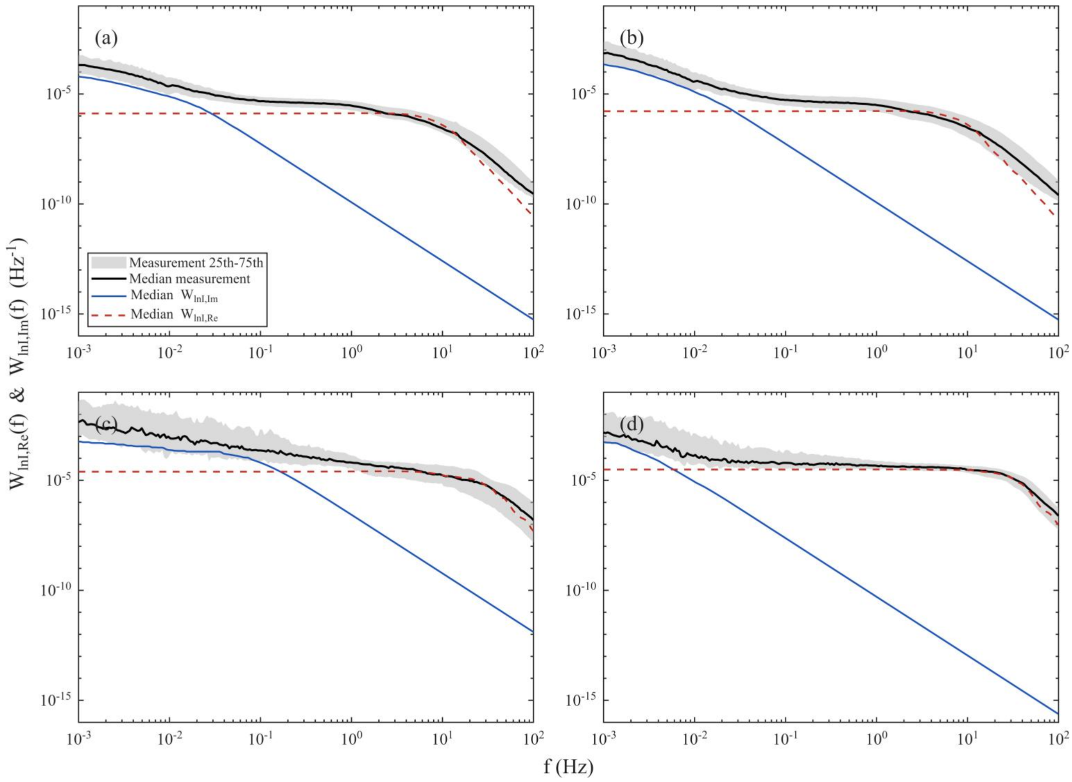

Figure R3 (reproduced from the revised manuscript Fig. 2). Median log-intensity spectra of the accepted 30-min LAS observations during (a) Siyuan Lake I, (b) Siyuan Lake II, (c) Sijiao Island I, and (d) Sijiao Island II. Black curves denote the measured median spectra and gray shading indicates the 25th–75th percentile range. Blue solid and red dashed curves represent the median theoretical contributions of the imaginary and real refractive-index components, respectively. The intersection of the two theoretical components defines the crossover between the low- and high-frequency regimes.

The Sect. 2.1, Page 4, lines 87-97:

"The LAS system (Aiutyl-LAS, Sciencesky)…These timescales have limited influence on the derived mean quantities."

The Sect. 2.1, Pages 5-6, line 125 (Table 1):

"Table 1. Specifications, temporal characteristics, and study-specific configuration of the observational instruments…mounting heights are reported relative to the local water surface."

The Sect. 3.2.1, Pages 13-14, lines 315-341:

"Figure 2 shows the campaign-level log-intensity spectra for the four observational periods…the intersection defines the crossover between the low- and high-frequency regimes."

Comment 2: It is unclear whether Eq. (5) represents turbulent transport, flux magnitude, net flux, total flux, surface emission or another effective flux. Because the equation contains only positive powers of structure parameters, it appears to return a magnitude without determining direction. Nevertheless, all results are positive and repeatedly interpreted as upward aerosol emission, while logarithmic axes prevent negative deposition fluxes from being displayed.

Continuous upward emission during all campaigns is physically unlikely, particularly under weak winds or stable conditions, when downward deposition should occur. The authors must state explicitly whether absolute flux magnitudes are reported and explain how flux direction is determined.

Equation (5) appears to be a free-convection approximation whose validity is mainly expected under strongly unstable conditions. Its application to daytime and nighttime periods, different seasons and a mechanically turbulent coastal site is not justified. The authors should either use a complete Monin–Obukhov similarity formulation or restrict the analysis to conditions satisfying the free-convection assumptions. A stability-stratified comparison based on (z/L), friction velocity and the air–water temperature difference is essential. The assumption that solar radiation automatically generates unstable daytime conditions above water is insufficient because the thermal inertia of the water surface may produce a different diurnal stability cycle.

Response: We thank the reviewer for this insightful comment. The LAS retrieval does not preserve the sign of aerosol transport because both the imaginary refractive-index structure parameter and the derived aerosol mass-concentration structure parameter are obtained from squared fluctuations. The revised manuscript therefore defines the reported quantity as the magnitude of turbulent aerosol mass exchange. It is not described as a net flux, total flux, upward emission, or deposition.

The former free-convection approximation has also been replaced by the complete stability-dependent Monin–Obukhov similarity formulation. For every 30 min interval, friction velocity and Obukhov length are obtained from the independently processed 20 Hz sonic-anemometer record, and the similarity function is evaluated at the concurrent stability parameter. The analysis therefore does not assume that daytime periods over water are automatically unstable and is not restricted to strongly unstable free-convection conditions.

The requested stability-stratified and weak-turbulence diagnostics are provided in Supplement Sects. S4–S5, including Table S2, Tables S3a–S3b, and Figs. S4–S6. Water temperature was not measured, so the air–water temperature difference cannot be reconstructed reliably; this unavailable variable is identified in Supplement Sect. S11 rather than inferred from solar radiation or air temperature.

