

Supplementary Materials

1. Correlation of CDSS and other station in winter (December in 2022 to February in 2023).

Welch's two-sided t-tests (unequal variances) were applied to hourly means data set of CDSS and other background stations. It illustrated that CDSS is significantly higher than other sites for both CO₂ and CH₄ (p<0.001, in table S1).

For CO₂, the relative differences range +3.1% to +51.6% (absolute +13.2 to +148.2 ppm); for CH₄, +5.9% to +28.5% (absolute +116 to +462 ppb). Significance is marked as: *, p<0.05; **, p<0.01; ***, p<0.001. The tests results are based on the hourly time series (sample sizes and SDs in table). Minor numerical differences are due to rounding and do not affect the conclusion.

Table S1 Welch's t-tests

Gas	Reference site	CDSS mean	CDSS SD	CDSS n	Ref mean	Ref SD	Ref n	Mean diff (CDSS – Ref)	Mean diff (%)	t	df	p (two-sided)	Significance
CH4	GSN	2083.454	56.498	2140	1924.801	582.007	2160	158.654	8.243	12.609	2200.064	0	***
CH4	LLN	2083.454	56.498	2140	1967.305	15.974	21	116.149	5.904	31.446	25.208	0	***
CH4	MKO	2083.454	56.498	2140	1909.972	312.997	2015	173.482	9.083	24.507	2137.576	0	***
CH4	MLO	2083.454	56.498	2140	1939.381	9.103	36	144.074	7.429	73.971	94.404	0	***
CH4	MNM	2083.454	56.498	2140	1788.905	728.929	2160	294.549	16.465	18.723	2185.181	0	***
CH4	RYO	2083.454	56.498	2140	1621.7	1020.371	2160	461.754	28.473	20.999	2172.361	0	***
CO2	GSN	435.288	7.642	2140	375.819	273.414	2160	59.47	15.824	10.105	2162.405	0	***
CO2	LLN	435.288	7.642	2140	422.051	1.778	21	13.237	3.136	31.393	27.901	0	***
CO2	MKO	435.288	7.642	2140	403.552	150.857	2015	31.736	7.864	9.432	2023.733	0	***
CO2	MLO	435.288	7.642	2140	420.109	1.328	36	15.179	3.613	54.963	84.432	0	***
CO2	MNM	435.288	7.642	2140	287.116	416.894	2160	148.172	51.607	16.516	2160.464	0	***
CO2	RYO	435.288	7.642	2140	356.818	309.844	2160	78.471	21.992	11.767	2161.651	0	***

Note: (1) MLO (Mauna Loa Observatory, Hawai'i, USA), MKO (Mauna Kea Observatory, Hawai'i, USA), LLN (Lulin Observatory, Taiwan, China), MNM (Minamitorishima, Ja-pa-n), GSN (Gosan Station, Jeju, Korea), RYO (Ryori Observatory, Iwate, Japan);(2) Gas denotes the measured species (CO₂ in ppm, CH₄

in ppb); Reference site is the background comparison site; CDSS mean, CDSS SD, and CDSS n are the CDSS hourly mean concentration, standard deviation, and sample size for winter (December 2022 to February 2023); Ref mean, Ref SD, and Ref n are the corresponding values for the reference site; Mean diff (CDSS – Ref) is the difference of means (same unit as the reference), with positive values indicating CDSS is higher; Mean diff (%) is the relative difference computed as $(\text{CDSS} - \text{Ref}) / \text{Ref} \times 100\%$; t and df are the Welch t statistic and degrees of freedom; p (two-sided) is the two-sided p-value; Significance gives the significance codes (*: $p < 0.05$; **: $p < 0.01$; ***: $p < 0.001$); a p-value displayed as “0” indicates $p < 0.001$ due to rounding; all results are based on hourly time series.

.

2. Robustness and Uncertainty Checks

To further investigate the methodological framework and associated uncertainties in the study, validation of Equation (4) is conducted through two experiments: One-At-a-Time (OAT) perturbation analysis and Leave-One-Event-Out (LOEO) analysis. These experiments primarily assess the stability of the input perturbations within the three-factor framework of Equation (4). The OAT method aims to confirm whether the derived provincial and sectoral emission contributions remain robust to variations in key input factors (i.e., trajectory-based spatial weights, source category weights, and emission inventory data). The LOEO analysis, in contrast, evaluates the impact of sample size changes on the results.

One-At-a-Time (OAT) Perturbation Analysis: In the OAT approach, each key input factor is perturbed individually by a predefined percentage (either increased or decreased) while keeping other factors constant. Specifically, the following inputs are perturbed:

The spatial weight (Q) for each province (j); The weight (W) for each emission source category (c); The winter emissions (E) (organized by province (j) as rows and category (c) as columns).

After each perturbation, the model is recalculated, and the resulting provincial and sectoral contributions are compared against the baseline (unperturbed) results. This process determines the sensitivity of the theoretical framework to input variations.

