



1 **Synoptic-scale organization of multi-day black carbon variability in a regional-**
2 **background environment over eastern China**

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Abstract

Black carbon (BC) variability reflects the combined effects of emissions, regional transport, atmospheric mixing, and removal processes, yet the temporal scales through which these processes organize receptor concentrations remain poorly constrained. Regional-background environments provide a useful perspective for identifying atmospheric controls because they are less dominated by immediate local emission fluctuations. Here, year-long hourly BC observations from a regional-background site in eastern China were combined with time-frequency analysis, backward trajectories, circulation-state transitions, and an independent 3–10 day time-domain evaluation to investigate the drivers of multi-day BC variability. BC exhibited enhanced variability at multi-day periods near one week, but the signal occurred intermittently rather than as a persistent weekly oscillation. Transport classification identified contrasting atmospheric states: Regional/slow flow was associated with higher BC ($1.30 \mu\text{g m}^{-3}$), weaker winds (1.68 m s^{-1}), and shallower planetary boundary-layer height (431 m) than northeastern/coastal (NE/coastal) flow ($0.90 \mu\text{g m}^{-3}$, 2.22 m s^{-1} , and 593 m, respectively). Transitions toward Regional/slow conditions were followed by BC increases, whereas transitions toward NE/coastal conditions produced decreases. The independently extracted 3–10 day BC component covaried with the persistence of these transport states (Spearman $\rho = 0.345$; segment-aware circular-shift $p < 0.001$). Together, these results show that the apparent near-weekly BC feature represents an emergent signature of recurring synoptic accumulation–ventilation states rather than a fixed weekly emission cycle. This study highlights the importance of incorporating atmospheric-state information when interpreting multi-day aerosol variability.

Keywords: Black carbon; Synoptic circulation; Atmospheric transport; Time-frequency analysis; Boundary-layer dynamics



1. Introduction

Black carbon (BC) is a combustion-derived, strongly light-absorbing aerosol that influences climate, atmospheric stability, visibility, and human health (Bond et al., 2013; Janssen et al., 2011; Ramanathan and Carmichael, 2008). Its atmospheric abundance is governed not only by emission strength but also by rapid changes in transport, dilution, vertical redistribution, aging, and wet removal (Bond et al., 2013; Ding et al., 2016; Ding et al., 2023; Liu et al., 2020a). Consequently, a high BC concentration at a receptor does not uniquely diagnose high local emissions. The same concentration enhancement may arise when a polluted air mass remains over a source-rich region, when horizontal ventilation weakens, when the planetary boundary layer (PBL) becomes shallow, or when removal is suppressed. Conversely, a change to cleaner or more rapidly ventilated flow can lower BC even if local emissions change little. This sensitivity makes BC an informative tracer of atmospheric processing, but it also means that concentration trends and short pollution episodes cannot be interpreted from emissions alone. The ability to disentangle these influences depends strongly on receptor environment. Sites dominated by nearby sources may integrate rapid emission fluctuations, whereas regional-background environments provide an opportunity to examine how transport, mixing, and removal processes organize aerosol variability over broader spatial scales.

Eastern China provides a particularly useful setting in which to examine this problem because regional-background observations in this highly heterogeneous emission environment can capture the integrated influence of regional transport and atmospheric-state variability. The Yangtze River Delta (YRD) contains dense urban, industrial, and transport emissions and is influenced by monsoonal circulation, frontal passages, coastal flow, continental outflow, and substantial seasonal changes in boundary-layer structure (Zhou et al., 2024; Zhuang et al., 2014). Previous studies have demonstrated that regional transport can substantially alter BC within the YRD atmospheric boundary layer and that similar surface stability can coexist with very different transport conditions aloft (Cheng et al., 2022). Three-dimensional observations and modeling further show that transported BC can reach the upper boundary layer of downwind regions before affecting the surface, emphasizing that surface concentration is the endpoint of a vertically structured transport process and is not a direct proxy for local emissions (Lu et al., 2023). More broadly, transboundary transport and aerosol–boundary-layer interactions can reinforce persistent haze over northern and eastern China (Huang et al., 2020). These findings establish the importance of regional transport, but they leave open a different question: over what temporal scale does the evolving transport–ventilation system organize BC variability at a receptor?

This temporal question matters because atmospheric processes are intrinsically multiscale. Boundary-layer growth and collapse can impose diurnal variability, precipitation can remove aerosol over hours,



frontal passages and stagnation episodes can persist for several days, and seasonal circulation changes alter the background state over months (Miao et al., 2020; Moteki et al., 2012; Zhang et al., 2010). A conventional correlation between BC and wind speed or PBL height collapses these scales into a single number. Similarly, a trajectory cluster analysis can identify air-mass types but does not by itself reveal whether changes among those types correspond to a characteristic timescale in the concentration record. Time-frequency analysis offers a route to bridge these perspectives by identifying when particular periods contribute strongly to variability while allowing for non-stationarity throughout the record (Grinsted et al., 2004; Torrence and Compo, 1998).

However, detecting a spectral feature is not equivalent to identifying its physical origin. A broad maximum near seven days, for example, could be misinterpreted as a weekly emission cycle simply because seven days is a familiar calendar period. In the atmosphere, repeated but irregular synoptic transitions can generate power over a similar range without any deterministic weekly forcing. A physically defensible interpretation therefore requires evidence that is independent of the spectral calculation: the feature should be temporally intermittent in a manner consistent with weather variability, should covary with ventilation at comparable scales, should differ among physically distinct transport states, and ideally should be recoverable directly from the concentration record using a method that does not depend on wavelet power.

Here, we investigate the processes controlling multi-day BC variability at a regional-background site in Hangzhou, eastern China. Unlike urban receptors that are often strongly influenced by nearby emission fluctuations, regional-background observations provide an opportunity to examine how regional transport, atmospheric mixing, and removal processes shape aerosol variability at the receptor scale. By integrating time-frequency analysis with transport analysis and circulation-state transitions, we identify the atmospheric factors governing multi-day BC fluctuations and evaluate whether the apparent near-weekly feature reflects periodic emissions or recurring synoptic accumulation–ventilation states. This study provides new insights into the interpretation of BC variability in regional-background environments and highlights the importance of considering atmospheric processes when assessing aerosol pollution and its controls.

2. Materials and Methods

2.1 Site, BC observations, and meteorological data

Hourly BC concentrations were measured during 2017 at a regional-background site in Hangzhou, eastern China (30.22°N, 119.70°E), using a seven-wavelength AE33 Aethalometer (Magee Scientific) operated at a sampling flow rate of 5 L min⁻¹. The AE33 determines aerosol light attenuation from particles collected on a filter tape and converts the attenuation signal to BC mass concentration. A dual-



spot algorithm is used to compensate for the filter-loading effect in real time (Drinovec et al., 2015). In this study, the instrument-reported BC concentration at 880 nm was used because absorption at this wavelength is predominantly associated with black carbon and is less affected by other light-absorbing aerosol components than at shorter wavelengths (Drinovec et al., 2015; Rajesh and Ramachandran, 2018). Quality control followed the monitoring workflow used for the original dataset: hourly measurements deviating by more than three standard deviations from a 24 h moving average were flagged as potential outliers for inspection. Periods without valid instrument output, including maintenance and instrument downtime, were retained as missing and were not replaced for concentration statistics. The annual record contained 8,322 valid hourly BC observations. Co-located wind speed, precipitation, relative humidity, and temperature were available at hourly resolution from the same site. Planetary boundary-layer height (PBLH) was obtained at approximately 3 h resolution from the Copernicus Atmosphere Monitoring Service (CAMS) reanalysis for the grid cell containing the site (Inness et al., 2019). Analysis-specific gap treatment and temporal aggregation are described in the corresponding subsections below. Seasons were defined as winter (December–February), spring (March–May), summer (June–August), and autumn (September–November).

