Airborne pollen, wildfires, and Saharan dust control atmospheric carbohydrates and shape their biogeochemical impacts in northern Mediterranean

Milinković et al.,

The manuscript entitled "**Airborne pollen, wildfires, and Saharan dust control atmospheric carbohydrates and shape their biogeochemical impacts in northern Mediterranean**" by Milinkovic et al. contains potentially valuable information on the sources and deposition of atmospheric carbohydrates at a central Adriatic coastal site (Martinska, Šibenik). The authors compare PM_{10}, wet deposition, and total deposition during high-pollen periods, Saharan dust intrusions, and biomass-burning events. Their main objective is to determine how these events influence carbohydrate composition and fluxes and to estimate the potential contribution of atmospheric deposition to coastal seawater carbon. The principal reported results are that pollen periods were associated with higher primary sugars, biomass burning with higher anhydrosugars and dissolved carbohydrates, and Saharan dust mainly with increased PM_{10} mass but limited carbohydrate enhancement. The authors further use deposition calculations to estimate possible short-term inputs of water-soluble organic carbon (WSOC) and carbohydrate carbon to the Adriatic coastal surface layer.

However, substantial revision would be required before the principal conclusions can be considered adequately supported. The main concerns involve overinterpreting statistical associations as evidence for source identification, insufficient statistical analysis, large uncertainties in deposition-flux calculations, and unsupported claims of biogeochemical impact. In several instances, the cited literature does not appear to support the statements in the manuscript, and in some cases, the cited studies report different values or even conclusions that are at odds with those presented. The references should therefore be checked systematically and used with greater accuracy. In addition, the Adriatic Sea should be considered not only as a receptor but also as a potential source of atmospheric carbohydrates through sea-spray production and enrichment of the sea-surface microlayer. Without paired measurements of seawater, the sea-surface microlayer, sea-spray aerosol, or marine tracers, the marine contribution cannot be excluded. The ecological relevance of the calculated atmospheric input also remains unclear because the study does not compare the deposited carbohydrate flux with the existing carbohydrate pool in the receiving seawater. If atmospheric deposition accounts for only a very small fraction of the ambient marine carbohydrate inventory, the claimed biogeochemical significance would need to be substantially moderated, and the scientific importance of the study more clearly justified.

Based on the general and detailed comments and observations provided below, I consider that both the data interpretation and the overall discussion require substantial revision and condensation. The concentration data for carbohydrates and the other measured parameters may still constitute a valuable contribution to Atmospheric Chemistry and Physics, provided that the conclusions are restricted to what is directly supported by the measurements.

However, the broader hypotheses and source-attribution framework proposed in the manuscript would require a more robust and comprehensive experimental design. In particular, future work should include direct assessment of the Adriatic Sea as a potential source of atmospheric carbohydrates, for example, through paired measurements of seawater, the sea-surface microlayer, sea-spray aerosol, and appropriate marine tracers. Without such evidence, the central source and biogeochemical interpretations remain insufficiently supported.

General comment on the title

The title does not appear to be fully consistent with the reported results. The phrase “airborne pollen, wildfires, and Saharan dust control atmospheric carbohydrates” implies that all three event types exert a substantial control on atmospheric carbohydrate abundance. However, the manuscript reports that Saharan dust episodes primarily increased PM_{10} mass (+183%), whereas WSOC and most carbohydrate classes did not differ significantly from background levels, and the authors conclude that Saharan dust had a limited influence on carbohydrate concentrations and deposition. Saharan dust, therefore, appears to represent a contrasting event with limited impact on carbohydrates rather than a controlling source. The title should be revised to reflect this distinction and should also avoid implying demonstrated biogeochemical impacts, which were not directly measured.

General comment on the Abstract

The Abstract requires substantial revision because it contains several imprecise terms, unsupported source attributions, and claims that extend beyond the measurements. It should clearly distinguish among carbohydrates, organic carbon (OC), organic matter (OM), and WSOC (or DOC), and consistently define the geographical scope as the central Adriatic coastal environment rather than the Mediterranean Sea as a whole. The Abstract also presents associations with pollen, fungal activity, Saharan dust, and biomass burning as demonstrated source contributions, although several of these interpretations are indirect and not independently validated. Finally, the calculated atmospheric inputs to seawater are hypothetical estimates based on assumed mixing conditions and a literature-derived DOC concentration; they do not demonstrate bioavailability, microbial stimulation, or biogeochemical impact. The Abstract should be rewritten to report the measured results first, clearly identify exploratory interpretations, and substantially moderate causal and ecological claims.

General comment on the Introduction

The Introduction requires substantial restructuring and clarification. At present, it moves through several loosely connected topics: atmospheric aerosol, OM, and OC, WSOC, atmospheric deposition, ocean stratification, carbohydrates in different Earth-system compartments, cloud microphysics, and marine biogeochemistry, without establishing a sufficiently clear progression toward the specific scientific question the study addresses. Consequently, by the end of the Introduction, the central motivation, the current state of knowledge, the unresolved research problem, and the relevance of the selected coastal site remain difficult to identify. The terminology and spatial scope are also inconsistent. The text shifts among OM, OC, WSOC, organic compounds, and carbohydrates without consistently distinguishing among compound mass, carbon mass, and analytically defined fractions. Similarly, it alternates between the global ocean, the Mediterranean Sea, marine systems, and coastal waters, although these environments differ substantially in stratification, mixing, atmospheric exchange, and biogeochemical response. The geographical and environmental scale relevant to the present study should therefore be defined consistently. Several background statements are overly broad or insufficiently qualified, including the classification of OC solely by water solubility, the occurrence of carbohydrates throughout the lithosphere, and the proposed enhancement of CCN activity by the solubility and abundance of carbohydrates in bioaerosols. These statements should be revised to use more precise atmospheric chemistry terminology and be linked directly to the measured quantities and processes investigated in this work.

The Introduction should be shortened and reorganized around a clear sequence:

(1) the relevance of atmospheric carbohydrate deposition to central Adriatic coastal environments;

(2) the established sources and deposition pathways of atmospheric carbohydrates;

(3) what is currently known about pollen, Saharan dust, and biomass-burning events;

(4) the specific knowledge gap concerning their comparative influence across dry, wet, and total deposition pathways at central Adriatic coastal sites; and

(5) explicit research questions or testable hypotheses.

The main problem with the Introduction is that it is broad but insufficiently focused: it presents much information, yet the specific causal chain and research gap are not clearly established. Therefore, I recommend a complete restructuring and rewriting of the Introduction. It should follow a clear logical sequence: scientific background, current state of the art, identification of the unresolved problem or knowledge gap, and a precise explanation of how the present study addresses this gap. The research motivation, spatial scope, hypotheses, and novelty should be stated explicitly, while broad or only indirectly relevant information should be shortened or removed.

General comment on the Materials and Methods

The Materials and Methods section requires major revision because several procedures are insufficiently described, key analytical and statistical assumptions are not justified, and methodological information is repeatedly mixed with results. In its present form, the section does not provide enough detail for independent reproduction or for a reliable assessment of the reported uncertainties. Essential information is missing for sample filtration and extraction; operational definitions of dissolved and water-extractable fractions; method validation; calibration; precision; limits of detection (LOD); recovery; matrix effects; trajectory configuration; event identification; deposition-flux calculations; and statistical analysis. In several cases, measurements obtained using different sample-preparation procedures and different operational size cut-offs are treated as directly comparable without adequate justification.

The analytical methods require clearer validation. The manuscript should consistently distinguish among PM_{10} WSOC, deposition DOC, total dissolved carbohydrates, dissolved monosaccharides, calculated polysaccharides, and water-extractable molecular biomarkers. Filtration and centrifugation do not define equivalent analytical fractions. Filter material, washing procedure, blanks, adsorption tests, extraction recovery, repeatability, calibration performance, LOQ, matrix-spike recovery, and measurement uncertainty should therefore be reported where applicable. For the hydrolysis-based carbohydrate method, the hydrolysis efficiency and potential monosaccharide degradation must be evaluated rather than assumed.