The Sect. 2.4, Pages 8-9, lines 190-202:

"Following Yuan et al. (2019), the aerosol mass flux was derived within the Monin–Obukhov similarity framework…all LAS-derived aerosol fluxes are interpreted as flux magnitudes."

Comment 3: The term "open water" is insufficiently defined and may be misleading. An urban campus lake and a sheltered coastal bay do not necessarily represent open-ocean or high-fetch conditions. The authors should clarify whether the term refers only to an unobstructed water surface and quantify fetch, surface homogeneity and the fraction of each footprint located over water. If substantial land contributions occur, the results should be described as exchange over heterogeneous lake–land or coast–water systems.

All inputs and validity criteria for the Kljun et al. (2015) model, the wind-coordinate transformation and the treatment of boundary-layer height should be reported. In addition to campaign-mean footprint climatologies, the authors should calculate an instantaneous water-footprint fraction and LAS–EC footprint-overlap metric. Otherwise, footprint mismatch can only be proposed as a possible explanation for the low correlations, not presented as a demonstrated cause.

Response: We agree that the original use of 'open water' could be read too broadly. The revised manuscript defines an 'open-water site' operationally as a measurement configuration whose path-weighted LAS footprint is dominated by water; the term does not imply open-ocean conditions, unlimited fetch, or a spatially homogeneous surface. The lake campaigns are now described as water-dominated but heterogeneous lake–land measurements, whereas the coastal footprints represent water almost exclusively. The normalized water contributions are 79.4 % and 75.1 % for Siyuan Lake I and II and 99.4 % and 99.6 % for Sijiao Island I and II, respectively.

A single geometric fetch is not reported because the relevant upwind source area changes with wind direction and stability from one 30 min interval to another. Instead, surface heterogeneity is quantified with the two-dimensional, path-integrated footprint and the water-surface fraction. Section 2.1 reports the 290 m lake path and 1300 m coastal path, while Sect. 2.5 defines how the point footprints are integrated along each optical path using the normalized scintillometer weighting function. Time-resolved water-footprint fractions are provided in Supplement Sect. S6, Table S4, and Figs. S7–S8 in addition to the campaign climatologies.

Section 2.5 reports the 30 min footprint resolution and all FFP inputs: effective measurement height, mean wind speed, friction velocity, Obukhov length, lateral-wind standard deviation, boundary-layer height, and wind direction. Because no time-resolved boundary-layer-height measurements were available, h was fixed at 1000 m; this is identified as an uncertainty, and the sensitivity to h is reported in Supplement Sect. S6, Table S4, and Figs. S7–S8.

The requested LAS–EC footprint-overlap metric is no longer applicable because the PWD50-based EC flux and all LAS–EC correlations have been removed. Consequently, the revised manuscript neither attributes low correlations to footprint mismatch nor presents footprint overlap as validation evidence. The footprint analysis is retained only to define the surface types contributing to the path-integrated LAS retrieval.

The Sect. 2.1, Page 3, lines 72-78:

"A total of four field campaigns were conducted at two open-water sites in eastern China…Hereafter, the two campaigns at each site are denoted I and II in chronological order (Siyuan Lake I and II; Sijiao Island I and II)."

The Sect. 2.5, Page 9, lines 215-219:

"The point footprints were calculated at a 30 min resolution…the fixed h remains an uncertainty in the present climatologies."

The Sect. 3.4, Page 22, lines 470-476:

"The footprint climatologies at Siyuan Lake were elongated along the LAS transect…while also incorporating a smaller contribution from the surrounding land."

The Sect. 3.4, Page 24, lines 502-508:

"The footprint climatologies at Sijiao Island were elongated along the 1.3 km LAS transect…with only trace contributions from the adjacent land."

Comment 4: Section 2.2 does not provide enough information to reproduce the EC calculation. The authors should specify the coordinate-rotation and detrending methods, time-lag determination, resampling and synchronization, spike detection, stationarity and integral-turbulence tests, low u* filtering, data-completeness criteria, flow-distortion treatment and high- and low-frequency spectral corrections. They should also discuss air-density and water-vapour effects, whether a Webb–Pearman–Leuning-type correction is applicable to the converted mass concentration, and how hygroscopic effects influence the covariance. Random uncertainty, instrumental detection limits and uncertainty in converting extinction into mass should be quantified. Zinke et al. (2024) may provide a useful methodological reference.

The ±3 standard-deviation filter relative to a 2 h moving average is insufficiently described. If applied to raw high-frequency variables, it could remove genuine turbulent excursions and attenuate covariance. Despiking of raw signals should be clearly distinguished from quality screening of averaged fluxes.

Response: We sincerely thank the reviewer for this suggestion. Since the aerosol EC calculation has been deleted, PWD50–sonic detrending, resampling, synchronization, time-lag selection, high- and low-frequency covariance corrections, and an aerosol-EC detection limit are no longer part of the revised method. Likewise, WPL-type density corrections and humidity corrections to a PWD50-derived turbulent covariance are no longer applicable.

The sonic anemometer remains necessary only for the LAS similarity calculation and footprint inputs. Its independent processing is now specified in Sect. 2.7: at least 90 % coverage per 30 min interval, explicit exclusion of the 160–200° local mast-shadow sector, normalized-MAD spike detection for the three wind components and sonic temperature, and double rotation. The former ±3-standard-deviation filter relative to a 2 h moving average has been removed. Raw LAS and sonic-anemometer spikes are now treated with an explicitly defined robust normalized-MAD rule before 30 min calculation, whereas rejection of a completed 30 min retrieval is governed by separate coverage, optical, meteorological, spectral, and sonic criteria. This distinguishes sample-level despiking from interval-level quality control and avoids using the former moving-average rule to remove genuine turbulent excursions.

The stationarity and integral-turbulence tests and weak-turbulence distributions are provided in Supplement Sects. S4–S5, Tables S2 and S3a–S3b, and Figs. S4–S6. The conditional LAS uncertainty budget is provided in Supplement Sect. S8, Tables S6a–S6b, and Fig. S10. An EC-specific uncertainty or detection limit is not reported because the aerosol EC product itself has been withdrawn.

The Sect. 2.7, Pages 10-11, lines 241-270:

"All measurements were processed at a common 30-min retrieval interval…meteorological screening used the same reproducible fog criterion defined above."