Leave-One-Event-Out (LOEO) Analysis: The LOEO analysis involves systematically removing each of the 13 identified regional transport events (after temporal alignment of online and sampling data) and recalculating the attribution model. For each excluded event, new provincial and sectoral contributions are derived and compared with the baseline to assess result stability.

Figure S1 presents the LOEO validation results. Each row represents a scenario where a specific event is excluded, while each column corresponds to a province. The color intensity indicates the relative change in a province's contribution compared to the baseline (white represents negligible change, light blue/orange represents minor deviations). The figure reveals that most cells remain near-white, with only a few event-province combinations showing slight color variations. This indicates that the results are not dominated by any single transport event, and the spatial attribution patterns are robust—removing any event does not significantly alter the overall ranking or magnitude of contributions from key provinces.

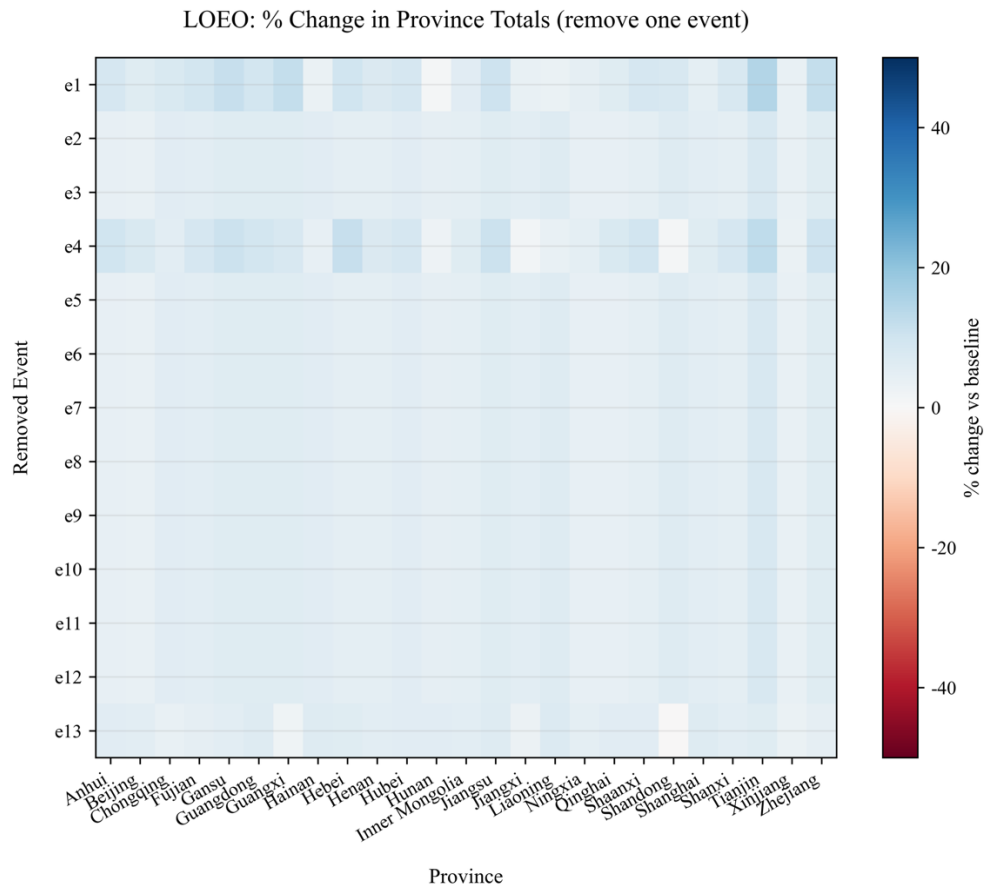


Figure S1. Leave One Event Out Provincial Share Change Heatmap

Figure S2 examines the stability of sectoral contribution results under the Leave-One-Event-Out (LOEO) procedure. For each of the 13 single-event exclusion scenarios, the contributions of the five major emission source categories (vehicle emissions, fuel combustion, solvent use, oil and gas production, and natural gas emissions) were recalculated and compared against the baseline. The figure presents the percentage contribution of each sector across all scenarios, allowing for a visual assessment of how the removal of individual events affects each sector's share.

The sectoral contributions exhibit only minor variations across different scenarios. The dominant source category, vehicle emissions (accounting for ~70% of the baseline contribution), remains the largest contributor in all scenarios. The removal of any single event does not alter the ranking of the top-contributing sectors. The second and third-ranking sectors—fuel combustion (~15%) and solvent use (~10%)—also show consistent contributions, with fluctuations typically within a few percentage points. For the two least significant categories (oil and gas production and natural gas emissions, each contributing only a few percent of the total), the exclusion of specific events leads to relatively larger percentage changes (with relative differences of up to ~45%), and even a rank reversal in two scenarios. However, these substantial relative fluctuations occur in categories with very small absolute contributions (~1–3% of total emissions) and do not affect the ranking of the major sectors.