2.2 Segment-aware wavelet analysis and red-noise significance

Continuous wavelet analysis followed the framework of Torrence and Compo (1998) using a complex Morlet wavelet ($\omega_0 = 6$). To retain the hourly grid without allowing long gaps to create artificial continuity, isolated gaps ≤ 24 h were linearly interpolated for the continuous wavelet transform (CWT), whereas gaps > 24 h were retained as true breaks. The 24 h threshold was selected to avoid filling gaps comparable to or longer than the target synoptic timescale while preserving the temporal resolution required for CWT analysis. Each resulting continuous segment was transformed independently, and segments were not concatenated before transformation. Each segment was assigned its own cone of influence (COI), and interpretation was restricted to coefficients inside the COI.

Wavelet power was defined as $|W(a,b)|^2$, and Fourier-equivalent periods up to 256 h were retained to encompass the target multi-day synoptic variability while avoiding interpretation of longer periods poorly constrained by the record length. Significance was evaluated against a first-order autoregressive [AR(1)] red-noise background. Lag-1 autocorrelation was estimated from valid adjacent observations without treating points separated by long gaps as adjacent. Local significance was evaluated at the 95% level. The global spectrum pooled valid coefficients across segments and inside their COIs; its significance threshold was estimated using 2,000 AR(1) Monte Carlo surrogate realizations processed with the same segment structure, COI exclusion, and sampling pattern as the observations.

Seasonal occurrence was calculated as the fraction of valid time-frequency points inside the COI



145 exceeding the 95% threshold within predefined period bands.

146 **2.3 High-BC episode composite and wavelet coherence**

147 High-BC episodes were defined as intervals in which hourly BC exceeded the annual 95th percentile
148 ($2.61 \mu\text{g m}^{-3}$) for at least 6 consecutive hours. The 6 h duration criterion was applied to exclude short-
149 lived transient spikes and focus on sustained pollution episodes. Events separated by <24 h were
150 merged, and boundary events lacking a complete -72 to $+48$ h window were excluded, leaving six
151 independent episode clusters (Table 1). Wavelet power near diurnal and multi-day periods
152 (approximately 24 and 168 h) was normalized to the mean during -72 to -48 h and composited relative
153 to episode onset. This composite was used only to describe how short- and multi-day variability coexist
154 around high-BC events.

155 Wavelet coherence between BC and wind speed and between BC and PBLH followed Grinsted et al.
156 (2004). Variables were placed on a common 3 h grid to match the temporal resolution of the PBLH
157 product; BC and wind speed were aggregated to 3 h means, and PBLH was interpolated to this grid.
158 For coherence analysis, a stricter gap criterion was used: only gaps ≤ 6 h were interpolated, whereas
159 longer gaps defined separate segments. A Morlet wavelet ($\omega_0 = 6$), 1/12-octave scale resolution, periods
160 up to 256 h, 95% significance testing, and segment-specific COIs were applied. Phase arrows indicate
161 relative phase relationships and are interpreted as evidence of temporal coupling rather than causality.

**Table 1. High-concentration episodes retained for composite analysis**

Cluster	Start time	End time	Duration (h)	Mean BC ($\mu\text{g m}^{-3}$)	Season
1	2017-04-06 15:00	2017-04-06 22:00	7	3.40	Spring
2	2017-09-25 13:00	2017-09-26 05:00	16	4.48	Autumn
3	2017-11-08 17:00	2017-11-08 23:00	6	3.40	Autumn
4	2017-11-27 08:00	2017-11-28 06:00	22	3.24	Autumn
5	2017-12-22 15:00	2017-12-24 00:00	33	3.68	Winter
6	2017-12-26 03:00	2017-12-28 05:00	50	3.38	Winter

2.4 HYSPLIT backward trajectories and transport regimes

48-h backward trajectories were calculated hourly with the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 1998) using GDAS1 meteorological fields. Trajectories arrived at 50 m above ground level to represent near-surface air-mass transport affecting the receptor. Trajectories with at least 45 endpoint hours and coverage to -48 h were retained. This criterion removed incomplete trajectories while maintaining sufficient spatial information for transport-regime classification. Endpoints were interpolated to common 3 h ages and converted to horizontal displacement vectors relative to the receptor. K-means clustering solutions from $k = 2-8$ were evaluated using silhouette and Davies–Bouldin indices together with physical interpretability, including differences in pathway orientation, transport distance, and regional meteorological characteristics. Three regimes were retained and labeled Regional/slow, northeastern/coastal (NE/coastal), and long-range continental transport based on path length, direction, and mean -48 h position (Fig. 7a).

2.5 Regime transitions and independent 3–10 day analysis

Changes between the two dominant transport regimes were identified in the hourly classification. Because Regional/slow and NE/coastal regimes represented the dominant contrasting circulation states associated with BC enhancement and reduction, respectively, transitions between these two regimes were further analyzed. Transitions separated by <24 h were declustered. BC anomalies were



composited from -24 to $+24$ h relative to each transition after subtracting the mean BC during the preceding 24 h to emphasize concentration changes relative to the pre-transition state instead of absolute differences among regimes. Windows with $<80\%$ valid BC were excluded.

Differences between transition directions were evaluated with the Mann–Whitney U test. To independently evaluate the multi-day variability identified from wavelet analysis, a fourth-order Butterworth band-pass filter retained BC variability between 72 and 240 h (3–10 days). Filtering was performed separately within long continuous segments after interpolation only of gaps ≤ 6 h, and 240 h were removed from both ends of each filtered segment to minimize edge artifacts. A 72 h transport contrast was calculated as the rolling fraction of Regional/slow trajectories minus the rolling fraction of NE/coastal trajectories, representing the evolving balance between accumulation-favorable and ventilation-favorable transport states. Sensitivity tests repeated this calculation using averaging windows of 24, 48, 96, 120, and 168 h to evaluate whether the inferred BC–transport relationship depended on the 72 h window selected for the main analysis.

Daily band-passed BC and transport contrast were compared using Spearman rank correlation. Statistical significance was assessed with 5,000 segment-aware circular shifts, in which each continuous segment was shifted independently by at least 14 days, thereby preserving within-segment autocorrelation while breaking the observed temporal alignment between BC and transport. Transport-contrast terciles defined NE/coastal-favored, intermediate, and Regional/slow-favored states; differences among states were evaluated using the Kruskal–Wallis test.

3. Results and Discussion

3.1 Characteristics and Temporal Scales of BC Variability

The annual mean BC concentration was $1.18 \pm 0.78 \mu\text{g m}^{-3}$, with pronounced temporal variability and clear seasonality (Figure 1). Winter showed the highest mean concentration ($1.58 \mu\text{g m}^{-3}$), approximately twice the summer mean ($0.77 \mu\text{g m}^{-3}$), while spring and autumn means were 1.18 and $1.14 \mu\text{g m}^{-3}$, respectively. The seasonal contrast is consistent with the combined influence of wintertime emissions, weaker atmospheric mixing, reduced wet removal, and more frequent continental or stagnant circulation, whereas the warm season is generally characterized by a deeper boundary layer and stronger precipitation processing. Similar seasonal enhancement of BC has been reported elsewhere in China (Cao et al., 2026; Ding et al., 2023; Ding et al., 2024; Feng et al., 2014), but the relative contributions of emissions and atmospheric processing vary substantially among regions. Here, the focus is on how atmospheric state conditions receptor concentrations; source-sector apportionment is outside the present scope.