Comment 5: The conversion factor R_{MN} is central to both LAS and EC estimates but is not adequately determined. Filter measurements represent conditioned aerosol mass, whereas LAS and the visibility sensor respond to particles at ambient relative humidity, which reached 100%. Hygroscopic growth can strongly increase particle size and extinction near saturation, while fog droplets may contribute to extinction without representing the PM_{10} mass collected on filters.

The authors should therefore evaluate the humidity dependence of extinction, scattering enhancement, hygroscopic growth and possible humidity–aerosol covariance rather than assume a constant R_{MN}. The possible contribution of water-vapour fluctuations to the real refractive-index structure parameter should also be discussed. Finally, the imaginary component should not be described as responding only to light-absorbing aerosol: the measured attenuation includes scattering, which is generally important or dominant for marine aerosol.

Response: We agree that a constant R_{MN} cannot represent the conversion between conditioned aerosol mass and ambient optical extinction over the observed humidity range. The revised framework therefore determines a separate continuous

R_{MN}(RH) relationship for each campaign from conditioned PM_{10}, visibility-derived imaginary refractive index, and concurrent RH. The campaign-specific treatment is necessary because the mass-to-optical relationship also depends on aerosol size distribution, composition, and optical properties. This empirical relationship represents the observed combined humidity dependence of conditioned mass and ambient extinction; it is not interpreted as a direct thermodynamic hygroscopicity measurement. No independent measurements of particle growth factor, scattering enhancement, aerosol liquid-water content, or size-resolved composition were available, so those processes cannot be separated individually. A turbulent humidity–aerosol covariance is also not calculated because the PWD50 does not provide independent fast-response extinction measurements.

Fog-affected observations with visibility below 1 km and measurements at the upper instrumental visibility range are excluded before calibration. Ambient humidity has already affected the measured extinction and the inferred imaginary refractive index; therefore, applying a second RH correction directly to that optical quantity would double count the same influence. Humidity instead enters the conversion through R_{MN}(RH).

The basis of Eq. (6) $R_{MN} = R_{\mathrm{ref}}\left[\frac{1-RH/100}{1-RH_{\mathrm{ref}}/100}\right]^{\gamma}$ can be stated through the optical quantity measured in this study. Zhou et al. (2020) use the established gamma parameterization for the RH-dependent enhancement of aerosol extinction or scattering. Written relative to a reference humidity, the optical enhancement factor is:

$$f_{\mathrm{ext}}(RH) = \left[\frac{1-RH/100}{1-RH_{\mathrm{ref}}/100}\right]^{-\gamma}, f_{\mathrm{ext}}(RH) = \frac{\beta_{\mathrm{ext}}(RH)}{\beta_{\mathrm{ext}}(\mathrm{Dry})}$$

$$n_{\mathrm{Im}} \propto \beta_{\mathrm{ext}}, \quad R_{\mathrm{MN}} = \frac{M_{\mathrm{a}}}{n_{\mathrm{Im}}} \Rightarrow R_{\mathrm{MN}}(RH) = R_{\mathrm{ref}}\left[\frac{1-RH/100}{1-RH_{\mathrm{ref}}/100}\right]^{\gamma}$$

The calibration used seven RH classes and required a significant PM_{10}–n_{Im} regression at $\alpha = 0.05$. In the Siyuan Lake II 60–70 % example, 280 of 292 hourly pairs were retained ($r = 0.87$), giving R_{MN} = 3835 kg m^{-3}. The estimates decreased with RH in all campaigns, and the continuous fits gave γ values of 0.750, 0.912, 1.518, and 0.838. For Sijiao Island II, the all-data sensitivity fit changed γ from 0.838 to 0.679 and changed R_{MN} by no more than 4.9 % within the observed RH range. This test addresses numerical stability within the data range, not thermodynamic interpretation.

Figures 5 and 6 show the hourly regression and RH-bin estimates used to identify the empirical response. Figure 7 shows the continuous campaign-specific fits applied in the flux conversion.

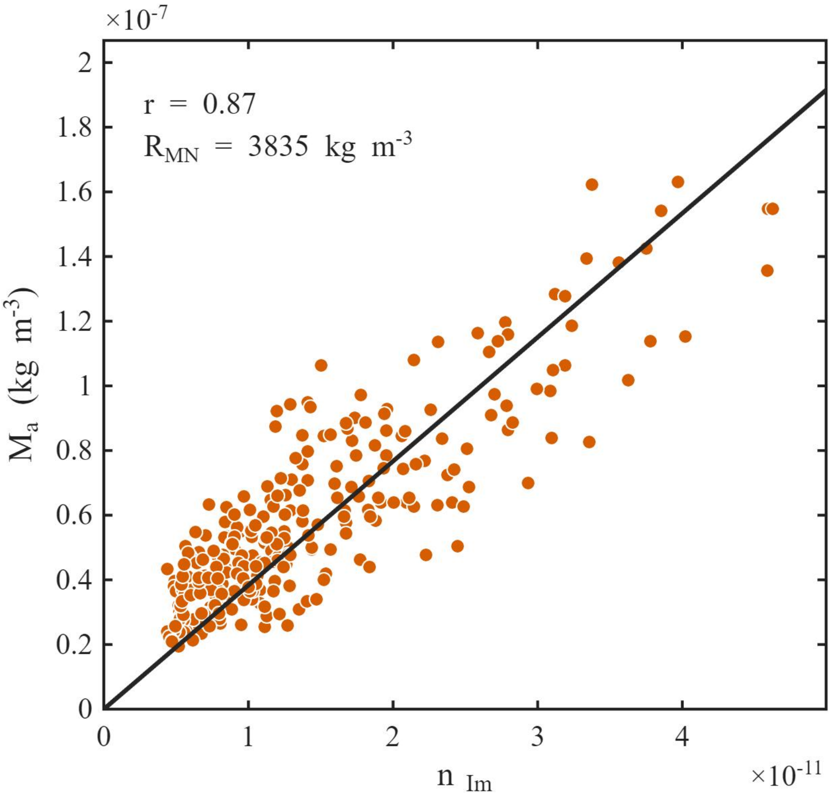

Figure R4 (reproduced from the revised manuscript Fig. 5). Example determination of R_{MN} for the 60–70 % RH interval during Siyuan Lake II. Orange points denote the hourly aerosol mass concentration M_a and the corresponding visibility-derived imaginary refractive index n_{Im}. The black line represents the linear regression of M_a against n_{Im}.

