Response to Anonymous Reviewer 2

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Original report: Anonymous Referee #2, 10 August 2026

Revised response based on the available observations.

Revised title: Do PM mass and aerosol optical properties differ between pollen-labeled impactor filters and dust-event-associated impactor filters at Hyytiälä?

We are grateful to Reviewer 2 for taking the time to read our manuscript carefully and provide detailed comments. We appreciate the thought and effort that went into the review. The suggestions have helped us improve the manuscript and explain our findings more clearly. We respond to each comment below.

Box category
- Reviewer comments
- Revisions, evidence and interpretation

Reading guide: quotations and original line references concern the submission; revised locations identify sections, tables and figures in the main manuscript (MM) and supplementary information (SI). Nominal, pointwise 95% percentile bootstrap intervals quantify sampling variability; measurement uncertainty is not separately propagated.

General comment 1. Independent pollen observations

> **Reviewer comment.** *The manuscript is well written, and the data analysis is methodologically sound, though some clarifications would be needed. However, I am not convinced that the measurement instruments used in this study really capture pollen concentrations. Could the validation of the result be done in collaboration with the Pollen forecasting unit at [UTU]?*

Response. Gravimetric and optical instruments do not directly measure pollen concentration. The nominal PM_{10} optical inlet limits large intact pollen; independently measured super-PM_{10} mass has no optical counterpart. Separate Hyytiälä Burkard samples were identified and counted by light microscopy, generally at genus or family level, by the UTU Aerobiology laboratory. They corroborate pollen during selected collections, not optical measurement of pollen concentration.

Manninen et al. (2014) report a sampler height of 20 m, approximately 3 m above the canopy, for 2003-2004; Pessi recalls the same tower through 2011 (personal communication, 2026). Six scanned April-May 2011 weekly records name Veijo Hiltunen as tape changer and log 10 L min^{-1} at the start and end. These handling and flow records do not independently verify the 2011 inlet position, concentration calibration, or sampling efficiency.

The pollen record contains 240 two-hour bins on 20 April-May 2011 days: 172 positive and 68 zero totals. We use fixed UTC+2 without daylight-saving adjustment. Odd-hour labels denote interval centers (01:00 represents 00:00-02:00); means and coverage use actual overlap with recorded collections. Observed zeros remain; missing days are unfilled. SI S8.1 documents this temporal support.

Seven retrospectively selected 2011 collections have complete two-hour pollen coverage and mean concentrations of 22.1-389 grains m^{-3}. They form three sampling blocks and were already in the original impactor inventory. Six unflagged neighbors also contain pollen during observed windows, but 13.8-33.4% coverage precludes complete-period negative controls.

This evidence establishes neither classification sensitivity or specificity nor stage-specific pollen presence. It cannot validate 2016-2022 note-only records: 55 of 76 eligible pollen-positive collections (72.4%) and 46 of 58 primary optical P collections (79.3%). Label-recording method and year coincide, preventing separation of their effects.

Pollen grains per gram divides separate, period-matched pollen concentrations by PM_{10} plus super-PM_{10} mass concentrations. It is descriptive, neither pollen mass fraction nor grains recovered from the impactor. The historical records above leave 2011 relative inlet position, transmission and calibration uncertain; temporal matching cannot establish comparable sampling efficiencies.

Changes in the main manuscript. Sects. 2.2-2.3; Sect. 3.4, Table 3 and Fig. 6; Sect. 4.2. Adds the independent observations, states the clock convention, and defines the descriptive ratio while limiting validation claims.

Changes in the supplementary information. S8.1-S8.3 and Table S5; S12, Table S7. Documents the microscopy and sampling records, temporal matching, partial controls, remaining metadata limits, and how pollen labels were recorded.

Pollen evidence and its scope. Exported concentrations equal 68.64 times the sum of four counted fields in 17 matches of exact date, taxon, and bin with four numeric inputs and zero residuals (SI S15.4). This verifies arithmetic, not the factor's physical area/volume basis, blank and year-field conventions, measurement uncertainty or exact 2011 relative inlet positions. Supplied concentrations and the descriptive cross-instrument grains-per-gram ratio retain these limits and the validation limits above; pollen mass fraction is not established.