The trajectory and event-classification methods are also insufficiently reproducible. The authors should report the complete HYSPLIT configuration, arrival times and time zones, local-time-to-UTC conversion, trajectory-height justification, vertical-motion option, temporal matching to the 48 h samples, and objective criteria for marine, continental, mixed, Saharan-dust, pollen, and biomass-burning classifications. Listing satellite, model, and institutional data sources without specifying the exact products, variables, access method, processing steps, and decision rules is inadequate. The selected trajectories and satellite images may illustrate particular cases, but they do not by themselves establish seasonal representativeness or surface-level influence.

The deposition-flux calculations depend strongly on assumed deposition velocities, particle-size associations, collection periods, and operational definitions. These parameters, unit conversions, literature basis, and associated uncertainty should be stated explicitly. Because the compound-specific deposition velocities were not measured in this study, **sensitivity analyses are needed to demonstrate** how strongly the conclusions depend on these assumptions.

The statistical workflow likewise needs stronger justification. A general log transformation of all variables should not be applied solely because some datasets failed a normality test. The authors should report post-transformation diagnostics, censoring frequencies, group sizes, independence assumptions, treatment of multiple comparisons, and the reasons for selecting Welch's t-test and Pearson correlation. The use of LOD/2 substitution should be evaluated for potential bias. The PCA procedure must describe scaling, centering, correlation or covariance matrix selection, component-retention criteria, sample-to-variable ratios, treatment of missing and censored values, and solution stability.

Finally, the section should be reorganized to include only the study design, sampling procedures, analytical methods, calculations, quality assurance, and statistical workflow. Observed meteorological ranges, seasonal averages, identified event dates, wildfire descriptions, trajectory classifications, and similar campaign findings are results and should be moved to the Results section or to clearly identified supplementary tables. Overall, the Methods should be rewritten as a coherent, reproducible workflow, with each measured quantity clearly linked to its sample matrix, preparation procedure, analytical definition, validation parameters, calculation, and uncertainty.

General comment on the Results and Discussion

The Results and Discussion substantially overinterpret the available measurements. In many places, statistical associations, numerical differences, literature-based expectations, and event classifications are presented as if they constituted direct evidence of source identification or causation. They do not. The study measures PM_{10} mass, WSOC, selected carbohydrate biomarkers, and dissolved-carbohydrate fractions in deposition samples. It does not directly measure pollen-derived material on the PM_{10} filters, fungal spores, source-specific marine tracers, total organic matter, microbial uptake, or biogeochemical responses in seawater. The conclusions must therefore remain strictly limited to the measured quantities. The pollen attribution is particularly overstated. Airborne pollen concentrations were obtained independently from a pollen-monitoring network and were not measured directly in the PM_{10} samples. Most intact pollen grains are larger than 10 µm and would not be efficiently sampled by a PM_{10} inlet, although pollen fragments and subpollen particles may enter the PM_{10} fraction. The reported correlations therefore, show that PM_{10} glucose and related biomarkers were elevated during periods of high regional pollen counts; they do not prove that intact pollen was the dominant source of the measured PM_{10} carbohydrates. The manuscript should distinguish clearly among intact pollen, pollen-derived fragments, plant debris, soil biological material, and other seasonally covarying sources. More fundamentally, the Adriatic Sea is treated largely as a passive receptor of terrestrially derived carbohydrates, whereas its possible role as an atmospheric source is insufficiently considered. This is not justified at a coastal sampling site. Marine microorganisms, phytoplankton, macroalgae, dissolved and particulate organic matter, and especially the sea-surface microlayer contain carbohydrates that may be transferred to the atmosphere through bubble bursting and sea-spray aerosol production. Sea-spray particles, including supermicron particles within PM_{10}, may therefore contain glucose, polyols, and other organic compounds depending on seawater and microlayer composition. A “local” source inferred from polar plots cannot automatically be interpreted as terrestrial or fungal; it may also originate from the surrounding sea. The manuscript provides no paired measurements of seawater, the sea-surface microlayer, sea-spray aerosol, or marine tracers to evaluate this potential marine contribution. Nor does it adequately compare carbohydrate concentrations under clearly marine and continental air-mass conditions. Consequently, marine sources cannot be excluded, and the reported atmospheric deposition cannot be assumed to represent entirely new terrestrial carbon entering the Adriatic Sea. Part of the measured material may reflect local sea–air recycling. Similar overinterpretation occurs for Saharan dust, biomass burning, and fungal sources. Dust periods mainly show the expected increase in PM_{10} mass, whereas most carbohydrate changes are not statistically significant. Biomass-burning periods provide the clearest source-consistent result from elevated levels of anhydrosugars, but this does not establish control over all carbohydrate classes or total organic matter. Higher summer sugar-alcohol concentrations are consistent with fungal spore emissions, but fungi were not measured independently, and alternative terrestrial and marine biological sources remain plausible. Literature can support the plausibility of an interpretation, but it cannot replace site-specific evidence. The biogeochemical section extends these uncertain source assignments into claims of marine impact. However, no concurrent measurements of seawater carbohydrates, microbial activity, respiration, bacterial production, or biological response were made. The reported changes are hypothetical mixing calculations based on an assumed water depth and a literature-derived DOC concentration, not observed ecosystem effects. This section should therefore be reframed as an exploratory estimate of potential atmospheric input rather than a demonstration of biogeochemical significance.

The PCA interpretation introduces an additional major inconsistency. The manuscript does not clearly describe the PCA input matrix, scaling and centering, treatment of below-LOD values, the number of

observations, or whether spring and summer were analyzed separately or encoded as separate variables. Figure 6 appears to show only variable loadings or a correlation circle, not sample scores or a true biplot. It therefore cannot demonstrate separation of spring and summer samples, nor can it identify or quantify atmospheric sources. The inclusion of Total MB, along with its constituent carbohydrate groups, also introduces mathematical redundancy and may artificially strengthen the observed covariance structure. Because the first two components explain only 54.4% of the total variance, the two-dimensional interpretation is incomplete. PCA can reveal patterns of covariance, but it cannot confirm pollen, fungal, marine, dust, or biomass-burning sources without independent tracers or source-specific measurements. The PCA-based source assignments should therefore be substantially reduced or removed unless a complete and reproducible PCA workflow, score plots, numerical loadings, explained variance, and validation analyses are provided. Overall, the Discussion should be shortened and rewritten to clearly distinguish between what was directly measured, what was statistically associated, what was inferred from the literature, and what remains hypothetical. Terms such as "confirmed," "dominant source," "governed," "significant contribution," "bioavailable carbon," and "biogeochemical impact" should be avoided unless supported by direct and source-specific evidence. The defensible conclusion is that certain carbohydrate classes were associated with predefined pollen, dust, and biomass-burning periods, while the actual source contributions, including a potentially relevant marine component, remain unresolved.

Specific comments: Abstract

L9–10: The statement that carbohydrates are "ubiquitous, water-soluble organic compounds" is incorrect because not all carbohydrates are water-soluble. Please revise this to refer specifically to water-soluble carbohydrates or to clarify that the study addresses selected water-extractable and dissolved carbohydrate fractions.

The terminology is also unclear. Organic matter (OM) and organic carbon (OC) are not equivalent, and carbohydrates cannot generally be described as tracers of "atmospheric organic matter sources" without identifying which compounds trace which sources. For example, anhydrosugars may indicate biomass combustion, while sugar alcohols are commonly associated with biological material, but carbohydrates as a whole are not a single source tracer.

The statements that carbohydrates participate in cloud chemical processes and provide readily bioavailable carbon are too general. Please specify which atmospheric carbohydrate fractions or processes are intended. The present study did not investigate cloud chemistry or directly measure microbial bioavailability.

L11–13: "The Mediterranean Sea" is too broad for a study conducted at one central Adriatic coastal site. Please define the geographical scope consistently, for example, as the central Adriatic coastal environment. The study investigates periods classified as high-pollen, Saharan dust, and biomass-burning events, rather than directly measuring the three discrete sources. The objective should therefore be phrased as assessing how carbohydrate concentrations and deposition differed during these event periods.