This conditioning is already preliminarily visible in the bulk meteorological relationships. Mean BC decreased from $1.57 \mu\text{g m}^{-3}$ when PBLH was $<50 \text{ m}$ to $0.89 \mu\text{g m}^{-3}$ when PBLH exceeded 1000 m , and BC was negatively correlated with wind speed ($r = -0.30$). A shallow PBL compresses emissions and transported aerosol into a smaller mixing volume, while weak wind reduces horizontal dilution and can increase residence time over a source-rich region (Ambade et al., 2021; Tiwari et al., 2013). These processes are physically complementary and operate through distinct mechanisms. A shallow PBL can elevate BC even under modest transport, whereas weak regional flow can maintain a polluted air mass over the receptor for longer than the lifetime of an individual local emission pulse. Vertical observations over Beijing demonstrate that PBL development redistributes BC between the surface and the upper boundary layer and that moist summertime conditions can remove substantial BC during vertical transport (Liu et al., 2020a). Thus, PBLH is not merely a statistical covariate but it influences the vertical volume available for dilution and modifies the pathway through which BC is redistributed and removed.

The global wavelet spectrum provides a different view of the same variability (Figure 2). It did not show a dominant and isolated diurnal peak; instead, BC power increased broadly toward multi-day periods and exhibited a pronounced maximum in the vicinity of one week. The breadth of this feature is scientifically important. A persistent 7-day emission cycle would be expected to produce a relatively narrow and persistent spectral signature. By contrast, atmospheric circulation systems have variable lifetimes, propagation speeds, and transition times. A sequence of stagnation, regional recirculation, frontal passage, and coastal ventilation events can therefore project power across several neighboring periods. We consequently interpret the 3–10 day range as a characteristic synoptic timescale, not as evidence for a deterministic seven-day oscillator.

The local wavelet spectrum reinforces this distinction (Figure 3). Significant multi-day power was confined to discrete intervals and did not form a continuous ridge through 2017. Strong structures appeared during parts of spring, early autumn, and late autumn/winter, while other periods showed little significant multi-day power. The internal data interruption was retained as a true break with segment-specific COIs not being bridged by long interpolation. This treatment matters because a long interpolation could create smooth low-frequency variability and artificially strengthen multi-day power. The persistence of significant structures within the observed continuous segments therefore supports an atmospheric origin of the multi-day feature and argues against a gap-filling artifact.

The seasonal occurrence analysis (Figure 4) further shows that the multi-day mode is conditional on season. Autumn contained the most frequent significant power in the 96–168 h range, while winter



exhibited significant variability over a broader range of scales, and summer contributed comparatively little. Such seasonal intermittency is difficult to reconcile with a fixed weekly emission schedule acting as the primary driver. Although human activity may contain weekly variability, such calendar effects would need to persist consistently across seasons to explain the observed pattern. Synoptic circulation, in contrast, is expected to be seasonally selective because the frequency of fronts, monsoonal flow, stagnation, precipitation, and boundary-layer states changes with season. A YRD-wide analysis of synoptic circulation likewise found that distinct circulation types produce distinct transport pathways, diffusion conditions, and PM_{2.5} levels, demonstrating that weather regimes strongly organize regional air pollution alongside emission strength (Liao et al., 2017).

The high-BC episode composite (Figure 5) adds another layer to this interpretation. Six independent high-BC clusters remained after merging nearby exceedances and excluding incomplete boundary events. The 24 h component tended to strengthen close to episode onset, whereas multi-day power evolved more gradually over the wider window. Given the small number of episodes and the substantial event-to-event variability, the composite is best viewed as evidence of nested variability not as a predictive sequence: rapid changes associated with boundary-layer cycling or short-lived emission/meteorological perturbations occur within a slower synoptic envelope. This nesting explains why an individual high-BC event can develop over hours even though the atmospheric state favoring accumulation persists for several days.

Taken together, Figures 1–5 establish two points that are easy to conflate but should be separated. First, BC has a characteristic multi-day timescale. Second, that timescale is non-stationary and seasonally selective. The second observation is what gives the first physical meaning. A persistent calendar cycle would be primarily a property of the emission schedule; an intermittent multi-day feature is more naturally a property of an evolving atmosphere. The remaining question is whether independent dynamical information actually identifies the atmospheric states responsible for this feature.

3.2 Synoptic Transport and Ventilation Effects on BC Variability

Wavelet coherence between BC and wind speed and between BC and PBLH provides the first independent dynamical evidence (Figure 6). Significant coherence was not uniformly distributed across the year or across periods. Instead, coherent structures appeared episodically from diurnal to multi-day scales, often during intervals in which the BC wavelet spectrum also contained enhanced power. Many significant regions showed a leftward phase component, broadly consistent with anti-phase behavior: BC enhancements were generally associated with periods of reduced ventilation, whereas stronger wind or higher PBLH tended to coincide with lower BC variability at comparable timescales. The precise phase relationship varies because concentration responds to transport history, mixing, and



removal rather than instantaneously to a single local meteorological variable. For this reason, coherence is used here primarily as evidence of scale-specific coupling, not as a literal time-lag estimator.

This result also resolves an apparent contradiction between the weak global 24 h BC peak and the obvious importance of boundary-layer cycling. The PBL itself has a strong diurnal cycle, but BC will only inherit a persistent 24 h signal when emission, entrainment, residual-layer exchange, and advection align consistently with that cycle. On days dominated by frontal transport or strong regional advection, the local diurnal mixing signal can be overwhelmed. Conversely, during stagnant periods, daily PBL collapse can strongly modulate surface BC. The wavelet framework therefore shows why a process can be physically important without producing a dominant global spectral peak: its influence is conditional in time.

The trajectory analysis makes the synoptic interpretation more explicit (Figure 7). Of 8,729 trajectories retained after quality control, 62.0% belonged to a regional/slow regime, 28.0% to an NE/coastal regime, and 10.0% to a long-range continental regime. Mean path lengths were approximately 543, 1005, and 1703 km, respectively. BC differed markedly among the three regimes (Kruskal–Wallis $p < 0.001$): mean concentrations were $1.304 \mu\text{g m}^{-3}$ under regional/slow flow, $0.902 \mu\text{g m}^{-3}$ under NE/coastal flow, and $1.278 \mu\text{g m}^{-3}$ under long-range continental flow. The contrast was also evident across multiple seasons, indicating that the annual difference was not solely caused by seasonal frequency differences among regimes. Thus, the annual difference is not simply caused by one regime occurring more often in winter and another in summer.

The most informative feature is that the shortest transport regime produced the highest mean BC. If transport distance alone controlled receptor loading, the long-range continental pathway might be expected to produce the largest concentration because it crosses a greater geographic domain. The observations show otherwise. Regional/slow flow had the weakest wind speed (1.68 m s^{-1}) and shallowest PBLH (431 m), whereas NE/coastal flow had the strongest wind (2.22 m s^{-1}) and deepest PBLH (593 m); long-range continental conditions were intermediate (1.94 m s^{-1} and 486 m). The regional/slow regime therefore combines two accumulation mechanisms: reduced horizontal ventilation and reduced vertical dilution. Because the YRD is itself a dense emission region, a slowly moving air mass does not need to travel far to accumulate BC. What matters is not simply how far the air mass has traveled, but how long it remains coupled to emissions and how effectively the atmosphere disperses or removes the accumulated aerosol.

This interpretation is consistent with previous work in eastern China. Cheng et al. (2022) showed that local accumulation and regional transport can both control BC in the YRD atmospheric boundary layer and, importantly, that similar stable surface conditions can conceal very different upper-air transport



structures. Lu et al. (2023) found that regional transport is a key control on the three-dimensional BC distribution over eastern China and that transported BC can arrive in the upper boundary layer of a downwind region before its surface manifestation. These studies caution against treating surface wind or trajectory origin as independent one-dimensional predictors. Our regime analysis complements this perspective by showing that the high-BC state is a coupled transport–ventilation configuration: slow regional residence and shallow mixing occur together.