Completed check. Label-recording and optical-processing-period checks (SI S15.3) retain 12 P and three D in flag-and-note years, with reportable direct intervals. Note-only years retain 46 P and five D, but 90.7% valid bootstrap draws fail the original 95% gate; direct intervals are withheld. Later-processing-period D occurs only in 2022 and fails the two-year gate. P-B intervals meet reporting gates in both periods, but differing interval outcomes do not test effect differences. The confounding between recording and year above remains; neither labels nor calibration are validated.

General comment 2. Organization and relevance

Reviewer comment. *The results lean heavily to massive Supplement, which makes it laborious to read. Could the readability to be increased with reorganizing figures and tables? Perhaps moving the NPF related text, irrelevant to dust and pollen, to supplement and bringing the more relevant results to main text.*

Response. The main manuscript brings together collection-category populations (Table 1), seven focused optical-property summaries (Sect. 3.1), PM-mass medians and distributions (Table 2; Figs. 3-4), independent pollen and grains per gram (Table 3; Fig. 6), full-range size distributions (Fig. 7), and seasonal and temporal sensitivities (Sects. 3.3, 3.6; Fig. 8). Processing and extended statistics remain in the SI; machine-readable tables retain complete wavelength comparisons.

The Introduction's three numbered questions receive corresponding Conclusion paragraphs, linked by the final Discussion paragraph. The original standalone sub-100 nm discussion and example figure are removed. Remaining small-particle text provides noncausal size context; SI S9 and S11.4 retain methods and scalar-definition limits.

Figure 1 separates filter labels, regional dust-calendar associations, and independent Hyytiälä pollen identified and counted by UTU. Figures 2-5 omit individual points, retain category counts, and use tighter y-axis ranges with shading only inside boxes. Their counts, and those in Figs. S2-S3, denote valid collection periods. Vector supplementary figures retain interval bars and coloured median markers without dark outlines. Across all

figures, complete sentences end with full stops; short labels do not. Numerical results are unchanged. Figure 6 places collection periods on a calendar axis, defines points as ratios of collection-average concentrations, and distinguishes duration segments from uncertainty intervals.

Changes in the main manuscript. Introduction; Sect. 2.4; Sect. 3; Sects. 4.4-5; Tables 1-3 and Figs. 1-8. Implements the organization and figure clarifications described above.

Changes in the supplementary information. S8-S9; S11; S14.2-S14.3. Groups supporting pollen, size, statistical, and optical details; Figs. S1-S3 replace the earlier dense diagnostic figure set.

Completed check. SI S10 removes the unused classifier procedure but retains its documented training-support failure; no optical classification accuracy was estimated or claimed.

Detailed comment 1. References at original lines 106-107

Reviewer comment. *Lines 106-107: How these references relate to pollen? Aren't they dealing with new particle formation?*

Response. The NPF studies distinguished repeated observations from atmospheric episodes; they establish neither pollen-specific definitions, grouping thresholds nor episode independence. Their placement could imply pollen identification, so we removed that argument and define collection units and temporal grouping directly.

Changes in the main manuscript. Introduction; Sect. 2.3; Sect. 2.5. Defines intact collection periods, temporal matching, and resampling units without relying on NPF classifications.

Changes in the supplementary information. S1-S2; S6; S11.2. Defines collection boundaries, midpoint labels, and alternative temporal groupings.

Detailed comment 2. Bootstrap blocks and collection intervals

Reviewer comment. *Lines 156-158: Did I understand correctly that the bootstrap blocks do not follow the impactor sampling intervals? How this affect the results? How it is defined which block the measurement value is allocated and are these numbers representative? Why not using the sampling intervals as blocks in bootstrap?*

Response. Calendar blocks are resampling units and need not coincide with impactor intervals; each intact collection remains one observation. Usable hourly optics receive

temporal-overlap weights over actual boundaries, yielding one value per variable per eligible collection. Midpoints assign collections to blocks and strata defined by year and month; they create neither independent daily PM-mass observations nor representativeness of full blocks or months. PM mass represents the recorded interval, optics its accepted common hours.

Reference uncertainty jointly resamples whole years across categories. Fixed 1, 2, or 3 day blocks assign each collection once by midpoint date to a grid anchored at 1 January 1970; boundary-crossing collections and stored measurements remain unsplit. Empty gaps remain; gap-aware correlations use actual calendar separations. Alternatives use median collection duration or cross-category runs separated by gaps of at least 24, 48, or 72 hours. Shared block and run multiplicities preserve joint temporal grouping (SI S11.2).