The sectoral attribution results are robust to the removal of individual events. The dominant role of vehicle emissions remains unchanged, and the overall hierarchy of sector contributions (Transportation > Fuel Combustion > Solvent Use > Others) is preserved across

all LOEO tests. While the minor categories show some sensitivity in relative terms, their absolute impact remains negligible. This consistency underscores the reliability of the identified key source sectors influencing Hong Kong’s greenhouse gas levels across scenarios, reinforcing confidence in the sectoral attribution results despite the limited event sample.

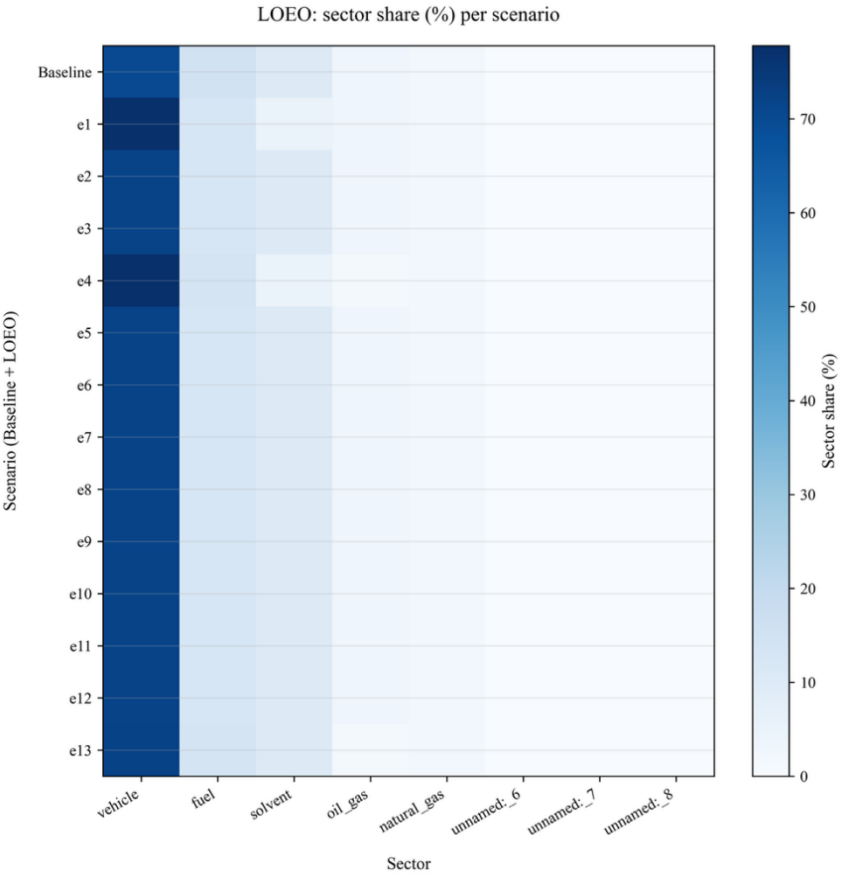


Figure S2 Leave One Event Out Sector Share Stability Matrix

Figure S3 summarizes the observed variations in provincial and sectoral contributions across all LOEO experiments. For each LOEO scenario (i.e., removal of a single event), the maximum absolute change in contribution for any individual province and the maximum absolute change for any individual sector (both expressed in percentage points of the total) were recorded. The figure plots the maximum percentage changes observed at both the provincial and sector levels for each scenario, with the two categories clearly distinguished.

The results indicate that, in most cases, the removal of a single event induces only limited changes. Out of the 13 event-removal scenarios, 10 exhibit near-zero variation in provincial contributions, while the maximum sector-level changes remain relatively low (approximately 2%). Only a few scenarios deviate from this pattern: for example, the removal of Event #4 yields the largest provincial contribution change (around 1%) and the most significant sector-level variation (around 8%). This confirms that the attribution results exhibit bounded sensitivity to individual events, with impacts remaining extremely limited.