L13–15: The relationship between the identified events and the deposition pathways is unclear. The Abstract should state whether the authors are comparing concentrations and calculated or measured deposition fluxes among event and background periods. It should not imply that the sources directly control the deposition mechanism unless this was specifically demonstrated.

L15–19: Statements that pollen was the "dominant source," that sugar alcohols indicated fungal contributions, and that biomass burning significantly increased dissolved carbohydrates require moderation. These findings are based mainly on event classification, correlation, biomarker interpretation, and limited sample numbers. They support source-consistent associations but do not constitute quantitative source apportionment. The phrase "highest anhydrosugars levels" should be corrected to "highest anhydrosugar concentrations."

L19–21: The wording concerning Saharan dust is internally unclear. The manuscript states that dust contributed insignificant organic-carbon fractions and had limited influence on carbohydrate deposition, but subsequently reports that pollen and dust episodes increased surface DOC by 0.2–0.7%. These

statements refer to different calculations and should not be presented as directly equivalent evidence. Moreover, the 0.2–0.7% values are not measured increases in surface-water DOC. They are hypothetical concentration increments calculated from atmospheric deposition under an assumed mixing depth and using a literature-derived ambient DOC concentration. This should be stated explicitly.

L22: The term "modest" is qualitative and undefined. Please report the numerical magnitude and clarify whether the comparison was statistically significant, environmentally relevant, or merely a model estimate.

L22–23: The claim that a high carbohydrate fraction indicates "labile" material and that episodic deposition delivers "important pulses of bioavailable carbon" is unsupported. Chemical lability and microbial bioavailability were not measured. These terms should be replaced by wording such as "potential atmospheric inputs of dissolved carbohydrate carbon."

L24: "Atmosphere–ocean interactions" is too broad and does not accurately describe the coastal Adriatic setting. Please specify whether the manuscript concerns atmosphere–sea or ocean exchange, atmospheric deposition to coastal or marine seawater, or coupled sea–air cycling. The Adriatic Sea should also be mentioned in the title.

Specific comments: Introduction

L27: The terminology is imprecise, and the sentence appears redundant. "Aerosol" refers to particles suspended in a gas, whereas particulate matter, including PM_{10}, is an operationally defined particle-size fraction. Please clarify what distinction the authors intend to make and revise the sentence accordingly. The plural form "aerosols" should be used only when referring to different aerosol populations, types, sources, or regions.

L29: Please define the abbreviation organic carbon (OC) once and use it consistently throughout the manuscript. At present, the terms organic matter (OM) and organic carbon (OC) appear to be used interchangeably, although they are not equivalent. OM refers to the total mass of organic compounds, whereas OC refers only to their carbon content. This distinction should be maintained consistently.

L30: The text appears to classify OC primarily according to water solubility. Water solubility is one useful operational classification, but it is not the only relevant way to describe atmospheric organic aerosol. Organic carbon may also be classified by origin (primary or secondary), source, volatility, oxidation state, polarity, functional-group composition, molecular size, optical properties, hygroscopicity, and cloud-forming properties. Please revise this passage so that water solubility is presented as one analytical classification rather than as the principal or exclusive division of OC.

L31: The citation and numerical range for the contribution of WSOC should be checked carefully. The stated range of 20–70% does not appear to represent a direct finding of Du et al. (2014), but rather values cited from earlier studies. The authors should identify the original sources, verify that the numerical range is reproduced correctly, and clarify whether it refers to WSOC's contribution to OC, organic aerosol mass, or another quantity. The comparison with the present study should also be made only if the definitions and analytical methods are comparable. More generally, the Introduction is difficult to follow because it is often unclear whether the authors are referring to organic matter, organic carbon, water-soluble organic carbon, total carbohydrates, or selected carbohydrate biomarkers. These terms should be defined clearly and used consistently.

L35: The wording suggests that dry and wet deposition may not be the only relevant transfer pathways. Please specify which deposition or exchange mechanisms are considered. If the authors refer only to dry and wet deposition, the sentence should be made more precise. If additional processes are intended, such as sea–air exchange, gas uptake, cloud processing, or sedimentation, these should be identified explicitly.

L36–38: The discussion shifts from the Mediterranean Sea to the broader concept of ocean stratification, even though the study focuses on a central Adriatic coastal environment. Please clarify whether the statement refers specifically to stratification in the Mediterranean Sea, the Adriatic Sea, or the global ocean. The consequences of stratification for atmospheric inputs differ substantially among the open ocean, semi-

enclosed seas, coastal waters, and shallow bays. The relevance of this general statement to the Martinska site should therefore be explained more directly.

L39: The statement that carbohydrates are key components of organic matter in the lithosphere is too broad. Carbohydrates are abundant in soils, sediments, pore waters, and biologically active near-surface environments, but they are not generally major components of rocks or of the lithosphere as a whole. Please specify the relevant compartments, such as soils, organic-rich sediments, or biologically influenced surface materials. The description of carbohydrate source markers should also be revised for precision. Anhydrosugars are produced primarily by the thermal degradation of cellulose and hemicellulose and are widely used as tracers of biomass burning. Sugar alcohols, such as mannitol and arabitol, are commonly associated with fungal spores, fungal biomass, and biologically active soil or plant debris, but they are not uniquely source-specific. Primary sugars may originate from pollen, plant fragments, algae, microorganisms, and biologically active soils. These source associations should be presented as indicative rather than exclusive.

L55–57: The explanation of cloud-condensation-nuclei activity is oversimplified. Hygroscopic growth and cloud-droplet activation are governed by Köhler theory and depend on particle size, soluble mass fraction, water activity, surface tension, molecular composition, and mixing state. High carbohydrate solubility alone does not necessarily imply strong CCN activity, particularly because many sugars are less hygroscopic than inorganic salts and some polysaccharides are only partly soluble. The authors should also clarify whether they refer to intact pollen grains and fungal spores, subpollen particles, biological fragments, or fine carbohydrate-containing aerosol particles, since these particle types differ substantially in size, composition, hygroscopicity, and activation behavior.

L61: The spatial scope becomes unclear because the text shifts among oceanic, marine, Mediterranean, coastal, and local environments. These terms are not interchangeable. The authors should define consistently whether the manuscript concerns the global ocean, the Mediterranean Sea, the Adriatic Sea, or the immediate coastal environment around Martinska. The Introduction should remain aligned with the actual scale of the measurements.

Specific comments: Materials and methods

Study area

L100: I would not describe Martinska simply as a rural site. Although the immediate surroundings are largely undeveloped, Šibenik is only approximately 2 km away and lies directly across the bay. Depending on wind direction, the site may be influenced by urban emissions, tourism, harbor and boating activity, domestic combustion, and local traffic. The classification of Martinska as a "rural site" therefore requires clarification. I suggest describing it as a coastal background site or as a rural coastal site with potential nearby urban influence. The authors should also explain whether wind-direction data were used to assess the possible influence from Šibenik.

L110–115: This passage presents regional wildfire statistics and year-to-year comparisons rather than methodological information about the study area. In its current form, it does not belong in the Materials and Methods section. The authors should also clarify the spatial relevance of county-wide statistics to the sampling site, because fire occurrence within the administrative region does not by itself demonstrate an atmospheric influence at Martinska.

Sample collection and processing

L123–125: The expression "samples of atmospheric aerosols (PM_{10})" is terminologically redundant and imprecise. PM_{10} represents an operationally defined airborne particle size fraction, whereas aerosol refers to the suspension of particles in air. Please revise this to "PM_{10} samples" or "PM_{10} aerosol particles."

L138–141: The description of the filtration procedure is incomplete. Please specify the membrane material, filter diameter, hydrophilic or hydrophobic character, and pre-washing procedure, including the volume and solvent used. Membrane composition is important because adsorption and recovery of dissolved

organic carbon and carbohydrates may differ substantially among PTFE, regenerated cellulose, PES, nylon, PVDF, and other materials. In addition, filtration through a 0.45 μm membrane provides an operational definition of the dissolved fraction; it does not strictly "remove the insoluble fraction," because colloids smaller than the nominal pore size may pass through and dissolved compounds may be retained by adsorption. Please revise the terminology and report any blank or recovery tests performed for the filtration step.