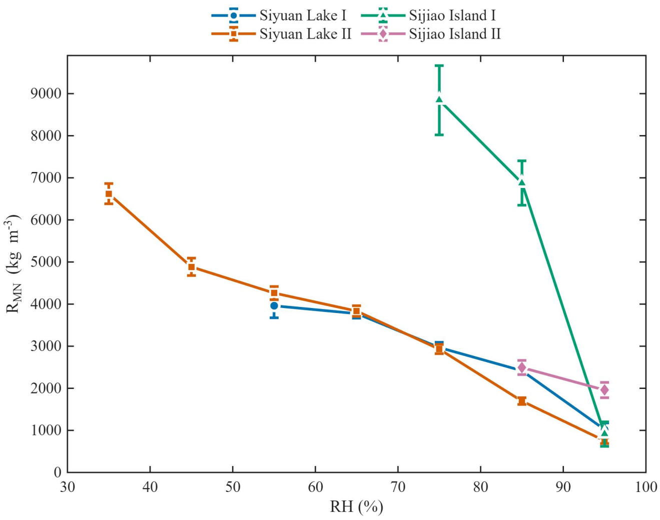

Figure R5 (reproduced from the revised manuscript Fig. 6). Variation of RH-bin R_{MN} estimates during the four observational campaigns. Symbols denote the R_{MN} estimates for individual RH intervals during Siyuan Lake I, Siyuan Lake II, Sijiao Island I, and Sijiao Island II, plotted at the nominal center of each RH bin; vertical bars show the corresponding 95 % confidence intervals.

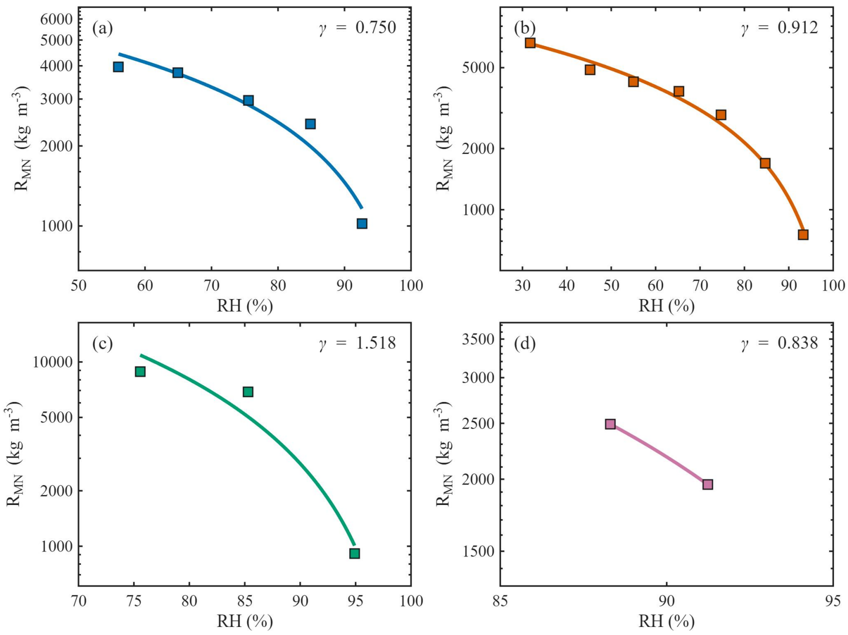

Figure R6 (reproduced from the revised manuscript Fig. 7). Continuous R_{MN} (RH) fits for **(a)** Siyuan Lake I, **(b)** Siyuan Lake II, **(c)** Sijiao Island I, and **(d)** Sijiao Island II. Squares denote the R_{MN} values obtained from the RH-bin analysis, and solid curves represent the campaign-specific fits using Eq. (7). The measured mean RH of each interval was used in the fitting. The fitted humidity-response parameter γ is shown in each panel.

We have also corrected the physical descriptions of the refractive-index components. The imaginary component represents aerosol-related attenuation by scattering and absorption and is not restricted to light-absorbing aerosol. The real component is described as thermodynamic and can include both temperature- and water-vapour-related refractive-index fluctuations.

The Sect. 2.3, Page 7, lines 156-158:

"Following Yuan et al. (2016, 2019), aerosol mass concentration is approximated as PM10…the atmospheric equivalent refractive index is expressed through the R_{MN}"

The Sect. 3.3, Pages 17-19, lines 379-429:

"Conversion of $C_{n,\mathrm{Im}}^{2}$ to the aerosol mass-concentration structure parameter requires the mass-to-optical conversion…vertical bars show the corresponding 95 % confidence intervals."

Comment 6: The filter-conditioning procedure requires justification. Conditioning glass-fibre filters for only 3 h at 45 °C without reported relative humidity does not conform to established ambient-PM reference procedures. EN 12341:2023 requires conditioning at 19–21 °C and 45–50% RH for at least 48 h and a balance resolution of no more than 10 µg. The relevant US EPA PM10 and $PM_{2.5}$ methods require at least 24 h of controlled temperature and humidity conditioning; the $PM_{2.5}$ method additionally specifies PTFE filters and microgram-level weighing. Heating loaded filters to 45 °C may remove ammonium nitrate, semi-volatile organic matter and particle-associated water, biasing the measured mass.

The authors should identify the standard or validated protocol followed and report the filter manufacturer, material and dimensions; sampling duration and sample numbers; whether $PM_{2.5}$ and PM_{10} were sampled simultaneously; flow calibration; transport and storage conditions; field and laboratory blanks; repeated weighings and mass-stability criteria; detection limits; and measurement uncertainty. If $PM_{2.5\text{-}10}$ was obtained by subtracting parallel $PM_{2.5}$ from PM_{10} measurements, this must be stated and the uncertainty propagated.

The deposition velocity calculation is not reproducible. Explicit equations are needed for aerodynamic and surface resistances, Brownian diffusion, inertial impaction and settling velocity because the cited formulations are not identical. All assumptions should be reported, including particle size distribution, density, shape factor, slip correction, hygroscopic growth, surface roughness, friction velocity, stability treatment and water-surface collection efficiencies. The numerical deposition velocities used for each fraction must also be given. Assigning one representative velocity to $PM_{2.5}$ and another to $PM_{2.5\text{-}10}$ may produce large errors because deposition velocity varies by orders of magnitude within these broad size ranges.