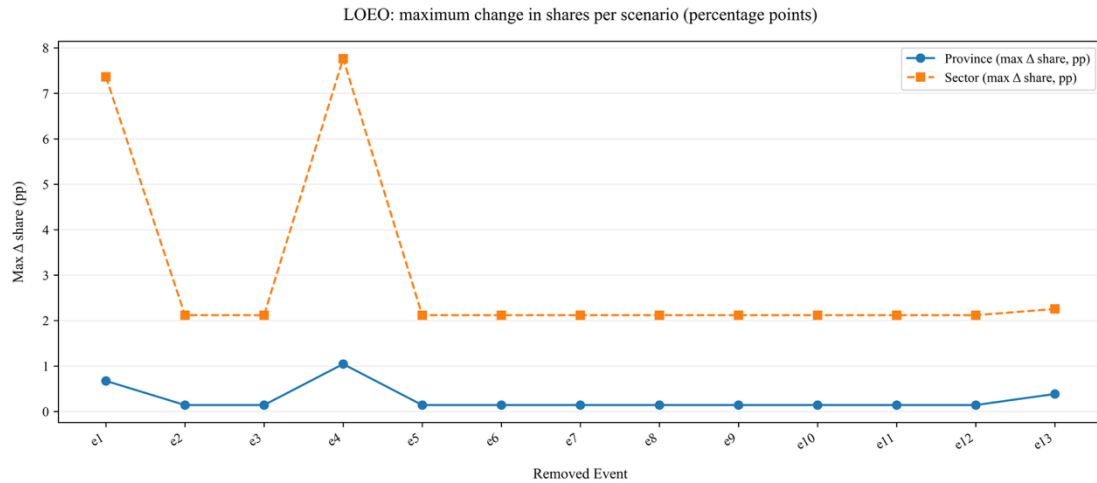


Figure S3 Leave One Event Out Variability Summary of Maximum Changes

Figure S4a summarizes the number of "flips" in provincial rankings under single-factor perturbations (rows/columns of Q, W, E) and perturbation magnitudes ($\pm 10\%$, $\pm 20\%$). Overall, $\pm 20\%$ perturbations are more likely to trigger flips than $\pm 10\%$, and factors directly related to transport flux or provincial emission intensity (e.g., row perturbations of Q or E) induce more flips than column perturbations of W or E.

It is important to note that these flips are concentrated in mid-tier provinces with baseline shares close to each other, while dominant provinces rarely experience flips. This suggests that, within reasonable input uncertainty ranges, the dominant pattern remains robust, and the overall hierarchical structure is preserved.

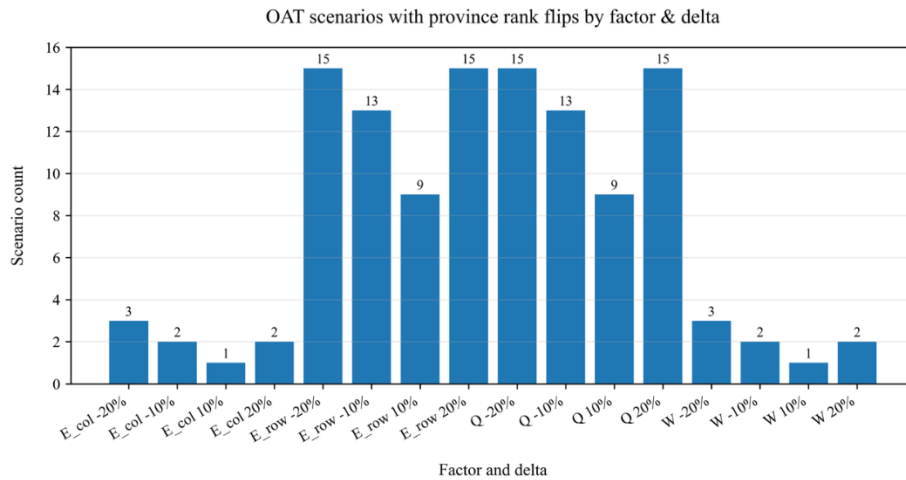


Figure S4a OAT rank - flip counts by factor and perturbation magnitude (provinces)

Sector-level rankings are highly stable under the one-at-a-time (OAT) sensitivity tests: applying $\pm 10\text{--}20\%$ perturbations with internal renormalization produces no rank reversals among dominant sectors and only occasional, adjacent swaps among minor sectors, accompanied by negligible changes in absolute contributions. Consistent outcomes from the leave-one-event-out (LOEO) check indicate that the inferred sectoral hierarchy is insensitive to plausible input uncertainty.

Figure S4b visualizes, via a heatmap, sector - level rank deltas across all OAT scenarios. Most cells are zero, with only sporadic ± 1 deltas for a few low-share sectors and no larger

jumps observed.

This indicates that sectoral ordering is effectively preserved even under stronger single-factor shocks, consistent with the near-zero flip counts in S4b.