L149–156: The terminology, operational definitions, and analytical comparability of WSOC and DOC require substantial clarification. In the PM_{10} samples, WSOC is defined as the water-extracted fraction of a 2.54 cm^2 portion of the quartz filter, obtained by extracting with 21 mL of water, followed by filtration through a 0.7 μm GF/F filter. In the wet and total deposition samples, DOC is defined as the organic carbon that passes through a 0.45 μm syringe filter. These measurements therefore differ in matrix, sample-preparation procedure, extraction step, filter material, and operational size cut-off. Simply renaming DOC as WSOC does not make the two quantities analytically equivalent. Please retain matrix-specific terminology, for example, $PM_{10,}$ WSOC, and deposition DOC, and explain the extent to which they can be compared. The authors should also justify the use of 21 mL of extraction water for a 2.54 cm^2 filter portion and report the corresponding solvent-to-filter-area ratio. Because extraction volume can affect both extract concentration and extraction recovery, especially for partially soluble or strongly particle-associated organic material, please state whether the procedure was validated by spike recovery, repeated extraction, or another recovery test. The potential influence of different filter materials and nominal cutoffs should also be discussed. Finally, please provide the calculation used to convert the extract concentration to atmospheric WSOC concentration, including the extracted filter area, total exposed filter area, extraction volume, sampled air volume, and field-blank correction.

Dissolved carbohydrates

L157–166: The description and validation of the dissolved-carbohydrate method are insufficient. Please define MBTH at first use as 3-methyl-2-benzothiazolinone hydrazone and specify all modifications made relative to the cited procedures, because the expression "slightly modified" is not reproducible. The concentrations of all reagents, reaction volumes, calibration range, number of calibration points, linearity, blank correction, replicate precision, and sample-storage conditions should also be reported. The authors report only a method detection limit (LOD) of 30 μg C L^{-1}, but do not explain how it was calculated, how many blank or low-level replicate measurements were used, or whether this value applies equally to DTCHO, DMCHO, and calculated DPCHO. The corresponding limit of quantification (LOQ) should also be provided, together with the calculation procedure. Because DPCHO is obtained by subtracting DMCHO from DTCHO, its uncertainty is larger than that of either measurement alone; uncertainty propagation and the treatment of negative or near-zero differences should therefore be described.

Most importantly, no recovery data are reported. The authors should demonstrate method recovery using dextran as a minimum validation standard, including the complete hydrolysis and color-development procedure, and preferably also evaluate additional representative mono- and polysaccharides. Recovery should be assessed in both reagent water and matrix-spiked wet- and total-deposition samples to identify possible matrix effects. Strong-acid hydrolysis at 100 °C may produce incomplete hydrolysis for some polysaccharides and degradation of released monosaccharides for others, so hydrolysis efficiency must be experimentally verified rather than assumed. Calibration using dextran alone may also not fully represent chemically diverse atmospheric carbohydrates, and this limitation should be discussed. Without recovery, precision, LOQ, uncertainty, and matrix-validation data, the accuracy and comparability of the reported DTCHO, DMCHO, and DPCHO concentrations cannot be adequately assessed.

Carbohydrate biomarkers

L168–196: The section provides several essential analytical details, including sample preparation, instrument model, column, NaOH eluent, electrode configuration, recoveries, and compound-specific LODs. However, important method-performance parameters remain missing. Please report the calibration range, number of calibration levels, calibration model, and linearity, limits of quantification, replicate

precision or repeatability, and overall analytical uncertainty. The reported recovery range of 87–106% should be presented in greater detail, including the compounds and matrices tested, mean recoveries, variability (relative standard deviations), and the number of replicates. The operational definition of the analyzed fraction also requires clarification. For HPAEC-PAD, the PM_{10} extracts and reconstituted deposition samples were centrifuged rather than filtered. Centrifugation at 3000 rpm does not provide a defined particle-size cut-off and cannot be assumed to yield the same dissolved fraction as the 0.45 μm or 0.7 μm filtration procedures used for DOC, WSOC, and dissolved-carbohydrate analyses. Please report the relative centrifugal force, rotor radius, or centrifuge model, and clarify whether the HPAEC-PAD results represent dissolved carbohydrates or water-extractable biomarkers in the centrifuged supernatant. The implications of these different sample-preparation procedures for comparisons among WSOC, DOC, DTCHO, and individual biomarkers should be discussed.

Meteorological conditions

L200–205: This subsection combines methodological information with campaign results. The data source, measured meteorological variables, temporal resolution, and data-processing procedure belong in the Materials and Methods section. In contrast, the observed ranges and seasonal averages of temperature, relative humidity, precipitation, wind speed, and wind direction are results and should be moved to the Results section. The authors should also define how “spring” and “summer” were assigned. It is currently unclear whether they used meteorological seasons, astronomical seasons, calendar months, or campaign-specific periods. The exact date ranges for each season should be specified, as the calculated seasonal means depend on this definition. Please also report the distance and representativeness of the Šibenik meteorological station relative to the Martinska sampling site, together with the original temporal resolution and the averaging procedure.

Air masses source region classification

L207–217 and Figure S3: The time basis used for the trajectory calculations is not specified. Please state whether the PM_{10} sampling start and end times were recorded in local time, CET/CEST, or UTC, and confirm that all HYSPLIT arrival times were entered in UTC, consistent with the GDAS meteorological data. Because the campaign spans periods affected by daylight saving time, the exact conversion from local time to UTC should be specified. The arrival times at the beginning, midpoint, and end of each 48 h sampling interval should also be reported, along with their time zone, and the authors should clarify how these trajectory times were synchronized with the sampling intervals, meteorological observations, pollen data, and identified Saharan dust or biomass-burning events. Figure S3 shows only one continental and one marine example, but the exact sampling dates, seasons, arrival times, and criteria used to select these cases are not provided. Please clarify whether these trajectories are intended only to illustrate the classification procedure or whether they are considered representative of the spring and summer transport regimes. Two selected examples cannot by themselves demonstrate seasonal representativeness. The authors should therefore identify the periods shown, explain why they were selected as “typical,” and provide the classification of all PM_{10} sampling intervals, including the number and proportion of continental, marine, and mixed or undetermined cases separately for each season. A supplementary table, trajectory-frequency, or cluster analysis would allow evaluation of the robustness of the classification and the representativeness of Figure S3.

Identification of specific atmospheric events

Section 2.6, L225–239: The procedure used to identify Saharan dust and biomass-burning events is not sufficiently described or reproducible. For the Saharan dust classification, the authors list HYSPLIT, MODIS, NASA Earth Observatory, ESA, CAMS, and EUMETSAT sources, but do not specify the exact products examined, variables used, spatial and temporal resolution, access dates, geographical domain, or whether the data were accessed through an API, downloaded as numerical fields, or evaluated by manual inspection of online imagery and reports. Please clarify which dataset served as the primary event-identification tool, which datasets were used only for confirmation, and the objective criteria required to classify a period as a Saharan dust event. The authors should also explain how the observations were

temporally matched to the 48 h PM_{10} sampling intervals and how Saharan dust was distinguished from local or regional mineral-dust resuspension. Similarly, for biomass burning, please define the criteria used to classify an event as "intense and/or prolonged" and explain how official fire records, MODIS observations, event location, duration, burned area, and air-mass transport were combined to determine whether a particular fire could influence the sampling site. The use of a fire record within Šibenik-Knin County alone does not demonstrate atmospheric influence at Martinska. The final sentences of this subsection report the identified event periods, the occurrence of several wildfires, and the affected vegetation types. These are the results of the event-identification procedure, rather than methodological information, and should be moved to the Results section or presented in a separate event-classification table. The Methods section should retain only the datasets, processing workflow, thresholds, and classification rules.

Figure S5 can visually support a dust classification, but a satellite image alone does not prove that dust was present at the surface level at Martinska. Vertical dust forecasts, surface PM_{10} response, lidar or aerosol optical data, and trajectory altitude would strengthen that attribution.