The filter-based calculation combines measured concentration with parameterized deposition velocity and is therefore a model estimate rather than an independent direct flux measurement. It should not be used as decisive validation of the LAS method. The authors must also show that the stated balance precision of 0.1 mg supports deposition fluxes reported to four decimal places after propagation of mass, volume and model uncertainties.

Response: We thank the reviewer for this comment. We agree that the former description did not distinguish the study-specific heating step from the controlled equilibration and weighing procedure. The filters were first pre-conditioned at 45 ± 1 °C for 3 h to establish a consistent dry baseline and were then equilibrated at 20 ± 1 °C and 50 ± 5 % RH for at least 24 h before weighing, following the weighing-related provisions of HJ 618-2011. The same procedure was applied to the PM2.5 and PM10 filters. Because the heating step is a study-specific modification, the resulting values are described as conditioned filter-based aerosol mass concentrations rather than EN 12341, US EPA, or other regulatory reference-method measurements. Possible loss of semi-volatile material during heating and adsorption of gas-phase organic compounds on bare glass-fibre filters are acknowledged as potential influences.

The revised methods identify the TH-150H sampler, nominal 100 L min−1 operation, $PM_{2.5}$ and PM_{10} impactors, nominal 2 h sampling periods, 90 mm Whatman GF/A glass-fibre filters, cumulative standard-condition sample volume, handling and individual bagging, field and laboratory blanks, conditioning conditions, and the EAB 125i balance. Valid $PM_{2.5}$ and PM_{10} concentrations were available for all 46 accepted sampling periods, of which 41 contained at least one concurrent valid 30 min LAS interval. In the dry-deposition calculation, $PM_{2.5}$ represents the fine fraction and PM_{10}−$PM_{2.5}$ represents the coarse fraction. The PM_{10} sample-level quality-control records are provided in Supplementary Table S1a. The temporally matched comparison of accepted filter-based and monitoring-station PM_{10} observations is documented in Supplement Sect. S3 and Fig. S3. The 46 matched periods were strongly correlated ($r = 0.918$, $p < 0.001$), while the filter measurements showed a positive mean offset of 16.327 μg m^{-3} relative to the station values. The station record is therefore used to represent temporal variation during periods without concurrent filter measurements rather than to assume identical absolute concentrations.

The TH-150H was operated using its built-in flow meter and recorded cumulative standard-condition volume. No independent flow-calibration instrument, calibration-frequency record, or calibration-bias record is available. Post-transport storage temperature and storage duration were likewise not recorded accurately. Blank-mass correction, duplicate weighing, the <0.4 mg mass-stability criterion, the method detection limit, an operational quantification threshold based on the observed sample volumes, and the propagated weighing-repeatability contribution are now reported. These limitations apply to both PM2.5 and PM10 measurements and are retained when the concentrations are used in the dry-deposition calculation.

We agree that the filter-based dry-deposition calculation is a model estimate rather than a direct flux measurement and that the deposition-velocity calculation must be traceable. The revised calculation uses the non-vegetated-water-surface parameterization of Petroff and Zhang (2010) as a single, internally consistent scheme. Its expressions for aerodynamic transfer, gravitational settling, Cunningham slip correction, Brownian diffusion, turbulent impaction, stability, roughness, and liquid-water collection efficiency are applied as defined in that study and are therefore cited rather than reproduced individually. Size-dependent deposition velocity was evaluated over 0.05–2.5 μm for the fine fraction and 2.5–10 μm for the coarse fraction and integrated over the corresponding lognormal mass distributions; $PM_{2.5}$ and $PM_{2.5\text{–}10}$ were not assigned single arbitrary velocities. The central distributions from Sayer et al. (2012) used volume-median diameters of 0.314 and 5.18 μm and natural-log widths of 0.50 and 0.72 for the fine and coarse modes, respectively, with spherical particles of density 1500 kg m^{-3}. Each 30 min calculation used the concurrent friction velocity, Obukhov length, air temperature, and pressure. Across the 154 matched intervals, the resulting fine- and coarse-fraction mass-weighted deposition velocities had medians (5th–95th percentiles) of 6.9×10^{-5} (6.4×10^{-5}–7.8×10^{-5}) and 1.83×10^{-3} (1.61 × 10−3–6.59×10^{-3}) m s^{-1}, respectively. Sensitivity to the unmeasured particle-size distributions and density is reported in Supplement Sect. S9, Table S7a and Fig. S11.

The former statement of 0.1 mg balance precision has also been replaced by the manufacturer-specified readability of 0.01 mg, repeatability of 0.03 mg (standard deviation), and linearity of ±0.08 mg within the ≤62 g semi-micro range. The revised methods further report the duplicate-weighing criterion, operational minimum loading, and propagated weighing-repeatability contribution. The dry-deposition median is reported as 0.058 μg m^{-2} s^{-1} rather than to four decimal places.

The revised comparison between LAS and dry-deposition calculation contains 41 filter samples matched to 154 valid 30 min LAS intervals. The Pearson correlation between the base-10 logarithms of the LAS and dry-deposition fluxes was 0.14 under the constant conversion and 0.53 under the RH-dependent conversion. The corresponding median LAS magnitudes were 0.086 and 0.099 μg m^{-2} s^{-1}, compared with a dry-deposition median of 0.058 μg m^{-2} s^{-1}. Because measured concentration is combined with a parameterized deposition velocity, this calculation is treated as a process-based reference and not as an independent flux validation.

The comparison is reproduced below to show the scale, pairing, and effect of the RH-dependent conversion.