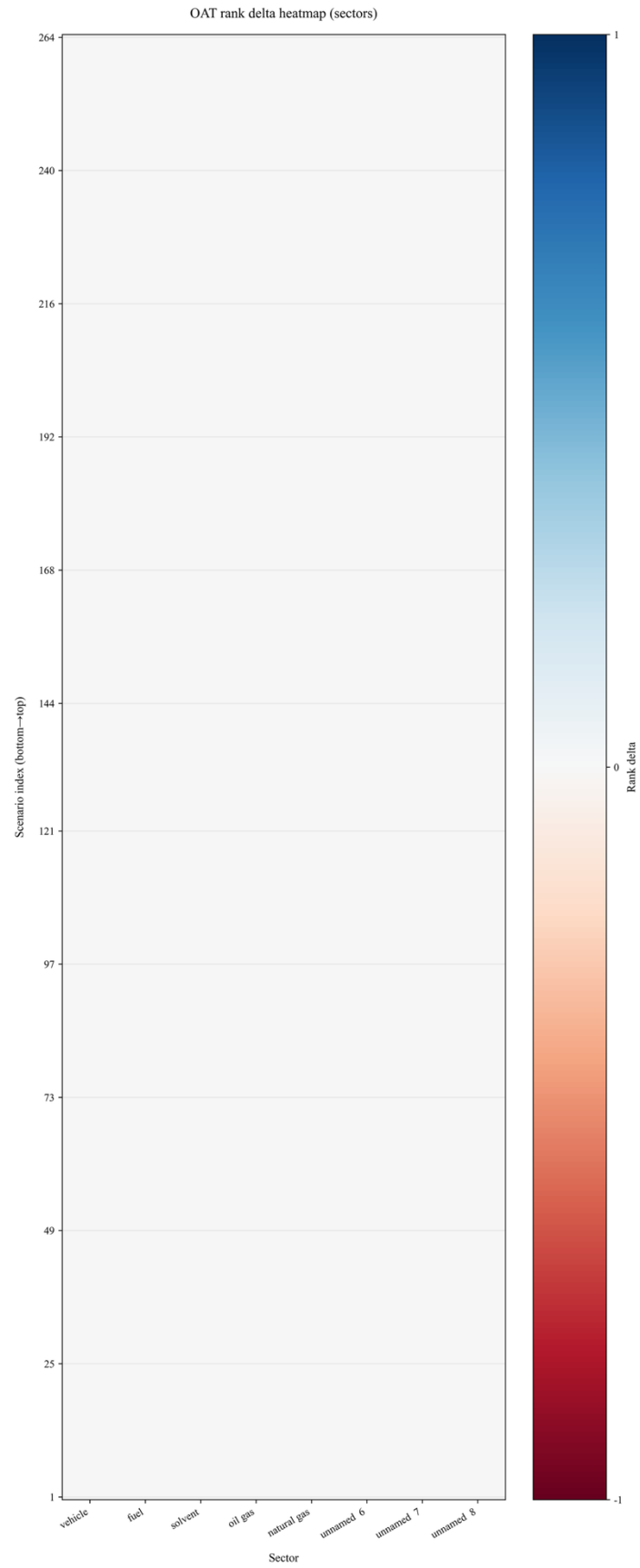


Figure S4b — OAT rank-delta heatmap (sectors)

Figure S4c's provincial rank-delta heatmap reveals that non-zero ranking changes are predominantly confined to mid-tier provinces, with ± 1 shifts being the most common. Meanwhile, the dominant provinces remain largely unchanged. This observation is consistent with the statistical finding in Figure S4a that "flips frequently occur among mid-tier provinces with similar shares," further supporting the robustness of the three-tier provincial hierarchy under perturbations.