Calculation of deposition fluxes

Section 2.7, L245–265: Please clarify the units and parameter definitions used in the deposition-flux calculations. For wet and total deposition, state explicitly whether V represents the measured rainwater volume or the Milli-Q rinse/reconstitution volume, specify the collector area used for each sample type, and explain how the sampling duration was assigned for event-based wet deposition and approximately weekly total deposition. For dry deposition, V_d is defined in m s^{-1}, whereas the adopted values are subsequently reported in cm s^{-1}. Please use consistent units or provide the corresponding conversions. Moreover, $C_d V_d$ yields a flux in μg m^{-2} s^{-1}; the conversion to the reported daily units should be explicitly included. The assignment of 2 cm s^{-1} to primary sugars and sugar alcohols and 0.1 cm s^{-1} to anhydrosugars is based on assumed coarse- and fine-particle associations rather than size-resolved measurements in the present study. Please clearly identify these as literature-based assumptions, specify the deposition velocity assigned to every reported compound or group, and provide a sensitivity or uncertainty analysis. The weighted WSOC velocity of 0.67 cm s^{-1} should likewise be presented as dependent on the assumed 70:30 fine-to-coarse partitioning.

Data analysis

Section 2.8, L267–277: The statistical workflow requires substantially clearer justification and validation. The authors state that, because some datasets deviated from normality, all variables were log-transformed. Please identify which variables failed the Shapiro–Wilk test, report the corresponding sample sizes, and demonstrate that the transformation improved both distributional symmetry and variance homogeneity. Normality and homoscedasticity should be reassessed after transformation rather than assumed. Applying one transformation (log transformation) to all variables should also be justified, particularly for variables containing zeros or a substantial proportion of values below the LOD.

Welch's t-test is suitable for comparing two independent group means when variances and sample sizes may differ, but the manuscript should explain why it was selected, specify the exact contrasts, report group sample sizes, state whether tests were two-sided, and confirm that independence and post-transformation assumptions were evaluated. Because multiple seasonal and event-related comparisons appear to have been performed, the authors should also address multiple-testing control. For comparisons involving more than two event categories or simultaneous effects of season and event type, a multifactor model or a robust or permutation-based approach may be more appropriate than repeated pairwise t-tests.

The use of Pearson correlation also requires justification. Please verify linearity, influential outliers, independence, and the adequacy of the transformed bivariate distributions. Where relationships remain non-normal, are nonlinear but monotonic, or are strongly affected by outliers, Spearman correlation should be considered for sensitivity analysis. Correlation coefficients, confidence intervals, exact p-values, and sample sizes should be reported.

Replacing all values below the LOD with LOD/2 may bias means, variances, correlations, and PCA results, particularly when censoring is frequent. Please report the fraction of <LOD observations for every compound and evaluate the sensitivity of the results to the selected substitution method.

Finally, PCA does not require univariate normality when used descriptively. The manuscript should instead report whether variables were centered and standardized, whether the covariance or correlation matrix was used, the number of samples and variables in each PCA, the treatment of missing and censored values, the component-retention criterion, and evidence that the solutions are stable. Separate PCA analyses for small event subsets may be particularly unstable and should be supported by sample-size information and, ideally, bootstrap or sensitivity analysis (suggestions).

Results and discussion

Atmospheric carbohydrate composition and seasonal variability

L280–286 and Table 1: The composition of the carbohydrate groups differs among matrices. For example, "sugar alcohols" comprises four compounds in PM_{10}, three in wet deposition, and five in total deposition, while "Total MB" is consequently also based on different analyte sets. This appears to result from matrix-specific exclusion of compounds for which more than 65% of measurements were below the LOD. Although matrix-specific exclusion may be justified, variables with different chemical compositions should not be presented under the same label and directly compared without qualification. Please explain the basis for the 65% threshold, provide detection frequencies for every compound and matrix, and clarify how below-LOD values were handled within each group sum. For direct cross-matrix comparisons, the authors should either use a common set of compounds or clearly identify the groupings as matrix-specific and avoid interpreting differences as if the variables were analytically equivalent.

Table 1 and Sect. 2.8: The manuscript states only that values are reported as "mean ± 1 standard deviation," but it does not specify what the standard deviation represents. Based on the reported sample numbers, it appears to describe variability among independent environmental samples rather than analytical replicate precision; this should be stated explicitly in the Methods and table captions. Analytical repeatability or measurement uncertainty should be reported separately. The relationship between the log-transformed statistical analysis and the descriptive statistics in Table 1 also requires clarification. Please state whether the tabulated means and standard deviations were calculated from the original untransformed concentrations or from log-transformed data. If calculated on the original scale, they are arithmetic means and between-sample standard deviations (SDs), whereas the statistical tests were apparently performed on log-transformed values (as described in the method). If log-scale means were back-transformed, the results should be identified as geometric means and accompanied by geometric SDs or back-transformed confidence intervals rather than symmetric arithmetic SDs. Given the strong skewness and small sample sizes, medians, interquartile ranges, and full ranges should also be reported.

L291–298: Table 1 shows no statistically significant seasonal difference in PM_{10} mass concentration between spring (14.6 ± 9.8 μg m^{-3}) and summer (15.8 ± 9.6 μg m^{-3}). The authors correctly describe the seasonal variability as insignificant, but subsequently refer to a "trend of increased PM_{10} mass during summer" and compare this trend with a previous study. A small nonsignificant difference should not be interpreted as evidence of a seasonal increase or as confirmation of a previously reported seasonal pattern. Please revise the wording to state that PM_{10} concentrations were comparable between seasons, with only a small numerical difference. If the comparison with the previous study is retained, provide the corresponding value and clarify whether the sampling periods, methods, site conditions, and event occurrence were sufficiently comparable. The statement that concentrations were lower than in the previous study should also be presented as a descriptive comparison unless statistical comparability can be demonstrated.

L298–300 and Table 1: The reported mean contribution of WSOC to PM_{10} mass of 9.4% cannot be reproduced directly from the campaign-average concentrations in Table 1, which yield approximately 8.8% when the ratio of the reported means is calculated. Please specify whether 9.4% represents the mean of

sample-specific WSOC/PM_{10} ratios, the ratio of the overall means, or another calculation, and provide the corresponding equation and variability or confidence interval.

L300–310 and Sect. 3.2: The discussion attributes the higher summer WSOC mainly to changes in dominant seasonal sources and, later, to stronger local and regional source influence under low-wind conditions. However, the manuscript does not adequately consider secondary atmospheric formation and oxidative aging. Enhanced summer photochemistry can oxidize primary and gaseous organic precursors, producing more oxygenated and often more water-soluble secondary organic compounds. At the same time, a deeper and more convective summer boundary layer may enhance dilution. The observed seasonal WSOC difference, therefore, cannot be interpreted solely in terms of stronger source emissions or local accumulation. Please discuss the combined effects of primary emissions, secondary formation, atmospheric aging, transport, and boundary-layer dynamics, and moderate source attribution unless these processes can be separated using additional tracers or measurements.

L324–334 and Table 1: The synthesis of seasonal patterns is difficult to follow and is not fully consistent with the statistical results, which makes this part confusing. According to Table 1, statistically significant seasonal differences occur only in PM_{10}, specifically for WSOC, primary sugars, sugar alcohols, and Total MB, whereas anhydrosugars do not differ significantly. No significant spring–summer differences are indicated for wet or total deposition. Therefore, statements that wet deposition "amplified" seasonal differences, that individual biomarker groups showed strong seasonal variation across matrices, and that spring precipitation exerted a strong influence on total deposition chemistry are not statistically supported and should be moderated. The statement that total biomarker amounts lacked a significant seasonal trend is also inconsistent with the significant PM_{10} Total MB difference and should be qualified by the matrix. Please distinguish systematically between statistically significant differences and numerical tendencies. A multi-panel figure displaying individual spring and summer observations for each matrix, together with sample sizes and significance levels, would make the contrasting and partly nonsignificant patterns substantially clearer.