The comparatively low BC under NE/coastal flow is likewise unlikely to reflect air-mass origin alone. Stronger winds and a deeper PBL increase dilution, while maritime or coastal pathways can reduce contact with intense continental sources. Precipitation and moist processing may further decrease BC during transport. Direct observations in South China show that trajectory class can strongly modulate BC wet scavenging (Ding et al., 2023), and vertical measurements over Beijing demonstrate substantial summertime BC loss during transport from the surface toward the top of the PBL under moist conditions (Liu et al., 2020a). Along the eastern Chinese coast, Guo et al. (2017) observed systematic changes in BC–CO relationships from coastal to marine environments, consistent with aging and removal during transport. These studies support a broader interpretation of the coastal regime as a ventilation and processing state beyond a simple directional classification.

The long-range continental regime illustrates the opposite complexity. Its mean BC ($1.278 \mu\text{g m}^{-3}$) was close to that of regional/slow flow despite a much longer mean path. Long-range transport can import pollution from distant source regions, but it simultaneously allows more time for dilution, deposition, vertical redistribution, and chemical aging. BC aging during transport is strongly influenced by the emissions encountered along the pathway, and polluted regions can modify the mixing state of BC originating elsewhere (Zhang et al., 2019). Consequently, long-range transport can deliver substantial BC without producing a simple monotonic relationship between path length and surface concentration. This is why the three regimes should be interpreted as integrated atmospheric states, not as a ranking from 'near' to 'far'.

Static regime means describe concentration contrasts but not the temporal ordering of concentration changes. The transition analysis therefore examines how BC evolves when the circulation state changes (Figure 7d). After declustering, 58 transitions from the NE/coastal to the Regional/slow regime and 66 transitions in the opposite direction were retained. Transitions from NE/coastal to Regional/slow conditions were followed by a mean 24 h BC increase of $0.179 \mu\text{g m}^{-3}$, whereas transitions from Regional/slow to NE/coastal conditions were followed by a mean decrease of $0.196 \mu\text{g m}^{-3}$. The BC responses differed significantly between the two transition directions (Mann–Whitney U test, $p < 0.001$). The near-mirror response directly links circulation evolution to subsequent concentration



evolution across matched transition events.

These transition composites characterize coordinated circulation changes; they do not isolate a single causal intervention. A circulation transition simultaneously modifies wind speed, PBLH, precipitation probability, air-mass history, and vertical transport, so the trajectory label is best understood as a compact descriptor of an evolving meteorological state. The analysis therefore tests whether reorganization between atmospheric states is followed by systematically different BC behavior.

The transition result is also consistent with the broader concept of regional haze organization by synoptic circulation. Huang et al. (2020) showed that transboundary transport can interact with aerosol–boundary-layer feedbacks to amplify long-lasting haze between northern and eastern China. Their study addressed a larger spatial scale and included aerosol feedbacks, while the common feature is that persistent pollution emerges from a coupled regional circulation–boundary-layer system. Our results show that this coupling also leaves a measurable multi-day imprint on BC at a regional-background receptor.

The spectral and trajectory analyses together suggest that recurring transport states contribute to the observed multi-day BC variability. To quantify this correspondence independently of wavelet power, we isolated the 3–10 day component directly from the BC concentration series and compared it with a continuous 72 h transport contrast derived from the relative occurrence of Regional/slow and NE/coastal trajectories (Figure 8). This analysis tests the specific expectation arising from the accumulation–ventilation interpretation that increasing persistence of the Regional/slow state relative to the NE/coastal state should be accompanied by a positive multi-day BC anomaly.

3.3 Transport-State Organization of Multi-Day BC Variability

Figure 8 provides a complementary time-domain test of whether the multi-day BC variability identified by the spectral analysis is systematically associated with transport-state persistence. To ensure that the derived 3–10 day BC component was not affected by data discontinuities, the band-pass filter was applied separately within each long continuous portion of the observational record. Only short gaps (≤ 6 h) were interpolated, and 10 days were excluded from both ends of each filtered segment to minimize edge effects, leaving 279 matched daily observations. The extended mid-year data gap was retained as a discontinuity, preventing information from being propagated across unobserved periods. This segment-wise treatment is important for multi-day filtering because interpolation across long gaps or filtering across segment boundaries could introduce artificial low-frequency structure. The retained 3–10 day component therefore represents variability resolved within continuous observational periods without reconstructing across missing intervals.

The daily 3–10 day BC component was positively associated with the 72 h transport contrast between



Regional/slow and NE/coastal states (Spearman $\rho = 0.345$; Figure 8b). Because both BC and transport-state metrics exhibit temporal autocorrelation, significance was evaluated using segment-aware circular shifts that preserved the temporal structure within each continuous data segment while disrupting the observed alignment between the two variables. The association remained significant under this autocorrelation-preserving test ($p < 0.001$) and was consistently positive in the two long continuous segments ($\rho = 0.382$ and 0.287 , respectively). Sensitivity tests further showed that the relationship was not specific to the 72 h averaging window: correlations remained positive and significant for windows from 24 to 120 h but weakened progressively with increasing window length and became nonsignificant at 168 h (Figure S1). These results show that the observed BC–transport coupling is robust to temporal autocorrelation, the discontinuity in the observational record, and the choice of transport-state averaging window. The progressive weakening at longer averaging windows further suggests that multi-day BC variability is more closely associated with the recent persistence of transport conditions than with circulation averaged over an entire week. The magnitude of this modulation was substantial (Figure 8c). Mean 3–10 day BC anomalies were $-0.169 \mu\text{g m}^{-3}$ under NE/coastal-favored conditions, $+0.017 \mu\text{g m}^{-3}$ under intermediate conditions, and $+0.146 \mu\text{g m}^{-3}$ under Regional/slow-favored conditions. The difference between the two extreme states was therefore $0.315 \mu\text{g m}^{-3}$, approximately one quarter of the annual mean BC concentration. This contrast represents the amplitude of multi-day BC modulation associated with the coupled differences among synoptic transport–ventilation states after slower seasonal variability has been removed.

The time-domain analysis provides an important complement to the preceding spectral, meteorological, and trajectory analyses. Wavelet analysis establishes the presence and temporal localization of multi-day BC variability, while coherence identifies periods during which BC covaries with ventilation-related meteorological variables. Trajectory clustering further shows that distinct air-mass histories are associated with systematically different BC concentrations, and the transition analysis demonstrates that reorganization between the two dominant transport regimes is followed by BC changes of opposite sign. Figure 8 connects these observations quantitatively by showing that the independently extracted 3–10 day BC component varies systematically with the relative persistence of Regional/slow and NE/coastal transport states. The interpretation is therefore supported by convergence among analytical approaches that characterize the same phenomenon from different perspectives, not by spectral evidence alone.

This convergence also helps distinguish the observed multi-day variability from a simple calendar-driven weekly cycle. A recurring weekly pattern in anthropogenic activity could produce spectral power near seven days, but such periodicity alone would not account for the systematic association between



BC and transport state. The 3–10 day BC component increased with greater persistence of Regional/slow conditions and decreased under NE/coastal-favored conditions, while transitions between these regimes were followed by BC changes of opposite sign. In addition, the near-weekly wavelet signal occurred intermittently rather than persistently throughout the year. These features identify recurring changes in transport and ventilation as an important contributor to the observed multi-day variability. Weekly variations in emissions may also contribute, but the combined spectral and transport evidence supports interpretation of the near-weekly feature as an emergent synoptic signal rather than a fixed seven-day emission cycle.