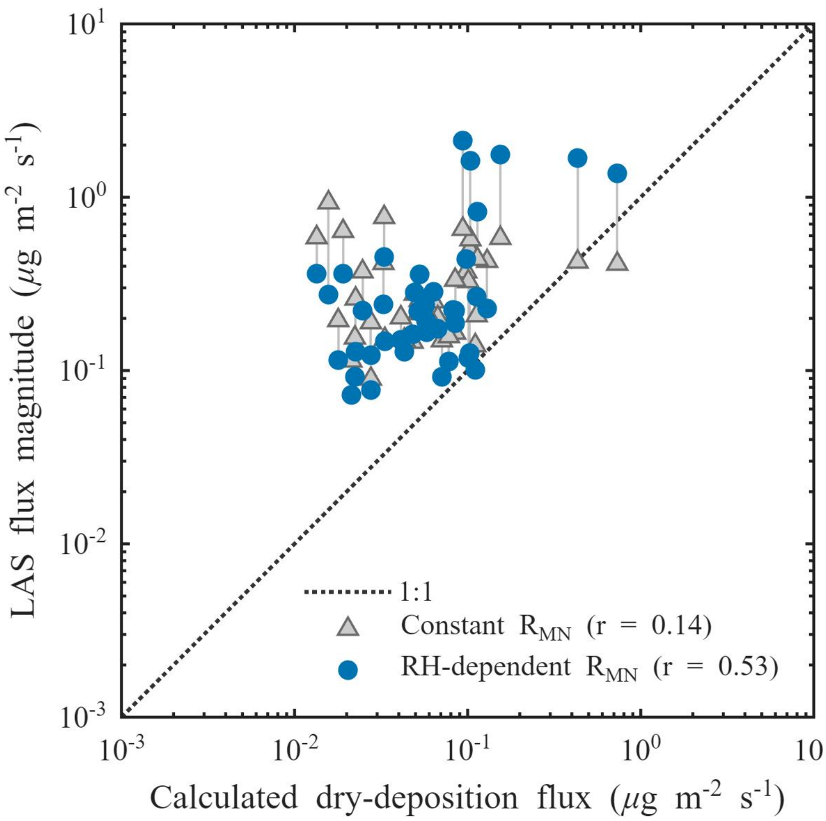

Figure R7 (reproduced from the revised manuscript Fig. 13). Comparison of the calculated dry-deposition flux with LAS-derived aerosol mass-flux magnitudes obtained using a constant R_{MN} of 3845 kg m−3 (gray triangles) and the RH-dependent R_{MN} (blue circles). Gray lines connect the two LAS calculations, and the dotted line denotes 1:1 agreement. Both axes are logarithmic.

The Sect. 2.1, Pages 4-5, lines 108-124:

"Filter-based aerosol sampling was also conducted during the four field campaigns…Potential biases from semi-volatile particle loss during heating and adsorption of gas-phase organic compounds on the bare glass-fiber filters cannot be excluded."

The Sect. 2.7, Page 11, lines 271-281:

"Quality control of the filter-based $PM_{2.5}$ and PM_{10} measurements followed the weighing-related provisions of HJ 618-2011…equivalent to 3.6–4.1 μg m^{-3} over the observed range of sampled-air volumes."

The Sect. 2.6, Pages 9-10, lines 221-239:

"A dry-deposition estimate was constructed as a physical reference for the LAS-derived aerosol mass-flux magnitude…where the PM2.5 and PM10 terms denote the corresponding concentrations."

The Sect. 3.4, Pages 26-27, lines 538-559:

"The dry-deposition calculation provided an independent reference for the LAS-derived flux magnitude…Both axes are logarithmic."

Comment 7: The reported meteorological averages are insufficient to assess whether the flux-method assumptions were satisfied. Mean wind speeds of approximately 0.96–1.19 m s-1 indicate that weak-turbulence periods may have been common during measurements. Campaign distributions should be provided for wind speed and its variability, friction velocity, standard deviations, turbulent kinetic energy, z/L, stationarity, water temperature, air–water temperature difference and boundary-layer height. Wave height or another sea-state indicator might be also useful at the coastal site. Because boundary-layer height is an input to the footprint model, its source and associated sensitivity should be documented.

The abstract and conclusions describe the method as operationally useful without specifying its performance envelope. The authors should provide a table defining the ranges of wind speed, turbulence, stability, relative humidity, extinction and received signal for which valid retrievals were obtained, together with data availability and rejection rates. Fog should be defined using a reproducible signal or extinction threshold rather than treated qualitatively.

Response: We thank the reviewer for this helpful suggestion. Table 3 now reports the observed 5th–95th percentile operating ranges of mean wind speed, friction velocity, stability, RH, extinction coefficient, and mean received-signal voltage among final valid 30 min flux retrievals. It also reports candidate intervals, retained AERISP intervals, final valid flux counts and availability, and mutually exclusive first-failure rejection counts and rates for precipitation, fog, received signal, spectral/AERISP retrieval, and other quality control.

Fog is now defined reproducibly as visibility below 1 km. Received-signal failure is defined separately as a 30 min mean LAS channel-1 voltage below 0.005 V or below 2 % of the campaign-specific reference, where the reference is the 90th percentile of window-mean voltage among intervals with at least 98 % raw LAS coverage. The table therefore distinguishes hydrometeor screening from loss of optical signal and from unsuccessful spectral separation.

The fuller distributions of u_* , z/L, wind variability, and other turbulent statistics are provided in Supplement Sect. S4, Table S2, and Fig. S4; the stationarity and integral-turbulence diagnostics are provided in Sect. S5, Tables S3a–S3b, and Figs. S5–S6; and the boundary-layer-height sensitivity of the footprints is provided in Sect. S6, Table S4, and Figs. S7–S8. Water temperature, air–water temperature difference, wave height, and whitecap coverage were not measured and are identified in Supplement Sect. S11. The boundary-layer height used in the primary footprint calculation is fixed at 1000 m because time-resolved measurements were unavailable.

The revised Abstract and Conclusions now state explicitly that LAS is feasible for retrieving path-averaged aerosol mass-flux magnitudes in open-water environments under adequate optical-transmission conditions.

The Sect. 3.4, Pages 21-22, lines 458-469:

"Final flux-retrieval availability was 83.9 %, 83.1 %, 45.9 %, and 97.7 %…rejection categories correspond to the first failed quality-control criterion."

The Sect. 4, Page 28, lines 587-592:

"These observations demonstrate that LAS can provide spatially integrated measurements…when optical conditions permit."

Comment 8: Validation statistics must be calculated from strictly paired LAS and EC observations. If EC measurements continued while LAS data were unavailable during fog, the unpaired periods should not affect comparisons of campaign means, MBE, RMSE or NRMSE. They may instead be used to quantify differences in operational availability.