Resampling individual intervals preserves measurements but loses neighbor dependence. Two-day grouping has 1,079 occupied blocks for 1,080 collections, nearly collection-level resampling. Raw point effects are unchanged, but fine and cumulative P-B scattering rank intervals exclude zero with short blocks and include zero with whole years or runs. We do not privilege the narrowest interval.

For raw and month-adjusted 550 nm scattering with shared two-day grouping, fine and cumulative direct D-P rank intervals exclude zero while median-difference intervals include zero. Whole-year intervals include zero throughout: inference depends on effect measure and resampling design (MM Sect. 3.6; SI S11.2).

Only eight primary D collections span five years, four in 2022. Common-month D-P optics retain three D and 23–24 P depending on property, without reportable year-bootstrap intervals. Validity gates do not establish calibrated coverage. Block allocation cannot resolve unequal optical retention (42.1% D versus 81.7% P; appendix).

Changes in the main manuscript. Sect. 2.3; Sect. 2.5; Sect. 3.6 and Fig. 8. Specifies assignment and shows how grouping changes inference.

Changes in the supplementary information. S4.1; S6; S11.2, Table S6; S11.5. Documents overlap weights, shared resampling units, support gates, and unavailable common-month intervals.

Completed check. Paired-design mass and leave-one-dust-year-out checks (MM Sect. 3.6; SI S15.3) share raw and month-adjusted draws within each design, re-estimating monthly backgrounds each time. Whole-year adjusted P-D median differences are 2.74 [-0.46, 5.76] μg m^{-3} for coarse mass, whose rank interval remains positive, and 3.83 [1.65, 7.27] for super-PM_{10}, which remains positive under both designs. All five dust-year omissions cover all categories. Omitting 2022 leaves four D collections: all 550 nm scattering intervals and 520 nm absorption median-difference intervals include zero; absorption rank intervals remain positive only in fine and cumulative fractions. PM_1 MAC_{520} also loses rank support for higher D: its D-P interval is $[-0.19, 1]$. Failed support gates are reported, not treated as agreement.

Handling-note sensitivity now quantified. Jointly omitting the five noted P collections (four in primary optics), with original support rules and shared resampling schedules (MM Sect. 3.6; SI S12 and S15.5), leaves positive month-adjusted super-PM_{10} P-D differences: 4.09 [1.52, 7.84] μg m^{-3} with whole years and 4.09 [2.14, 6.81] with two-day blocks. Adjusted coarse mass is 3.02 [-0.53, 6.24] with whole years; its rank interval now also includes zero. The PM_{10} 520 nm absorption D-P median difference stays 1.51 Mm^{-1}, but its whole-year interval becomes [-0.02, 2.96]; its rank interval remains positive. Thus some inferences change.

Small-denominator sensitivity. Here, P_p denotes the p^{th} percentile. Minimum and pooled P_1 and P_5 mass screens remove only B, leaving P-D comparisons unchanged by construction. Under the P_5 screen, fine B MSC's maximum falls from 1142.61 to 16.39 m^2 g^{-1}, its median from 3.23 to 3.21. Exploratorily omitting each category's smallest positive mass (SI S15.6) retains MSC rank and median intervals favoring D in all fractions, but fine MAC at 520 nm loses rank support: its D-P rank interval is [-0.18, 0.88]. All paired whole-year direct effect-shift intervals include zero. Changed zero exclusion or mechanically unchanged pooled-screen results demonstrate neither a change nor equivalence between effects.

These checks quantify influence, not measurement validity. The 18 May 2016 printer-off note has stored weights; notes establish neither invalidity nor final QC acceptance. Revised primary selection and figures remain the reference. The recovered 660–637 nm fit does not independently validate every wavelength or era; screening thresholds are not detection limits. Bootstrap intervals have no physical error budget (reading guide). The efficiencies MSC_λ and MAC_λ describe sampled mixtures, not intrinsic pollen or dust optical constants.

Detailed comment 3. Cliff's delta and effect magnitude

Reviewer comment. *Line 158: As Cliff's δ is not among the commonly known methods, you should briefly introduce it and justify its use. In addition, is there some threshold value where δ is considered small or large?*

Response. Cliff's delta (MM Sect. 2.5; SI S6, Eq. (S13)) compares two randomly selected category observations:

$$\delta(A, B) = \Pr(A > B) - \Pr(A < B).$$

Values range from -1 to 1; ties contribute zero and positive values favor the first category. Rank dominance requires no normality assumption and complements physical-unit median differences. Without ties, $\delta = 0.4$ means 70% of between-category pairs favor the first category, not a 40% concentration increase. Delta does not measure source contributions, and ranks do not remove temporal dependence.