The physical interpretation of these results requires viewing the identified transport states as more than trajectory directions. The Regional/slow state combines shorter transport pathways with weaker local winds, shallower PBLH, and elevated BC, whereas the NE/coastal state is characterized by a different transport pathway, stronger winds, deeper mixing, and lower BC. The observed concentration differences are therefore consistent with the combined influence of air-mass history, residence time, horizontal dispersion, vertical dilution, and removal. Such coupling is consistent with the established role of synoptic circulation in organizing particulate pollution over the YRD (Liao et al., 2017). It also illustrates why individual meteorological variables may provide an incomplete description of BC variability: local wind speed and PBLH are themselves partly manifestations of the larger circulation state that simultaneously influences air-mass origin and transport history.

The significance of the 3–10 day band therefore lies not simply in its numerical timescale, but in its association with recurring changes in transport and ventilation. Under persistent Regional/slow conditions, weak ventilation and shallow mixing favor elevated BC and increase the relevance of atmospheric conditions over preceding days. Consequently, an elevated concentration observed at a receptor cannot necessarily be attributed to same-day local emissions alone. Conversely, lower BC during strongly ventilated NE/coastal conditions does not necessarily indicate a corresponding reduction in local emissions. This distinction is particularly relevant when short observational periods are used to evaluate emission interventions: a control period coinciding with unusually favorable ventilation may exaggerate the apparent emission-related decrease, whereas persistent accumulation-favorable conditions may obscure an actual reduction. Meteorological normalization can account statistically for part of this variability, but explicit transport-state information may provide additional context because air-mass history and regional transport are not fully represented by local meteorological variables alone.

The same interpretation provides a process-based rationale for explicitly considering regional conditions when interpreting receptor concentrations. Persistent Regional/slow transport accompanied



by weak ventilation and shallow mixing identifies the conditions under which regional air-mass history becomes especially relevant. The 3–10 day variability characterizes the timescale over which recurring synoptic transport and ventilation states organize BC variability in the present observations; it does not represent a fixed atmospheric residence time.

Several limitations define the scope of this interpretation. First, the descriptive CWT uses interpolation for isolated gaps ≤ 24 h. Although long gaps are segmented and the central multi-day relationship is independently evaluated using the stricter ≤ 6 h criterion, local wavelet power near interpolated intervals should still be interpreted cautiously. Second, HYSPLIT trajectories are model-derived air-parcel histories whose uncertainty generally increases backward in time. The single 50 m arrival height used here emphasizes near-surface transport and cannot represent all elevated transport pathways documented in eastern China (Cheng et al., 2022; Lu et al., 2023). Third, k-means clustering necessarily discretizes a continuous circulation field. The continuous transport contrast used in Figure 8 reduces dependence on individual cluster assignments, but alternative clustering algorithms and synoptic classifications should be examined in longer datasets.

The one-year, single-receptor record constrains climatological generalization. Reproduction of the transport–BC relationship in both continuous observational segments supports its internal robustness, while multi-year observations are needed to evaluate the recurrence and interannual stability of the identified 3–10 day timescale. Changes in emissions, monsoon strength, precipitation, and synoptic frequency could alter both the magnitude and characteristic timescale of the response. The relative prominence of multi-day organization may also depend on receptor environment. At urban sites, stronger and more temporally variable local emissions may enhance sub-daily and diurnal BC variability and partially obscure the synoptic signal resolved at a regional-background receptor. Quantitative partitioning of local and regional emissions, horizontal transport, vertical mixing, and removal would require additional chemical tracers, source-resolved BC measurements, receptor modeling, or tagged chemical-transport simulations.

The analytical framework extends beyond an exact seven-day periodicity or the specific trajectory classes identified at this site. Its broader utility lies in combining time-frequency analysis with meteorological coupling, circulation-state transitions, and an independent time-domain reconstruction of the relevant pollutant variability. Applied to longer records and other transport-sensitive pollutants, this framework could help determine whether an observed spectral feature has a plausible atmospheric basis and identify the transport and ventilation conditions associated with its occurrence.

3.4 Comparison with contrasting urban environments



To examine whether the multi-day BC variability identified at the Hangzhou regional-background site was site-specific, we further analyzed concurrent BC observations from two independent urban stations representing contrasting climatic regions in China: Benxi in Liaoning Province, northeastern China, and Nanning in Guangxi Province, southern China.

The annual mean BC concentration at Benxi was approximately twice those observed at Hangzhou and Nanning, consistent with the stronger influence of wintertime heating-related emissions and generally less favorable wintertime dispersion conditions in northern China (Jiang et al., 2024; Liu et al., 2020b; Ran et al., 2016). Despite differences in regional meteorology, emission structure, and urban characteristics, both additional sites exhibited enhanced wavelet power within the 3–10 day band (Figure S2). However, unlike the Hangzhou regional-background site, the corresponding spectral maxima at Benxi and Nanning did not exceed the 95% confidence level, suggesting that the synoptic-scale signal was less pronounced at urban locations where local emission variability may obscure regional atmospheric signatures.

Nevertheless, enhanced wavelet power was consistently concentrated within the 3–10 day band at both urban sites. When evaluated relative to variability within the 1–10 day range, the 3–10 day component accounted for 81.7% and 79.4% of the integrated power at Benxi and Nanning, respectively, comparable to the 85.3% observed at Hangzhou. These results suggest that multi-day BC variability at synoptic timescales is not restricted to the Hangzhou background environment and can also be detected at contrasting urban sites, although with weaker statistical expression. The stronger and statistically significant expression observed at Hangzhou likely reflects the reduced influence of local emission perturbations at a regional-background site, allowing meteorological influences on BC accumulation and removal to emerge more clearly.

4. Conclusions

This study identifies a characteristic multi-day component of BC variability at a regional-background receptor in eastern China and associates it with recurring synoptic transport and atmospheric ventilation states. The observed near-weekly spectral maximum was intermittent and did not form a persistent seven-day oscillation and was accompanied by scale-dependent coupling between BC, wind speed, and planetary boundary-layer height. Trajectory analysis distinguished contrasting transport states: Regional/slow conditions combined weaker horizontal ventilation and shallower boundary-layer mixing with elevated BC, whereas NE/coastal conditions were associated with stronger ventilation, deeper mixing, and lower concentrations. Transitions between these states were followed by BC changes of opposite sign, linking circulation reorganization to subsequent concentration evolution.



512

513 A complementary time-domain analysis provided independent quantitative support for this
514 interpretation. The independently extracted 3–10 day BC component covaried with the relative
515 persistence of Regional/slow and NE/coastal transport states, and the relationship remained significant
516 after accounting for temporal autocorrelation and was reproduced in both long continuous
517 observational segments. The association was robust to transport-state averaging windows from 24 to
518 120 h but became nonsignificant at 168 h. Together, these results indicate that the apparent near-weekly
519 feature is better interpreted as an emergent signature of recurrent synoptic accumulation and ventilation
520 states than as evidence of a fixed weekly emission cycle. The 3–10 day range should therefore be
521 regarded as a characteristic timescale of atmospheric variability resolved in this record, not as a BC
522 residence time or a universally applicable periodicity.

523

524 The regional-background setting is important to this interpretation. Relative to more emission-
525 influenced urban receptors, a background receptor allows changes associated with air-mass history,
526 regional transport, boundary-layer mixing, and removal processes to be examined with less interference
527 from rapidly varying local sources. The prominence of multi-day variability observed here may
528 therefore depend on receptor environment. Whether similar synoptic organization persists, weakens, or
529 is masked by stronger local forcing at suburban and urban sites remains an important question for multi-
530 site investigation.