The exact NRMSE formula and normalization, sample number, median, standard deviation, quartiles, range and confidence intervals should be reported for every comparison. Because fluxes span approximately two orders of magnitude, their distributions should be assessed before using arithmetic means; median, geometric-mean or logarithmic analyses may be more appropriate. Paired statistical tests, regression slopes and intercepts with uncertainty, and methods accounting for error in both variables, such as Deming or orthogonal regression, should be considered. Bland–Altman analysis, a concordance metric and treatment of temporal autocorrelation would provide a more informative method comparison. Campaigns should first be assessed separately before pooling.

Improved agreement after 2 h averaging may result from smoothing and does not demonstrate that averaging resolves footprint differences. The 30 min data should remain the primary validation, with 2 h results presented as a sensitivity analysis. Similarly, the difference between correlations of (r=0.442) and (r=0.346) does not establish superior LAS performance without confidence intervals and a test for dependent correlations. Barbecue-smoke periods should be handled using objective exclusion criteria or a predefined sensitivity analysis rather than only a post hoc interpretation based on field notes.

Response: We agree with the reviewer's statistical concern. However, the PWD50 temporal-response limitation means that no defensible aerosol EC reference can be constructed, so a paired LAS–EC validation is no longer attempted. All paired and unpaired campaign means, MBE, RMSE, NRMSE, regressions, confidence intervals for LAS–EC statistics, 2 h sensitivity tests, pooled analyses, Bland–Altman and concordance comparisons, and tests of dependent correlations have been removed. The former interpretation that agreement improved after temporal smoothing or because of footprint matching has also been deleted.

We agree that medians are more appropriate than arithmetic means for the broadly distributed LAS flux magnitudes because they are less sensitive to large values. The revised analysis uses the physically defined 30 min LAS retrieval window without a post hoc 2 h averaging choice. Campaign distributions are summarized by medians and interquartile ranges, and daytime/nighttime contrasts are calculated from medians. Candidate and final valid sample counts are reported separately for each campaign in Table 3, reproduced in our response to Comment 7.

The new dry-deposition analysis is separate from the withdrawn LAS–EC validation and is used only as a process-based reference. The four campaigns are pooled because the analysis is a controlled paired comparison of two R_{MN} treatments applied to the same 41 filter-sampling periods, with all other retrieval inputs held unchanged. Arithmetic means of concurrent valid 30 min values represent each filter-sampling period. Across those period means, the $\log_{10}$ Pearson correlation increased from 0.137 to 0.532; the correlation difference was 0.395, with p = 0.0179 from a paired label-swap test and a bootstrap 95 % confidence interval of 0.124–0.606. The RH-dependent treatment did not reduce the median absolute fold deviation, so these statistics are interpreted as stronger correspondence of variation rather than validation of absolute agreement. These statistics are reported in Supplement Sect. S9 and Table S7b.

The barbecue-smoke event was localized near the former point-based EC system and affected only the immediate vicinity of that instrument; it was not considered to have materially influenced the LAS signal integrated over the full optical path. Because the EC comparison has been removed, those EC points are no longer part of the revised analysis.

The revised primary results are reproduced below. At Siyuan Lake, the campaign medians were 0.091 and 0.123 $\mu g\ m^{-2}\ s^{-1}$, with interquartile ranges of 0.058–0.150 and 0.075–0.231 $\mu g\ m^{-2}\ s^{-1}$. At Sijiao Island, the medians were 0.399 and 0.114 $\mu g\ m^{-2}\ s^{-1}$, with interquartile ranges of 0.174–0.936 and 0.073–0.161 $\mu g\ m^{-2}\ s^{-1}$. The figures retain the accepted 30 min records and show gaps at failed quality-control intervals; campaign comparisons use medians and interquartile ranges rather than 2 h averaging or EC agreement.

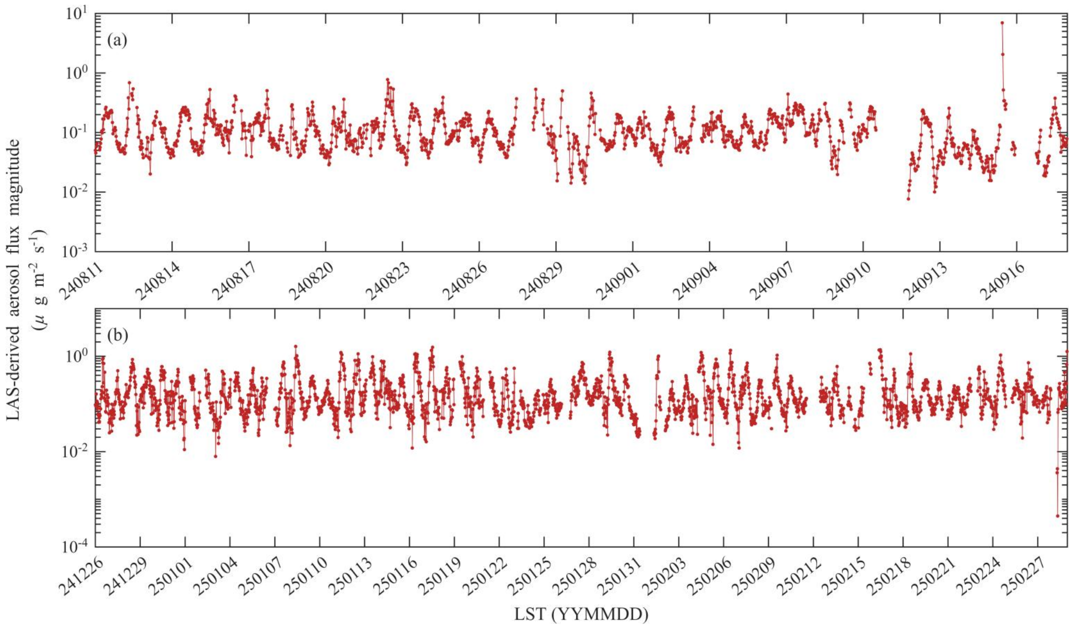

Figure R7 (reproduced from the revised manuscript Fig. 10). Temporal variations of the 30 min LAS-derived aerosol mass-flux magnitude during (a) Siyuan Lake I and (b) Siyuan Lake II. The vertical axis is logarithmic, and breaks denote intervals excluded by the quality-control criteria.