Generic small, medium, and large thresholds do not establish physical importance, so magnitude and uncertainty are reported separately. Zero rank dominance need not mean identical distributions; rank and median intervals can differ in zero exclusion. Under the reading guide's nominal pointwise uncertainty, agreement across wavelengths is not independent confirmation.

Changes in the main manuscript. Sect. 2.5. Gives the probability definition and direction of Cliff's delta, its physical-unit complement, and the pointwise uncertainty and inference limits.

Changes in the supplementary information. S6, Eq. (S13). Defines the pairwise statistic, avoids generic magnitude thresholds, and states inferential limits.

Detailed comment 4. Aitken-particle concentrations

Reviewer comment. *Figure 5: It is hard to believe that pollen would increase the number concentration of Aitken mode particles, as indicated in panel a.*

Response. Higher Aitken-mode number concentrations do not establish pollen production of these much smaller particles. Labels are independent of Aitken number but can select seasons, weather and mixtures; category differences establish neither a common source nor a causal pollen effect.

Figure 7 presents full-range number, projected-area and volume category means without whole-curve significance claims. Secondary material, transported mixtures and seasonal and meteorological covariation remain possible, unidentified explanations. A published size convention cannot verify the separately supplied Aitken scalar's historical integration limits.

Changes in the main manuscript. Sect. 2.6; Sect. 3.5; Fig. 7. States the size assumptions and descriptive, noncausal interpretation.

Changes in the supplementary information. S9; S11.4. Distinguishes the converted full-range curves from the separately supplied scalar descriptors and their metadata limitations.

Interpretation of the supplied size scalars. Original scalar integration limits and geometric definitions remain unrecovered. We use operational descriptors under declared units, without unverified bin edges or geometric meanings; the separate heuristic is not their assumed generator. The full-spectrum curves above use documented diameter, projected-area and volume transformations (MM Fig. 7; SI S9), which do not independently validate the scalars. Both retain the noncausal interpretation above.

Detailed comment 5. Redundancy of original Section 3.4

Reviewer comment. *Section 3.4. is rather redundant if the focus in the manuscript is in dust and pollen.*

Response. As General comment 2 explains, we removed the standalone sub-100 nm section and example figure. Section 3.5 retains only accompanying small-particle context and its limitations.

Changes in the main manuscript. Sects. 3.4-3.5. Replaces the original standalone process discussion with the selected pollen evidence and a concise size-distribution interpretation.

Changes in the supplementary information. S9 and S11.4. Retains the methods and interpretation limits described in Detailed comment 4; earlier NPF-related panels are omitted.

Current documentation and reproducibility. MM Code and data availability and AI-use sections; SI S14.4-S15 and AI-use section. Both documents include AI-use sections and a version-pinned Binder link. Verification on 27 October 2026 completed four hosted jobs and exactly reproduced eight main and three supplementary images, including $\sigma_{abs,520}$, MAC_{520}, revised labels, numerical formatting and Figure 6. The GitHub release includes matching data and code archives and the receipt. The supplied local archives also include the subsequent figure wording and punctuation updates. The default workflow uses frozen processed inputs, replays the dust-overlap diagnostic and retains saved bootstrap intervals. It does not rerun raw-data analysis or the locally run reconciliation and sensitivity scripts supplied separately in *Evidence-Resolution*.

Appendix. Record reconciliation and selection checks

Both versions compare optics and PM mass over impactor collection intervals. The recovered original run reproduces these totals and links records to the revised inventory. B = neither indicator, P = pollen only, D = dust only, P+D = both.

Population	B	P	D	P+D	Total
Original merged inventory	1,545	21	15	0	1,581
Original finite PM mass, each fraction	1,527	21	15	0	1,563
Revised eligible inventory	1,474	71	19	5	1,569
Revised primary optics	1,010	58	8	4	1,080
Revised PM_1 mass	1,457	70	19	5	1,551
Revised $PM_{1\text{-}10}$ mass	1,425	70	19	5	1,519
Revised PM_{10} mass	1,449	70	19	5	1,543
Revised super-PM_{10} mass	1,389	71	19	5	1,484
Revised full-range size spectra	1,454	71	19	5	1,549

Count sources: original SI Tables S1 and S4; revised MM Table 1. Derived optics may have fewer finite values than the primary optical population. Fraction-specific populations overlap and must not be summed.