531

532 More broadly, this study demonstrates the value of moving from timescale detection to process-based
533 interpretation when analyzing high-frequency atmospheric-composition records. A spectral feature
534 alone cannot identify its physical origin because similar timescales may arise from emissions,
535 meteorology, transport, or their interactions. Combining time-frequency analysis with scale-specific
536 meteorological coupling, trajectory-defined atmospheric states, circulation transitions, and direct time-
537 domain testing provides a framework for evaluating whether an observed temporal feature has a
538 physically consistent atmospheric basis. Application to multi-year records, contrasting receptor
539 environments, and other transport-sensitive aerosol species will help determine how characteristic
540 pollutant timescales vary with synoptic circulation, season, and the balance between local and regional
541 influences.

542



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546

547 **Competing interests**

548 At least one of the (co-)authors is a member of the editorial board of Atmospheric Chemistry and
549 Physics.

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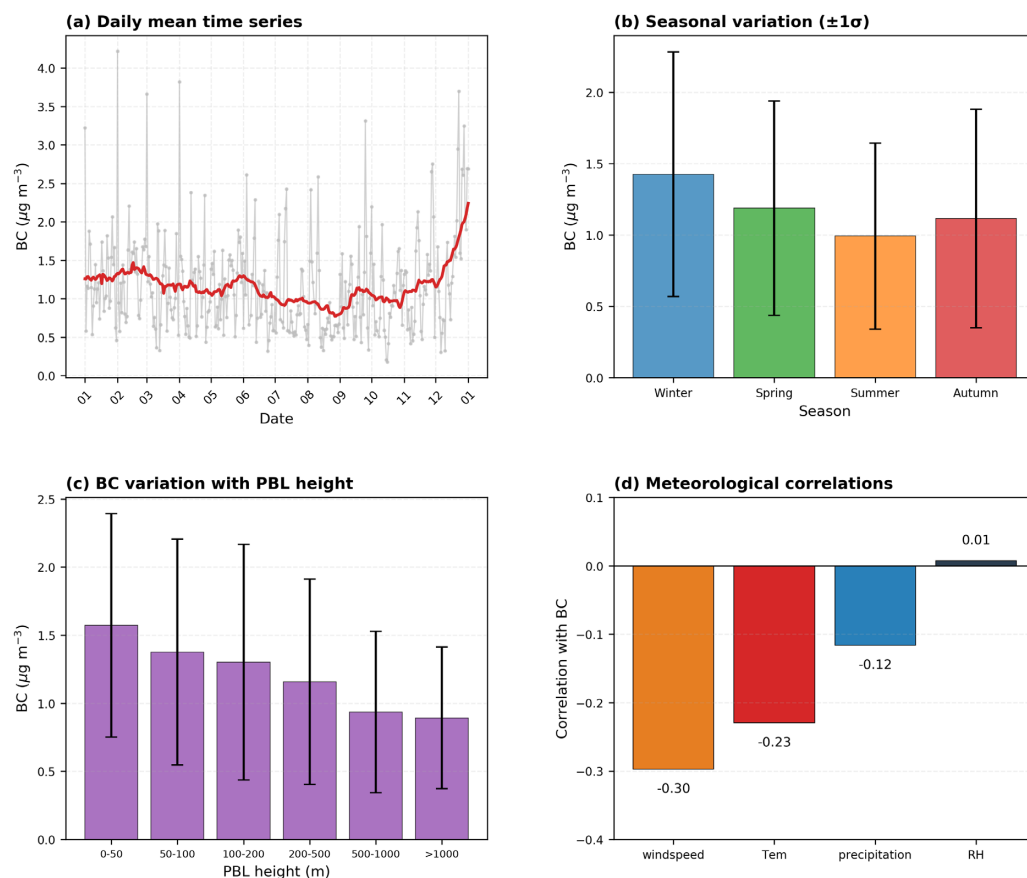
551 **Data availability**

552 The underlying time series used in this study, including hourly BC, co-located hourly surface
553 meteorology, and PBLH, are publicly archived in Mendeley, at DOI:10.17632/2bbrmffbpr.1.

554



555 Figures and captions



556

557 Figure 1. Overview of BC variability and associated atmospheric conditions at the regional-background
 558 site in Hangzhou. (a) Daily mean BC concentrations and 30-day running mean. (b) Seasonal variation
 559 of BC concentrations. (c) BC concentrations across different PBL height ranges. (d) Correlations
 560 between BC and meteorological variables.

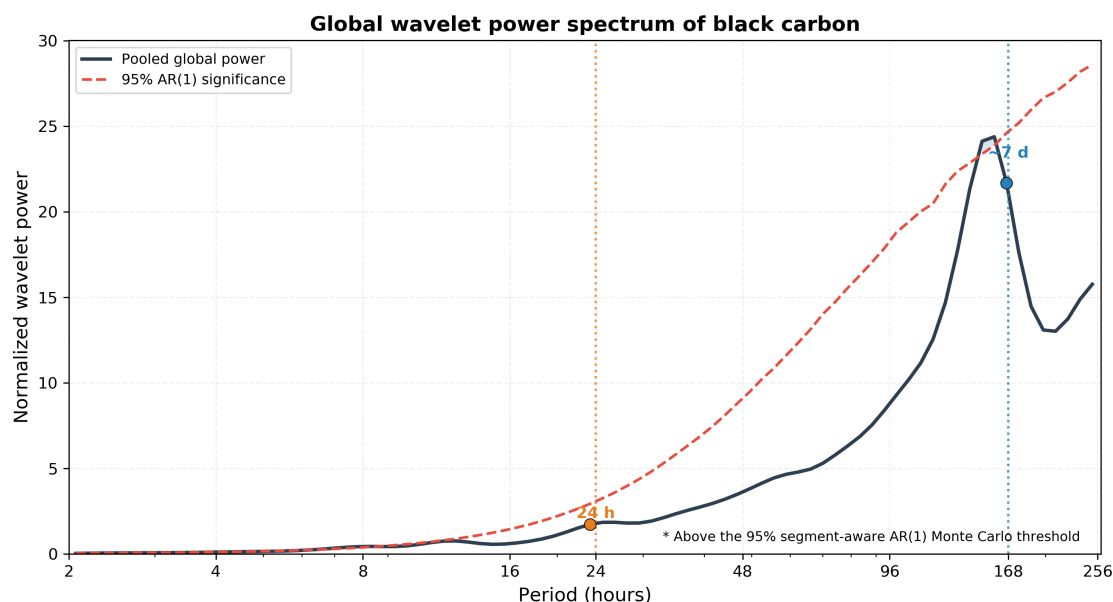


Figure 2. Global wavelet power spectrum of hourly BC concentration. The solid line shows the pooled global wavelet power and the dashed line the 95% AR(1) red-noise significance threshold. A broad multi-day maximum occurs near the one-week timescale, whereas the 24-h component is weak. The multi-day maximum is interpreted as a characteristic timescale and not as a persistent periodic cycle.