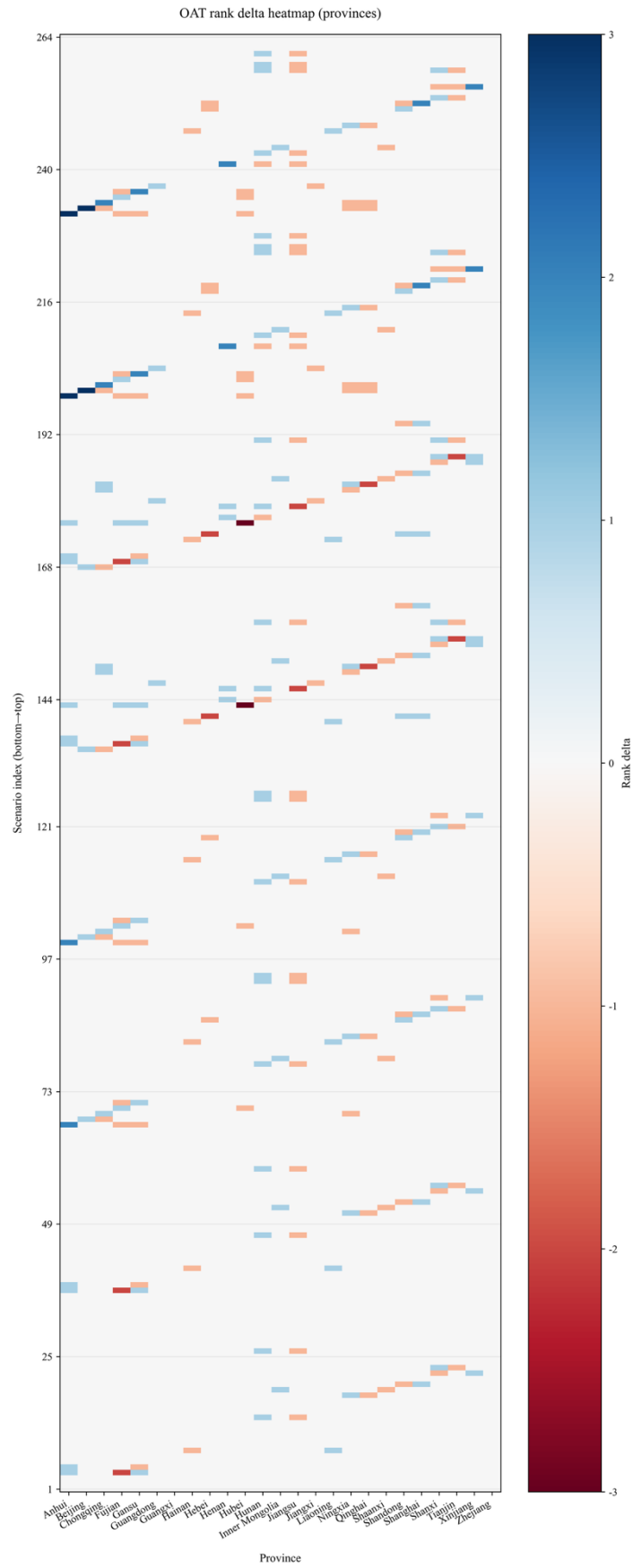


Figure S4c — OAT rank-delta heatmap (provinces)

Figure S4d presents the distribution of ranking flips across provinces under all OAT scenarios. The results show a right-skewed distribution: the majority of provinces experience few flips, while a small number of mid-tier provinces (with similar baseline shares) exhibit frequent adjacent-rank swaps under multiple perturbation combinations. In contrast, dominant provinces show very few flips, further validating that the key conclusions are insensitive to input uncertainty.

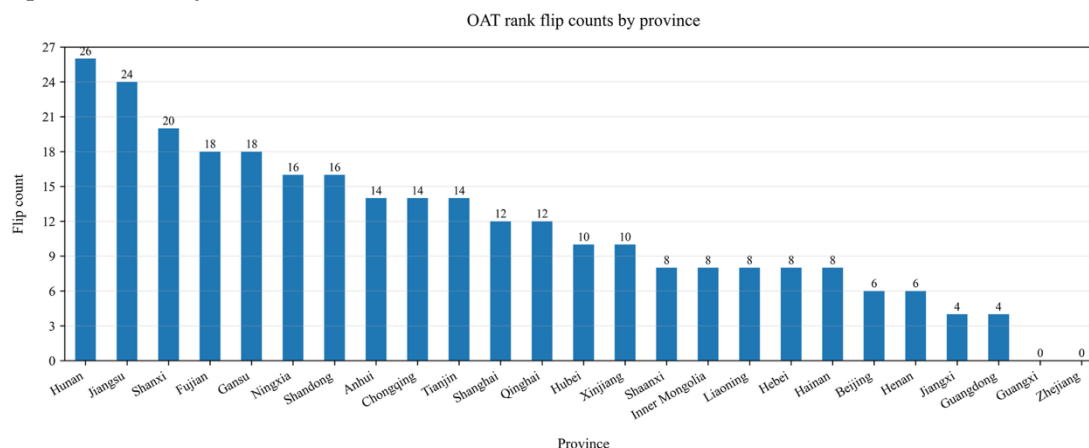


Figure S4d — OAT rank - flip counts by province

Figure S5a illustrates the impact of small ($\pm 10\%$) single-factor perturbations to the input parameters on the attribution results. Each subplot focuses on one input factor (spatial weight Q at the provincial level, source category weight W , or emissions E), showing how provincial and sectoral contributions change when the factor is increased or decreased by 10%.

In these experiments, the provincial contribution shares were recalculated after individually adjusting Q for each province by $\pm 10\%$ (with Q normalized afterward). Similarly, each source category's weight W was perturbed by $\pm 10\%$ (with normalization), and the emissions E for each province or category were scaled by $\pm 10\%$ to observe the resulting changes.

The attribution results exhibit extremely weak sensitivity to $\pm 10\%$ input variations. The maximum change in any province's share is approximately 8–10%, typically occurring when the perturbed factor directly corresponds to the province (e.g., increasing E or Q for a high-emission province by 10%). Crucially, for such small perturbations, no significant ranking shifts in provincial contributions are observed, with only occasional swaps between provinces with nearly identical baseline contributions. The dominant contributing provinces remain unchanged and continue to dominate. At the sector level, the changes in sector shares are even smaller—usually within 5%—and no sector ranking changes occur under the $\pm 10\%$ tests. Overall, applying a 10% increase or decrease to any single input leads to only minor adjustments in the output, leaving the qualitative features of the attribution landscape intact. The OAT ($\pm 10\%$) experimental results confirm that the framework is robust to minor input uncertainties. Whether the trajectory counts, source apportionment weights, or emission inventories contain errors of around 10%, the conclusions are not substantively affected: the key contributing provinces and sectors remain identifiable, and their relative importance is largely preserved.