Pollen ascertainment. Original SI Table S15 required flag 555. Revised SI Table S7 records 21 flag-and-note collections in 2011-2014 and the 55 note-only cases detailed in General comment 1: **76 pollen-positive = 71 P + 5 P+D**. That answer gives primary optical P counts by recording method; four further pollen-positive optical collections are

P+D. The seven fully covered collections match original 2011 entries P1-P7 within timestamp precision; independent pollen data add no impactor collections. Of the original 21 P, 20 remain P and one becomes P+D; of 15 D, 12 remain D and three become P+D. The 55 note-only additions comprise 51 original B becoming P, one B becoming P+D, and three D becoming P+D.

Dust ascertainment. The 24 revised dust-positive collections (19 D + 5 P+D) overlap 18 catalog dates; the 12 primary optical dust-positive collections overlap nine. With the older calendar, replacing the midnight-within-interval test with positive full-day overlap changes 15 to 20. Restoring 2 February 2014, 6 June 2014 and 11 February 2018 changes 20 to 24 because 6 June overlaps two collections. This separates calendar and overlap-rule effects without altering the complete-calendar baseline or establishing receptor composition.

Measurement selection. Primary optics requires at least 75% coverage for ten channels in both inlets and valid paired PM_1, PM_{10}, and coarse mass (revised MM Sects. 2.3-2.5; SI S4-S5). Original PM_1 and coarse scattering counts were 1,481 (1,447 B, 21 P, 13 D) and 1,474 (1,440 B, 21 P, 13 D; original SI Table S4). These selections differ from revised fraction-specific mass selection, which excludes negatives. In PM_1, coarse, PM_{10}, and super-PM_{10} order, removing 2, 28, 5, and 69 negatives from 1,563 gives 1,561, 1,535, 1,558, and 1,494 (original SI Table S7), still 10, 16, 15, and 10 above revised totals. The crosswalk resolves these differences: each fraction loses ten no-foil zeros; PM_{10} and coarse mass withhold five inconsistent PM_{10} totals (rows 1508-1512); coarse mass also excludes row 2038, whose negative parents yield a positive subtraction. Both blanks were already missing; no replacement values were inferred. Size spectra independently require 75% complete-spectrum coverage (SI S9).

Category-specific optical retention. Percentages use revised MM Table 1. Mutually exclusive coverage and mass exclusion reasons appear below and in SI S15.1.

Category	Eligible	Primary optics	Excluded	Retained
B	1,474	1,010	464	68.5%
P	71	58	13	81.7%
D	19	8	11	42.1%
P+D	5	4	1	80%

Aggregate balance. 76 + 24 - 5 = 95 revised collections carry either indicator, versus 36 originally. The 59 additional labeled and 71 fewer background collections give a

net inventory reduction of 12. The exact-time crosswalk establishes 12 removals and no additions: rows 1621-1630 had no foils; rows 1438 and 1460 were blanks. The original 23 APSD episodes (15 dust and eight pollen) were temporal clusters, not collection counts; the seven fully pollen-covered collections form three blocks.

Completed check. All 1581 original records match unique revised workbook intervals exactly. The removals detailed above are all old B; no records were added. The crosswalk identifies category transitions, all five overlap records, dust-calendar effects and every fraction-specific mass-count difference (SI S15.1). This numerical consistency cannot independently certify the exact originally submitted file version.

Completed check. Category-specific exclusions and common-population contrasts (SI S15.1-S15.2) show B losses of 415 for coverage only, 44 for paired mass only and five for both; P losses are 12 and one respectively; all 11 D and one P+D losses concern coverage. Mass and optics share collection IDs for each property. With finite 550 nm scattering in the primary all-channel population (58 P and eight D), coarse mass has a P-D median difference of 3.74 [-1.16, 6.61] μg m^{-3} and a positive rank interval; scattering's median difference is -2.61 [-5.38, 0.3] Mm^{-1}. All 93 intervals for changes in each mass-effect measure between full and matched populations include zero. This establishes neither equivalence nor absence of selection bias; missing optics remain unknown.