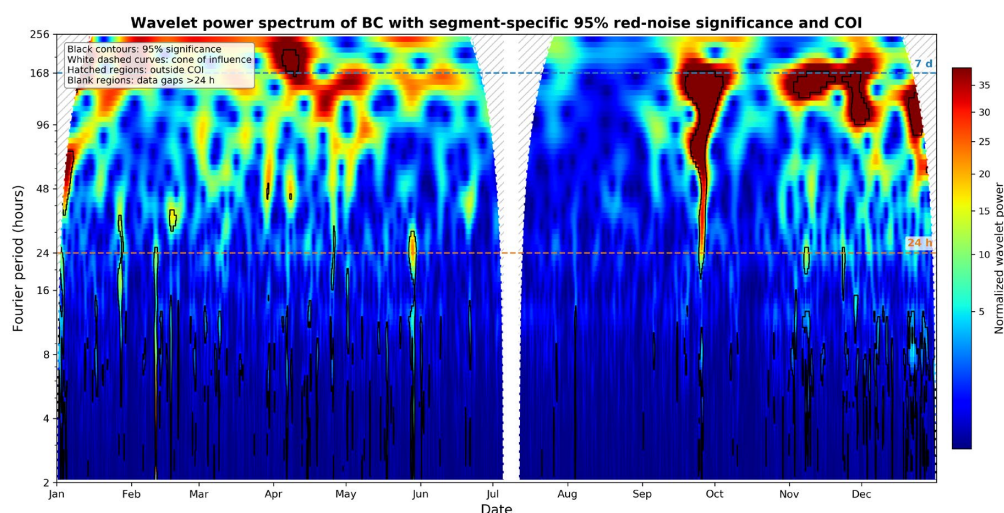
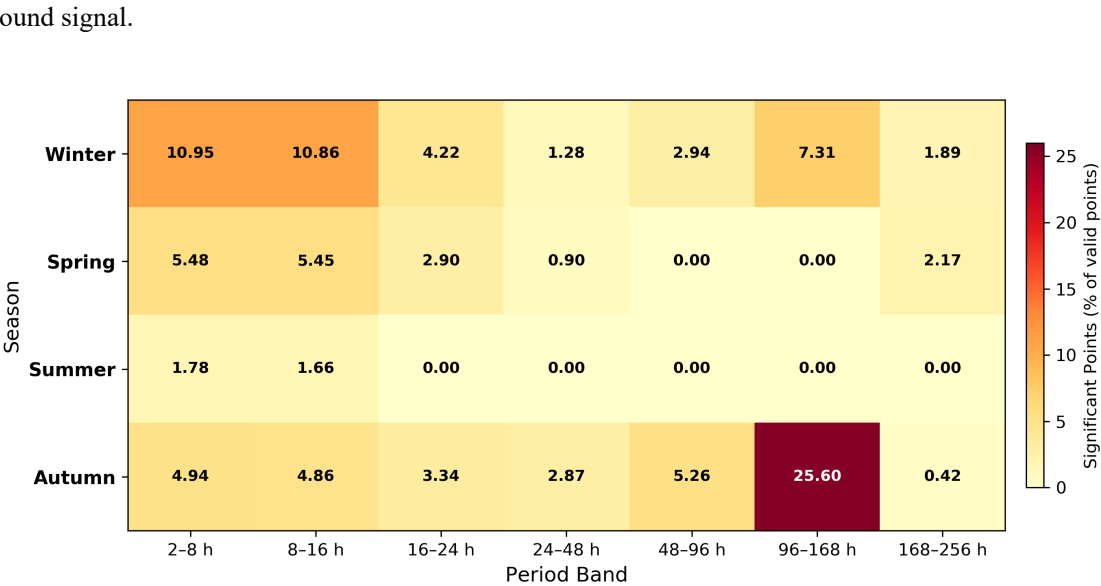


Figure 3. Segment-aware wavelet power scalogram of hourly BC concentration. Black contours denote 95% significance against an AR(1) red-noise background. White dashed curves and hatching denote the segment-specific cone of influence. Blank vertical intervals indicate unresolved gaps longer than the interpolation threshold. Significant multi-day variability is intermittent, with no persistent year-

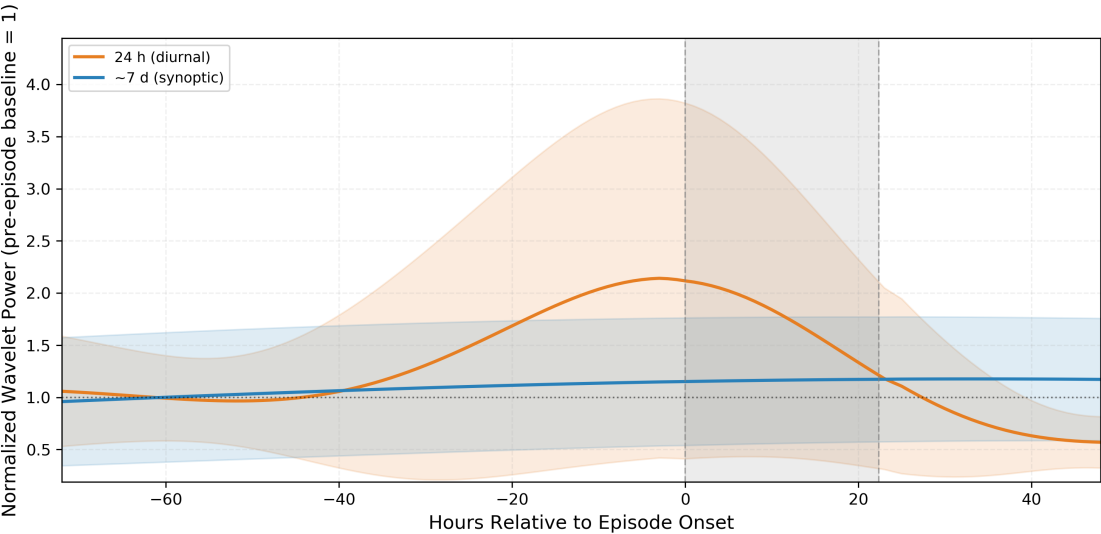


573

574 Figure 4. Seasonal distribution of significant wavelet power across period bands. Values are the

575 percentage of valid time-frequency points inside the cone of influence that exceed the 95% red-noise

576 significance level.



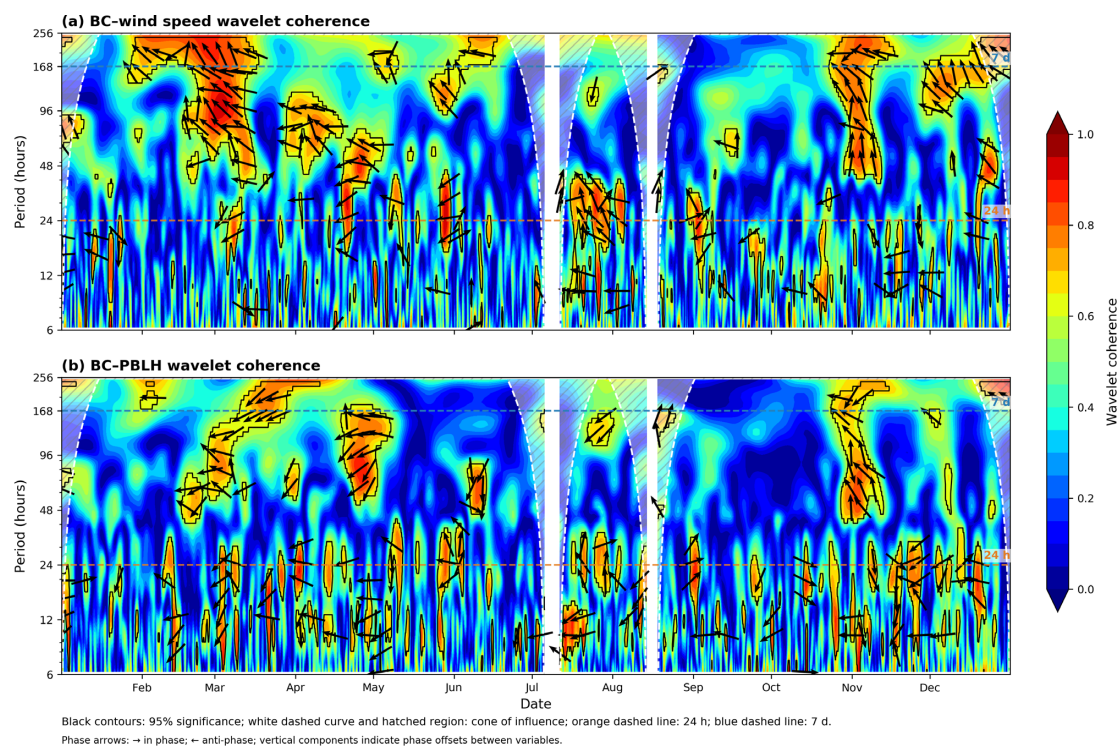
577

578 Figure 5. Composite evolution of 24-h and ~7-day wavelet power around six high-BC episode clusters

579 aligned at episode onset. Power is normalized to the -72 to -48 h pre-episode reference interval;

580 shading denotes ± 1 standard deviation across episodes. The composite describes episode-centered

581 evolution and is not used as a predictive precursor metric.