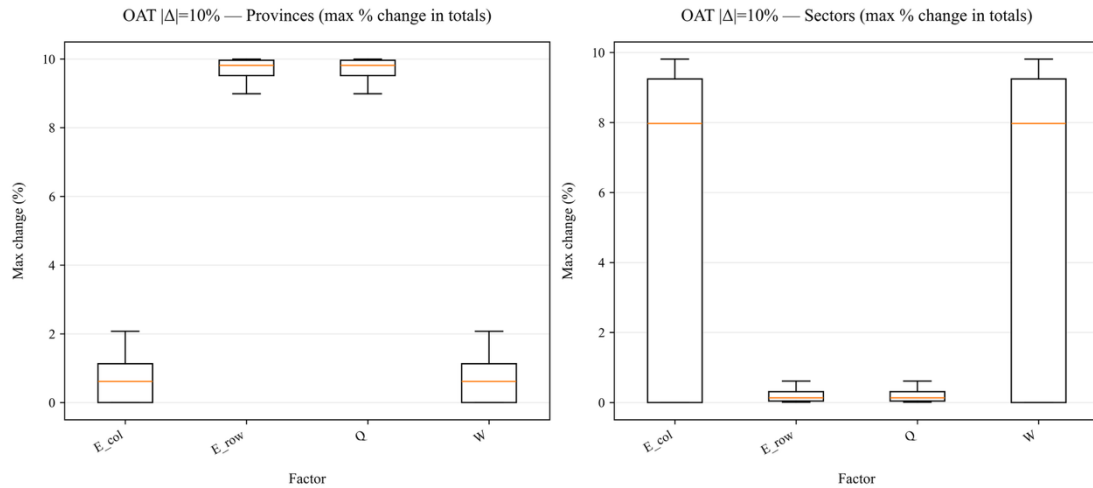


Figure S5a One At a Time Sensitivity Ten Percent Provincial and Sector Boxplots

Figure S5b extends the single-factor sensitivity analysis to a larger $\pm 20\%$ perturbation range. Following the same procedure as in Figure S5a, each key factor (Q, W, or E) was individually increased or decreased by 20%, and the resulting changes in provincial and sectoral contributions were plotted. This analysis tests whether the linear and stable characteristics observed at $\pm 10\%$ persist under higher perturbation magnitudes.

Even with $\pm 20\%$ input variations, the attribution results remain qualitatively stable. Compared to the $\pm 10\%$ case, the magnitude of provincial contribution changes increases—with some provinces experiencing maximum shifts of nearly 20%, commensurate with the input perturbation scale. In specific $\pm 20\%$ scenarios, rank swaps occur between provinces with initially similar contributions, but the overall provincial hierarchy is preserved: notably, the top two contributing provinces (e.g., Guangdong, Jiangxi) consistently retain their leading positions across all tests.

At the sector level, the relative changes are more pronounced for smaller categories (up to 10–20%), while dominant sectors (e.g., vehicle emissions) exhibit minimal fluctuations. Crucially, sector rankings remain unchanged even under $\pm 20\%$ perturbations: the same dominant sector (vehicle emissions) and the second-largest sector (fuel combustion) maintain their respective positions in all scenarios. The relationship between input changes and output responses remains approximately linear, indicating no nonlinear sensitivity at the 20% level.

The results demonstrate that the $\pm 20\%$ OAT experiments confirm the robustness of the attribution framework under higher input uncertainty. Even if a key input parameter is over- or underestimated by up to 20%, the calculated contributions for regions and sectors do not undergo drastic shifts. The core pattern—identifying which provinces and sectors are the largest contributors—remains largely intact, further validating the reliability of the overall conclusions derived from this method within reasonable uncertainty bounds.

This outcome addresses potential concerns by showing that the framework's underlying multiplicative independence assumption does not mask higher-order sensitivities: the response of contributions to input changes is proportional and predictable.

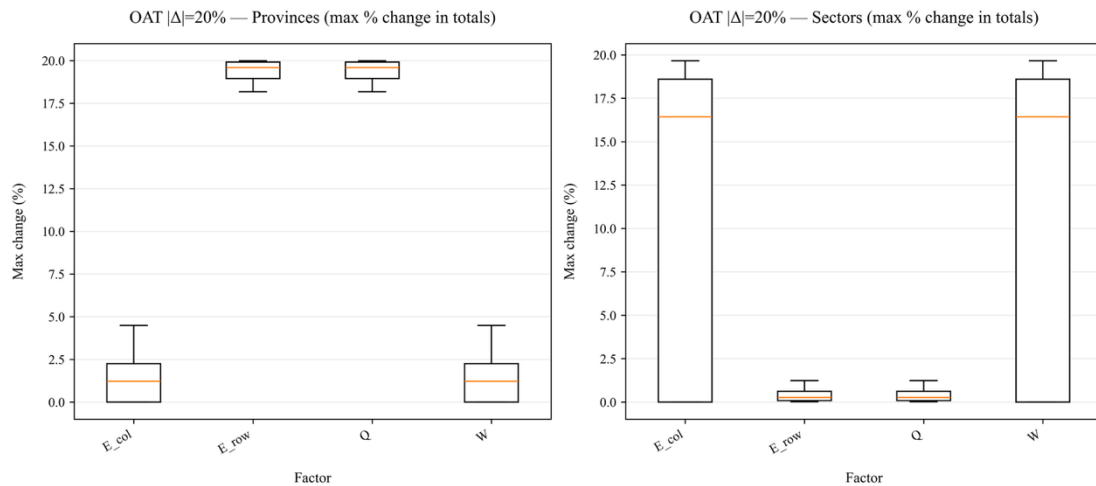


Figure S5b One At a Time Sensitivity Twenty Percent Provincial and Sector Boxplots