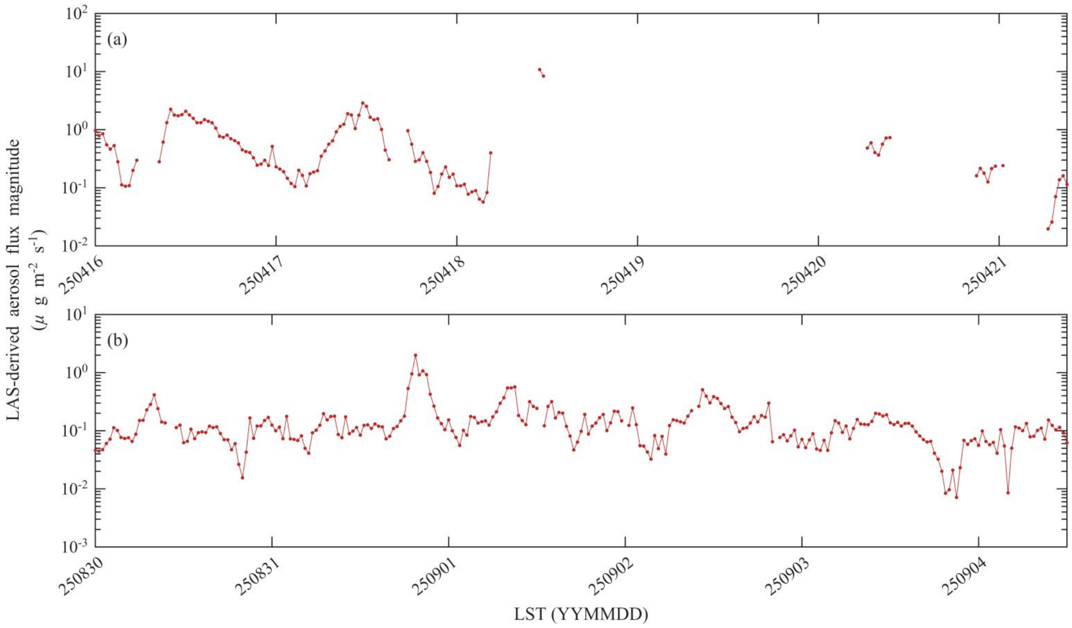

Figure R8 (reproduced from the revised manuscript Fig. 12). Temporal variations of the 30 min LAS-derived aerosol mass-flux magnitude during (a) Sijiao Island I and (b) Sijiao Island II. The vertical axis is logarithmic, and breaks denote intervals excluded by the quality-control criteria.

The Sect. 3.4, Page 24, lines 499-501:

"Figure 10. Temporal variations of the 30 min LAS-derived aerosol mass-flux magnitude…breaks denote intervals excluded by the quality-control criteria.”

The Sect. 3.4, Page 26, lines 535-537:

“Figure 12. Temporal variations of the 30 min LAS-derived aerosol mass-flux magnitude…breaks denote intervals excluded by the quality-control criteria.”

Comment 9: Daytime maxima and coastal events are repeatedly attributed to sea-spray emission or bubble bursting, but no aerosol composition, size distribution, sea-salt tracer, wave-state or whitecap data are presented. The urban lake and coastal bay may also be influenced by continental and local anthropogenic aerosol.

If sea-spray emission is proposed, fluxes should be analysed against wind speed while controlling for stability and water-footprint fraction. Separate upward and downward EC fluxes and PM2.5/PM10 partitioning would also be informative. Without compositionally or size-resolved evidence, the observed optical-flux variability should not be attributed specifically to sea spray.

Response: We agree that the available measurements do not support source-specific attribution. The revised text separates the observations—aerosol mass-exchange magnitude and the accompanying variations in u_* and M_*—from possible mechanisms. Definitive statements attributing daytime maxima or coastal events to sea-spray emission, wave breaking, or bubble bursting have been removed. The discussion now states that the coastal aerosol population may include marine, transported, and local contributions.

The wind-speed analysis cannot by itself establish a sea-spray source without wave state, composition, size distribution, or a directional flux. We therefore report the observed changes in wind speed, u_*, stability, M_*, and water-footprint fraction but do not interpret their covariation as source identification. A partial regression controlling for stability and footprint would not resolve the missing source tracers and would be especially underconstrained for the short coastal campaigns.

Separate upward and downward aerosol EC fluxes are unavailable because the PWD50-based EC calculation has been removed and the LAS retrieval preserves magnitude but not direction.

The Sect. 3.4, Page 23, lines 486-494:

“Both lake campaigns…did not account for the repeated daytime flux maxima.”

The Sect. 3.4, Page 25, lines 515-529:

"Both coastal campaigns…do not permit a definitive attribution of the underlying aerosol process."

Comment 10: "Available upon request" is inadequate for a methodological validation study whose conclusions depend strongly on signal processing, quality control, averaging and data selection. In accordance with FAIR principles, used data should be deposited in a repository with a persistent identifier. If public release is not possible during review, the authors should provide reviewer access and commit explicitly to publication of the data and code upon acceptance.

Response: We agree. 'Available upon request' has been removed. The revised manuscript supplies the persistent Zenodo identifier.

Comment 11: The manuscript contains many minor typos and glitches. I suggest a specialised English proofreader should check the text. I'm not qualified to check the language.

Response: We thank the reviewer for this recommendation. The manuscript has been rechecked for grammar, spelling, technical terminology, units, symbols, equation and figure references, and internal consistency.

Comment 12: Reference problems

- "Seinfeld and Pandis (2016)" is cited at line 154 but absent from the reference list.
- Wilczak et al. (2001) concerns sonic-anemometer tilt correction and does not support the aerosol covariance equation as cited.
- Yang et al. (2016) concerns sulfur dioxide attribution, not aerosol-particle EC fluxes.
- Kiehl and Briegleb (1993) is not an appropriate source for the statement that oceans cover 71% of Earth and are a major aerosol source.
- Twomey (1977) supports aerosol–cloud albedo effects, not the direct scattering/absorption statement as currently written.

Response: We thank the reviewer for identifying these reference problems. Each item has been checked against the statement it was intended to support.