583

584 Figure 6. Wavelet coherence between BC and (a) wind speed and (b) PBLH. Black contours indicate
585 95% significance; white dashed curves and hatching denote the segment-specific cone of influence;
586 reference lines mark 24 h and 7 days. Phase arrows indicate the relative phase between paired variables.

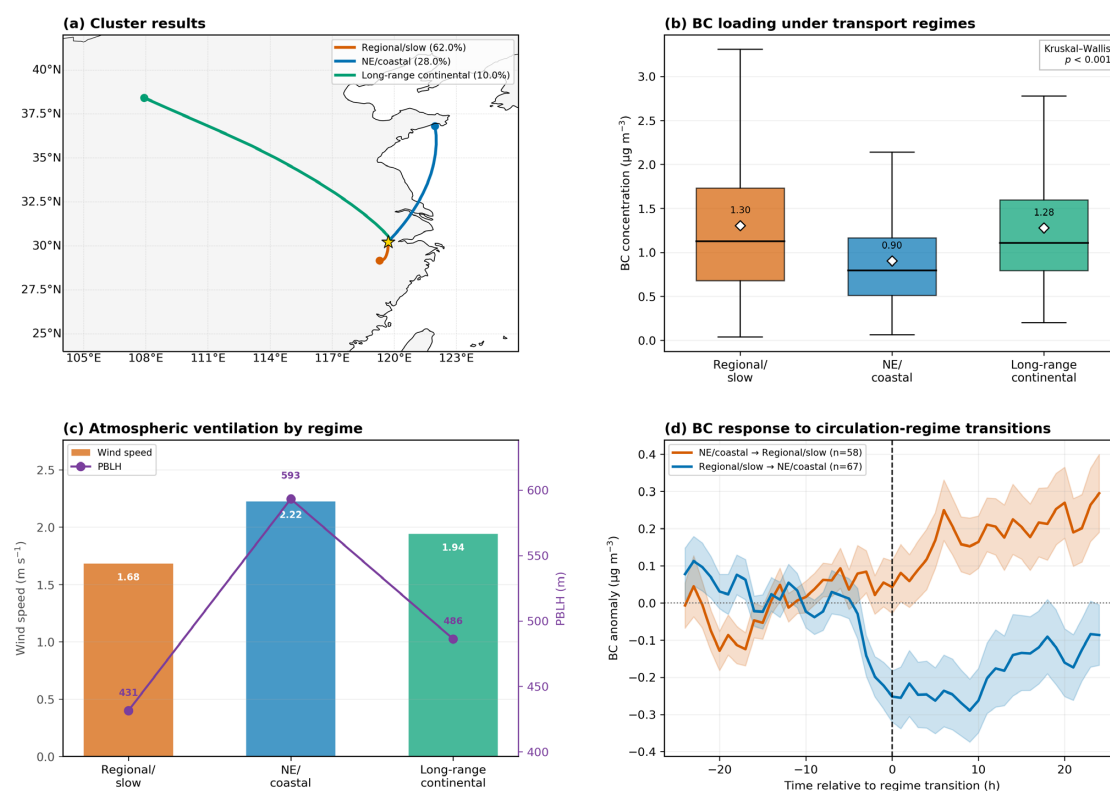


Figure 7. Transport and ventilation regimes associated with BC variability. (a) Cluster results showing mean 48-h trajectories for the three annual transport regimes. (b) BC distributions by regime; Kruskal–Wallis $p < 0.001$. (c) Mean wind speed and PBLH by regime. (d) Composite BC anomalies around transitions between NE/coastal and Regional/slow circulation states.

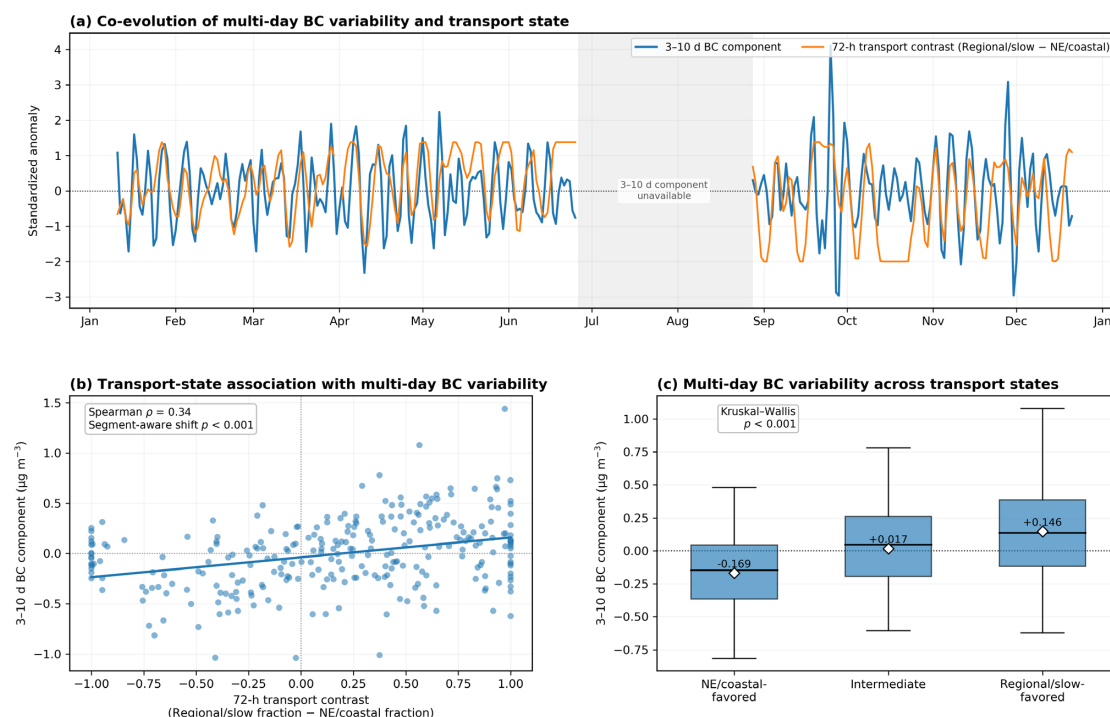


Figure 8. Synoptic transport-state modulation of the independent 3–10-day BC component. (a) Co-evolution of the standardized 3–10-day BC component and the 72-h Regional/slow-minus-NE/coastal transport contrast; the shaded internal interval denotes dates for which the 3–10-day component is unavailable because of the long observational gap and conservative filter-edge exclusion. (b) Daily association between transport contrast and the 3–10-day BC component (Spearman $\rho = 0.345$; segment-aware circular-shift $p < 0.001$). (c) Distribution of the 3–10-day BC component under NE/coastal-favored, intermediate, and Regional/slow-favored conditions; Kruskal–Wallis $p < 0.001$.



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