Figure S6 provides an overall overview of the model's robustness by directly comparing the baseline attribution results with those from perturbed scenarios. In this figure, the contributions of each province in the baseline scenario are plotted against their corresponding values under OAT (one-at-a time) perturbation scenarios (e.g., each point represents a province's baseline contribution versus its contribution after a $\pm 20\%$ input perturbation). A 45° reference line is included to indicate perfect agreement. The figure also displays the range of contribution values (minimum–maximum) for each province and sector across all sensitivity tests.

The results show that the major contributing provinces cluster in the higher contribution range and lie close to the diagonal, indicating only minor deviations in their contribution values under perturbations compared to the baseline. In contrast, the contributions of smaller provinces exhibit slightly more dispersion but remain confined within a narrow band.

For each province, the total variation in its contribution across all test scenarios is minimal relative to its baseline percentage. Similarly, the contribution ranges for each sector reveal that dominant sectors experience negligible changes, while minor sectors, though showing relatively larger spans, still have very small absolute contributions.

Figure S6 synthesizes the sensitivity analysis results, confirming the robustness of the three-factor attribution model's outputs. The proximity of the scatter points to the diagonal indicates that the contribution distributions under perturbed scenarios closely resemble the original case. The limited vertical deviation ranges for all provinces and sectors across tests suggest that uncertainties are bounded.

From a practical perspective, this means our key conclusions—identifying the regions and sectors most influential on the target area (e.g., Hong Kong) under typical regional transport event constraints—remain valid.

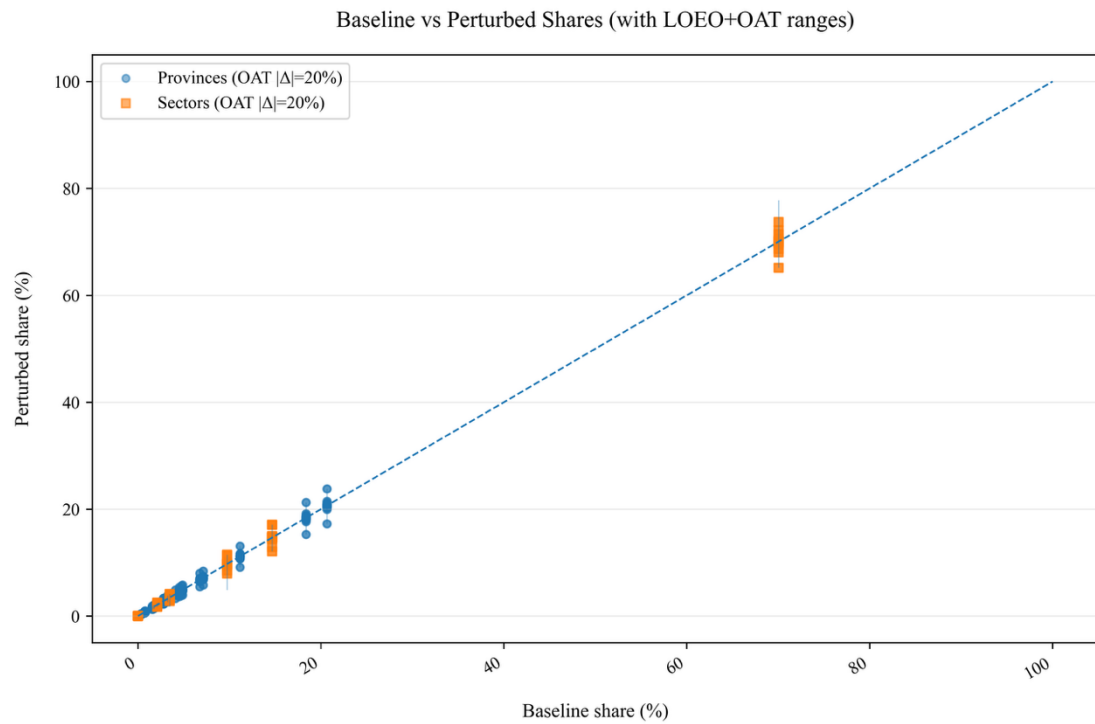


Figure S6 Overall Robustness Baseline Versus Perturbed Shares With Ranges