Moreover, the Methods section defines a single significance level of $p < 0.05$, whereas the tables (1, 2, 3, and 4) distinguish between $*p < 0.05$ and $**p < 0.01$. Please clarify that $p < 0.01$ is an additional reporting category indicating a smaller p-value, rather than a separately selected decision threshold. Exact p-values should be reported wherever possible instead of only asterisk categories, together with effect sizes and confidence intervals. More importantly, the manuscript performs numerous comparisons across matrices, seasons, compounds, and event types, but no procedure for controlling multiple testing is described. Please state the total number of tests performed and apply or justify an appropriate multiplicity correction. Comparisons based on very small groups, including $N = 3$, should be interpreted cautiously and not presented as robust solely because $p < 0.05$ or $p < 0.01$.

The wet-deposition subsets are extremely small, in some cases containing only two, three, or five samples. These observations may document precipitation events that occurred during the campaign, but they cannot be assumed to reliably represent seasonal wet-deposition behavior. With such small and highly variable event-based datasets, means, standard deviations, and Welch-test p-values are strongly influenced by individual events and have low statistical power. Comparisons based on $N = 2$ should not be treated inferentially, and results based on $N = 3$ or $N = 5$ should be interpreted as exploratory. Please report all individual event values, precipitation amount and duration, volume-weighted concentrations, and event-specific fluxes, and substantially moderate claims of seasonal differences or representativeness.

Figure 3, Figure S3, and Section. 2.5: Figure S3 presents only one illustrative continental and one illustrative marine trajectory case, whereas Figure 3 statistically compares WSOC concentrations for samples classified as CONT and MAR over the full campaign and by season. The link between these figures is not sufficiently documented. Please provide the sampling dates and number of observations included in every Figure 3 boxplot, together with a complete table assigning each 48 h PM_{10} sample to continental, marine, mixed, or undetermined air masses. The objective classification criteria and treatment of disagreement among the beginning, midpoint, and end trajectories should also be reported. Figure S3 should be described explicitly as illustrative only and should not be presented as evidence that the two

selected cases are representative of the full seasonal dataset. In addition, the term “contribution” in the Figure 3 caption is too strong: the figure shows WSOC concentrations associated with classified air-mass sectors, not quantitative source contributions.

Figure 3 and L335–340: The interpretation of higher WSOC under continental trajectories as evidence that continental air masses are a stronger source of atmospheric organic matter than marine air masses is too strong. Figure 3 shows receptor concentrations associated with trajectory sectors, not emission strengths or quantitative source contributions. Continental trajectories may include terrestrial biological, urban, harbor, traffic, combustion, and regional sources, while lower WSOC under marine flow may partly reflect stronger ventilation and dilution. Boundary-layer height should also be considered, because differences in vertical mixing can substantially affect near-surface concentrations independently of source strength. Incorporating boundary-layer height, together with wind speed and other meteorological parameters, would help distinguish source effects from dilution and accumulation. The conclusions should therefore be reframed as sector-associated concentration differences rather than direct evidence that continental sources emit more organic matter than the Adriatic Sea.

L348–350: The discussion of PM_{10} dispersion should also consider boundary-layer height. A shallow boundary layer promotes near-surface accumulation, whereas a deeper boundary layer enhances dilution. This may partly explain concentration differences attributed to marine and continental air masses.

L350–357: The interpretation repeatedly invokes dilution, accumulation, and atmospheric stability, yet boundary-layer height is not considered. Because near-surface WSOC concentrations depend strongly on the depth of the mixed layer, high concentrations at low wind speeds do not, by themselves, prove dominant local emissions; boundary-layer-height data should be included, or its absence clearly acknowledged as a limitation.

Figure 4: The bivariate polar plots show mean WSOC concentrations as a function of wind direction and wind speed, but they do not directly represent atmospheric stability, boundary-layer height, or source distance. Therefore, interpretations based on stable conditions, local sources, and distant transport should be moderated or supported by additional meteorological information.

Figure 5 and L362–368: Figure 5 is insufficiently discussed. Please describe the monthly wind sectors and concentration patterns systematically and report the number of samples contributing to each panel, because monthly polar plots based on 48 h samples may contain few observations. High Total MB concentrations at low wind speeds indicate reduced ventilation but do not, by themselves, demonstrate stable conditions or dominant local sources. Boundary-layer effects and other meteorological controls should be considered. In addition, Total MB combines chemically distinct biomarker groups; the authors should identify whether the March and August maxima were driven by primary sugars, sugar alcohols, or anhydrosugars and relate these patterns to pollen, biomass-burning events, trajectories, and seasonal biological activity. The discussion should also address the other months rather than focusing only on March and August.

L371–390, Figure 6 and PCA methodology (Methods section): The purpose, construction, and interpretation of the PCA are insufficiently described and require major revision. Figure 6 presents separate vectors for spring and summer versions of each variable, such as WSOC_SPR and WSOC_SUM, but the structure of the input matrix is not explained. Please specify what constituted the rows and columns of the PCA dataset, the number of observations, and whether a combined PCA or separate seasonal PCAs were performed. If individual PM_{10} samples were the observations, season should normally be used as a grouping variable for the sample scores rather than converting each analyte–season combination into a separate variable. Figure 6 is a loading or correlation-circle plot, not a biplot, because no sample scores are shown. Consequently, the statement that PC1 “clearly separated spring and summer samples” is not demonstrated. Please provide a score plot or a true biplot with individual samples identified by season, together with the numerical loading matrix, eigenvalues, explained variance, communalities or variable contributions, and a scree plot. The Methods should also state the software and function used, whether data were centered and scaled, whether the covariance or correlation matrix was analyzed, how missing and below-LOD values were handled, and how the retained components were selected. The inclusion of Total MB alongside the primary-sugar, sugar-alcohol, and anhydrosugar groups introduces mathematical redundancy, as Total MB

is derived from the same variables. This can artificially strengthen correlations and should be avoided or explicitly justified. More fundamentally, PCA reveals the covariance structure but does not, by itself, identify or quantify atmospheric sources. Clustering of variables should therefore not be interpreted as proof of common or dominant sources without independent evidence of sources. Because PC1 and PC2 explain only 54.4% of the total variance, the robustness of the two-dimensional interpretation should also be moderated. The authors should clarify what additional scientific insight the PCA provides beyond the direct seasonal comparisons.

Figure 6, as currently presented, is not sufficient to support either seasonal sample separation or source attribution. The most serious issue is the apparent treatment of spring and summer analyte values as separate variables rather than showing individual sample scores grouped by season.

L391–397 and Table 2: The purpose and design of the event-based comparison need clearer explanation. Table 2 appears to be intended to test whether concentrations during pollen, Saharan dust, and biomass-burning periods differed from non-event conditions for PM_{10}, wet deposition, and total deposition. However, the text refers broadly to "source influence," whereas the analysis demonstrates only statistical associations with predefined event periods and does not establish causality or quantify source contributions. Please state the specific expected response for each event type and parameter. The definition of "background" also requires clarification, including how overlapping pollen, dust, biomass-burning, and rainfall periods were handled and whether samples could belong to more than one category. Because sample sizes differ strongly among matrices and some wet-deposition groups contain only one or two observations, these comparisons should be treated as exploratory. The term "deposition events" should also be replaced by "atmospheric events" or "event periods."

Lines 410–413: The statement "beyond the increase in PM_{10} mass" is unclear and appears inconsistent with Table 1, which shows comparable seasonal PM_{10} concentrations in spring and summer. Table 1 presents a seasonal comparison, whereas Table 2 apparently compares event-classified pollen samples with background samples; this distinction should be stated explicitly. Please report the PM_{10} values and statistical results supporting the claimed increase during pollen episodes. In addition, clarify whether pollen-event samples were compared with spring-specific background samples or with a pooled March–September background dataset. Because high-pollen episodes occurred predominantly in spring, comparison with a pooled seasonal background may confound the pollen effect with seasonal and meteorological variability. The conclusion should therefore be limited to higher WSOC and carbohydrate biomarker concentrations during pollen-classified samples unless an independent pollen-related increase in PM_{10} mass is demonstrated.

Lines 415–431 and Table 3: The statistical presentation and source attribution require substantial clarification. The Methods define statistical significance at $p < 0.05$, whereas Table 3 marks only $p < 0.01$. Although $p < 0.01$ is compatible with an alpha level of 0.05, the manuscript should consistently define and report both significance levels, provide exact p-values where possible, and clarify whether the tests were two-sided and whether multiple-testing correction was applied. **The Table 3 caption should refer to statistically significant correlations, not regressions**. The correlations are based on only 14 pollen-event PM_{10} samples. Please report how daily pollen counts were temporally aggregated and matched to the 48 h integrated PM_{10} samples. The strong glucose–Total MB correlation should not be interpreted as independent-source evidence, because glucose is itself included in Total MB, producing a part–whole correlation. In addition, the text refers to a correlation between pollen and "glucose or primary sugars," although Table 3 reports only glucose; the actual correlation for summed primary sugars should be provided, or this statement should be removed. The statement that sugars are "released into atmospheric aerosols" is terminologically unclear. It would be more accurate to state that sugars are emitted into the atmosphere either in association with primary biological particles or as part of atmospheric particulate matter. Furthermore, this study analyzed bulk PM_{10} and did not resolve the fine and coarse fractions; therefore, literature-based statements regarding the size distribution of glucose should be presented as contextual information rather than as direct evidence from this dataset. Limited agricultural activity near the site also does not preclude contributions from local or transported soil and dust.

Most importantly, correlations do not “confirm” pollen as the dominant carbohydrate source. They support an association between airborne pollen and glucose during the selected pollen episodes. Still, they do not quantify source contributions or rule out covariance with season, meteorology, plant debris, soil resuspension, dust, or overlapping events. The conclusion should therefore be softened to state that pollen or pollen-associated biological material was a likely important source during these periods. Finally, please resolve the apparent inconsistency between N = 14 in Table 3 and N = 94 later in the text for the same reported correlation, r = 0.912.

Table 3: The term “statistically significant regressions” is incorrect. Table 3 presents a Pearson correlation matrix containing pairwise r-values, not regression results. Please replace “regressions” with “correlations.” If regression analysis was actually performed, the Methods and Results must specify the dependent and independent variables and report the regression coefficients, intercepts, R^2, residual diagnostics, and exact p-values.

There is also a related interpretative issue. Although the authors correctly describe the analysis as a Pearson correlation analysis, they subsequently use causal wording, stating that pollen increased carbohydrate concentrations and confirming pollen as the dominant source. The reported correlations demonstrate statistical covariation between pollen and glucose or Total MB, **but they do not establish causality,** rule out common seasonal or meteorological drivers, or quantify pollen's contribution relative to other potential sources.

Lines 436–455: The exclusion of marine contributions to atmospheric glucose is not sufficiently supported. The cited literature demonstrates that marine carbohydrates, including glucose, can be transferred from seawater to sea-spray aerosol**. In particular, Zeppenfeld et al. reported glucose as an important component of seawater carbohydrate pools, while Rocchi et al. identified glucose as a relevant organic constituent affecting sea-spray aerosol production**. **These studies therefore establish the plausibility of marine glucose rather than providing evidence for its exclusion**. The absence of a significant Pearson correlation between glucose and wind speed does not demonstrate that marine emissions were unimportant. Sea-spray production depends nonlinearly on wind speed, wave breaking, fetch, wind direction, sea state, and sea-surface biological composition. Moreover, the 48 h PM_{10} integration period may obscure short-lived sea-spray episodes. Excluding a marine contribution would require more direct evidence, such as correlations with sea-salt tracers, comparison between marine and continental air-mass categories, onshore wind-sector analysis, or size-resolved chemical measurements. The predominance of particles larger than 1 µm reported by Clemente et al. is also not a unique indicator of terrestrial origin because sea-spray aerosol contributes strongly to the supermicron fraction. Since the present study analyzed only bulk PM_{10}, the external size-distribution result should not be treated as direct evidence for the source of glucose at this site. Likewise, the weak correlation between mannitol and glucose (r=0.373) does not establish a shared terrestrial source, since both species may covary with season, meteorology, or transport, even though they arise from distinct biological sources. The conclusion should be softened to state that the results support an important terrestrial biological contribution without excluding a potentially marine contribution. Finally, please correct the apparent inconsistency in the pollen–glucose correlation, reported with N = 14 in Table 3 but N = 94 at line 455.

Pollen measurements and attribution: Please clarify that airborne pollen concentrations were obtained from an independent pollen-monitoring network and were not measured directly in the PM_{10} filter samples. The Methods should provide the pollen sampler type, counting and identification procedures, monitoring site coordinates, sampling height, distance from the Martinska PM_{10} site, data completeness, and the method used to match daily pollen counts to the 48 h PM_{10} samples. This distinction is particularly important because most intact pollen grains are larger than 10 µm and would not be efficiently sampled by a PM_{10} inlet. The observed relationship may therefore reflect pollen fragments, subpollen particles, other co-emitted plant material, or shared seasonal conditions rather than direct collection of intact pollen grains in PM_{10}. The conclusions should be framed as an association with high regional pollen periods unless pollen-derived material was independently identified on the PM_{10} filters.

Lines 457–477: The section demonstrates mainly that Saharan dust episodes increased PM_{10} mass, while most measured carbohydrate variables and WSOC were not statistically different from background. This is a limited but valid result. However, several subsequent interpretations extend beyond the evidence. The phrase "moderately higher" should not be used as evidence of an event effect where differences are not statistically significant, and sample numbers are very small, particularly for wet deposition. Please report exact sample numbers, effect sizes, confidence intervals, and analytical uncertainties, and distinguish clearly between the absence of a statistically detectable difference and evidence of no source contribution.

The statement that the results support efficient wet scavenging of carbohydrates is not demonstrated by two wet-deposition samples and the detection of biomarkers in only one sample. Likewise, the presence of DTCHO without detectable target monosaccharides may reflect untargeted oligo- or polysaccharides, but it may also result from differences in analytical scope, sensitivity, hydrolysis response, or matrix effects. These alternatives should be discussed.

The conclusions that dust acted primarily as a carrier phase, that natural background sources dominated the observed organic matter, and that dust did not contribute substantial organic loads are largely based on the literature rather than on measurements from this study. The authors measured PM_{10} mass, WSOC and selected carbohydrate variables, but did not measure total OC, total organic matter, insoluble organic carbon, mineral tracers, or sea-salt tracers. Accordingly, conclusions should be restricted to the measured quantities.

Terminology should also be standardized. "Organic load," "organic matter," "organic carbon," "WSOC," "Total MB" and "carbohydrate-derived organic loads" are not equivalent. Please define each term and use it only for the corresponding measured quantity. **In particular, the manuscript should not state that Saharan dust made no significant contribution to "atmospheric organic matter" when total organic matter was not measured.**

Lines 479–519: The biomass-burning discussion contains a direct statistical contradiction and several overextended source interpretations. The opening statement that "all measured parameters … increased significantly" conflicts with the following sentence, which states that the differences were not statistically significant. Please distinguish numerical increases from statistically significant differences and report absolute concentrations, sample numbers, variability, confidence intervals, and exact p-values. The very large percentage increases in wet-deposition WSOC, Total MB, and DTCHO are difficult to interpret due to the small sample size and potentially low background values. The increase in PM_{10} anhydrosugars during biomass-burning periods is consistent with enhanced combustion influence and provides the clearest source-specific result. However, anhydrosugars were below detection in wet and total deposition, whereas WSOC, Total MB and DTCHO are not unique biomass-burning tracers. Consequently, the attribution of the increase in deposition to biomass burning remains indirect and should be presented cautiously. The negative spring correlation between anhydrosugars and temperature is consistent with increased residential wood-combustion activity during colder periods, but it does not "confirm" residential heating as the dominant source. Temperature covaries with atmospheric stability, boundary-layer development, transport, and season. Similarly, the coincidence of high summer anhydrosugar concentrations with documented wildfires supports wildfire influence, but source attribution requires clarification of fire location, distance, timing, wind direction, and plume transport to the sampling site. The statement that strong winds were associated with air stagnation is physically inconsistent and should be revised. Strong winds generally enhance ventilation, whereas stagnation is associated with weak winds and limited dispersion. If different meteorological phases occurred during the fire episode, these should be described separately. Finally, the conclusion that summer wildfires "governed atmospheric carbohydrate levels" is too broad. The evidence specifically supports elevated PM_{10} anhydrosugars during wildfire periods, not control of all atmospheric carbohydrate classes. Likewise, "continuous impact of biomass burning" is not demonstrated by the presented event-based analysis. The discussion should be shortened and restricted to analyte-specific findings and to the actual added value beyond the already established use of anhydrosugars as biomass-burning tracers.

The attribution of elevated spring anhydrosugars to residential heating requires further support. The reported mean spring temperature was 19.0 ± 4.2°C, which does not intuitively indicate strong local heating demand. Although intermittent heating may still occur during colder March–April days or nights, the authors should identify the specific high-anhydrosugar samples, report the corresponding daily minimum or nighttime temperatures, and provide evidence regarding the local heating season and fuel-use practices. Alternatively, elevated anhydrosugars may reflect regional or long-range transport from cooler continental areas where residential biomass combustion remained active. The negative correlation with temperature alone cannot distinguish increased local emissions from changes in air-mass origin, boundary-layer depth, or atmospheric dilution. The source attribution should therefore be elaborated and softened unless additional evidence is provided.

Section 3.2.4, Other biogenic sources: The conclusion that fungal activity was the dominant source of summer sugar alcohols is insufficiently demonstrated. The present evidence consists mainly of higher summer PM_{10} sugar-alcohol concentrations, a bivariate polar-plot pattern interpreted as local, and a positive relationship with temperature. These observations are consistent with fungal spore emissions, since mannitol and arabitol are widely used as fungal biomarkers, but they are not source-specific and do not independently confirm the presence of fungi. No fungal spore counts, microscopy, DNA, ergosterol, β-glucans, or other microbiological indicators were measured. Please report the actual temperature-correlation coefficient, exact p-value, and sample size, and replace "fungal spore activity" with more precise terminology such as "fungal-spore emission or aerosolization." The statement that summer enhancement excludes vegetation as the dominant source is also not justified. Sugar alcohols can originate from fungal spores, plants, leaf litter, soil biological material, and mixed vegetation-associated sources, all of which may increase or be more readily aerosolized under warm and dry summer conditions. These source categories are not clearly separable using seasonality alone. The marine environment is notably absent from this discussion despite the coastal setting. A local source, inferred from a polar plot, could originate either from nearby land or from the Adriatic Sea. Marine microorganisms, phytoplankton, macroalgae, and sea-surface microlayer organic matter may contribute carbohydrates and polyols to sea-spray aerosol. The authors should examine whether high sugar-alcohol concentrations occurred under onshore winds or marine air masses and, where available, compare them with sea-salt tracers such as Na^+, Cl^-, and Mg^{2+}. Without such evidence, a marine contribution cannot be excluded. Future work could strengthen source identification through paired measurements of PM_{10}, bulk seawater, and especially the sea-surface microlayer, together with wave state, onshore fetch, marine biological indicators, and sea-salt tracers. Tidal stage may also be explored where shoreline exposure provides a plausible emission mechanism, although the 48 h PM_{10} integration period will average over several tidal cycles. The conclusion should therefore be softened to identify fungal emissions as a plausible contributor rather than a demonstrated dominant source.

Section 3.3 and Table 4: This section is very difficult to follow because a large number of values, percentage changes, and significance statements are presented without a graphical overview. Table 4 combines calculated dry-deposition fluxes, event-based wet-deposition fluxes, and open-collector total-deposition fluxes, although these quantities were obtained by different methods and over different integration periods. Please clarify that "total deposition" represents direct collection by the Bergerhoff sampler and is not the numerical sum of the separately estimated dry and wet fluxes. The temporal correspondence among the 48 h PM_{10} filters, individual rain events, and approximately weekly total-deposition samples should also be explained. The calculated dry-deposition fluxes depend directly on fixed literature deposition velocities and assumed particle-size associations. Since the manuscript acknowledges uncertainties approaching 100%, these uncertainties should be propagated into the reported dry fluxes and their implications discussed. Event-to-background differences smaller than this uncertainty may not be physically distinguishable. The wet-deposition comparisons are based on extremely small sample numbers, in some cases approximately N = 3 per group. Large numerical or percentage increases should therefore not be interpreted as evidence of efficient scavenging when the differences are not statistically significant. Wet flux is also strongly influenced by precipitation amount and duration; demonstrating scavenging efficiency would require paired aerosol–rain measurements, washout ratios, or precipitation-normalized

analyses. Table 4 should be separated into carbon-based quantities, expressed as μg C m^{-2} d^{-1}, and biomarker compound masses, expressed as μg compound m^{-2} d^{-1}. These are not directly comparable and should not be discussed collectively as "organic matter." I strongly recommend adding a figure showing event-to-background flux ratios with uncertainty intervals and sample numbers, separately for dry, wet, and total deposition. This would make the principal result visible: pollen predominantly enhanced primary-sugar deposition; biomass-burning periods produced the largest, though often statistically uncertain, increases in WSOC and dissolved-carbohydrate fluxes; and Saharan dust had little detectable effect on the measured carbohydrate fluxes.

Section 3.4, Biogeochemical significance: This section substantially overinterprets a simple theoretical mixing calculation as evidence of biogeochemical significance. The study did not measure DOC, dissolved carbohydrates, microbial activity, or any biological response in the receiving Adriatic seawater. The reported changes, therefore, represent hypothetical concentration increments derived from atmospheric fluxes, an assumed instantaneous mixing depth of 1 m, and a literature-derived ambient DOC concentration, rather than observed changes in the marine environment. This distinction should be stated explicitly throughout the section and in Table 5. The physical basis for the fixed 1 m mixing depth is not provided, although the resulting percentage change scales inversely with this assumption. Coastal mixing depth varies with stratification, wind, waves, convection, freshwater input, and circulation. A sensitivity analysis across plausible mixing depths is required, and the current estimates should be presented as scenario-dependent values, with the assumed 1 m depth yielding substantially larger concentration increments than in deeper-mixing scenarios. The term "bioavailable carbon" is not justified by the analytical measurements. WSOC denotes an operationally defined water-soluble carbon fraction and is not necessarily biologically available. Similarly, DTCHO identifies dissolved carbohydrate equivalents but does not establish chemical lability, microbial uptake, or degradation rate. No incubation, respiration, bacterial production, enzyme activity, or carbon-uptake measurements were performed. The terminology should therefore be revised to "potentially bioavailable" or, preferably, to the directly measured quantities: atmospheric WSOC and DTCHO input. The marine system is treated primarily as a passive receptor, although the Adriatic Sea contains endogenous, dynamically varying DOC and carbohydrate pools generated by phytoplankton, bacteria, macroalgae, extracellular polymer production, and coastal inputs. No concurrent seawater carbohydrate measurements were made, and the comparison is against total DOC from a separate study rather than against the local dissolved carbohydrate pool during the events investigated. The atmospheric contribution should therefore be evaluated relative to the existing marine DOC and, where possible, dissolved-carbohydrate pools. A possible marine origin and recycling of some atmospheric carbohydrates should also be considered. Sea-spray aerosol and sea-surface microlayer material can transfer marine carbohydrates into the atmosphere, which may subsequently undergo redeposition. In the absence of paired seawater and aerosol measurements, marine aerosol tracers, or a robust analysis of marine-sector air masses, the calculated deposition cannot be assumed to represent entirely new terrestrial carbon entering the Adriatic Sea. Finally, the source-specific interpretation compounds the uncertainties associated with event classification, biomarker specificity, trajectory analysis, and PCA-based interpretation discussed throughout the manuscript. Consequently, many of the source attributions appear more definitive than the available evidence supports. Pollen-derived material was not identified directly on PM_{10} filters, fungal emissions were not independently measured, dust-related carbohydrate increases were mostly non-significant, and biomass-burning wet-deposition comparisons were based on very small sample numbers. These uncertain assignments should not be carried into the biogeochemical calculation as established sources. The section should therefore be reframed as an exploratory estimate of potential atmospheric carbon input, rather than as a demonstration of marine biogeochemical impact.