Dear editor Roya,

Thank you for your email and for giving us another opportunity to revise our manuscript. We sincerely appreciate your constructive comments. In response to your concerns regarding potential changes in background isotopic values and their associated uncertainties, we have carefully addressed these issues in our revision. Specifically, we have incorporated a multi-isotope MixSIAR model for comparative analysis in Taiyuan. Below is our detailed response to your comments:

Major Comments

Please add a discussion to the manuscript on the limitations of the sample size, statistical significance tests for the results tabulated in Table S1, and a discussion of uncertainties/caveats/biases that the results have, especially during the dust evens and at sites where the local isotopic signatures have a similar mean to the various Gobe desert source samples.

Answer: Thank you for your suggestion. A more discussion of uncertainties/caveats/biases that the results have been added in revised manuscript. Please refer to Section 4.3. Figures S3, S4 and S5 as well as stable isotope analysis using MixSIAR in R (Text 2) have been added in supplementary Materials.

In this study, we applied a two-endmember mixing model comparing compound-specific nitrogen isotopic values of CAAs from Gobi dust source with those in urban aerosols during non-dust periods (representing the atmospheric background values), rather than those in local dominant plants, road dust and anthropogenic activities sources. Our methodology relies on the fundamental assumption that the nitrogen isotopic composition of CAAs in urban aerosols during the dust period, which derived from local dominant plants, road dust and anthropogenic activities, does not significantly differ from background atmospheric values.

For the Beijing, Tianjin and Shijiazhuang sampling sites, meteorological conditions (wind speed, relative humidity, and temperature) did not differ significantly ($p > 0.05$) between dust and non-dust periods (Figures S3 and S4). Under these stable conditions, local urban emission sources remained consistent, maintaining unchanged atmospheric background values. Therefore, the application of a two-endmember mixing model - utilizing isotopic values of compound-specific CAAs from Gobi dust sources and urban aerosols during non-dust periods - provides a scientifically robust approach for quantifying Gobi dust contributions in Beijing, Tianjin and Shijiazhuang.

At the Taiyuan sampling site, wind speeds during dust events were significantly higher than during non-dust periods ($p < 0.05$; Figures S3–S4). These strong winds may enhance entrainment of local plant debris and road dust into aerosols, potentially modifying baseline $\delta^{15}N$ signatures of CAAs. This suggests CAA $\delta^{15}N$ values during the non-dust period (atmospheric background) may not fully represent local signatures during dust events, potentially affecting source apportionment. To evaluate this effect, we applied the MIXSIAR model (Stock and Semmens, 2016; Song et al., 2021) with $\delta^{15}N$ values of both glycine and leucine from local and the Gobi dust sources (Table S1, details were provided in Supplementary Materials, Text2). The MIXSIAR model showed that in Taiyuan, the relative contributions of local dominant plants, road dust, and anthropogenic activities sources to aerosol CAAs averaged 1.6 ± 2.6%, 46.2 ± 20.9% and 45.8 ± 12.2%, respectively, during the non-dust period. The dust period exhibited modified contribution profiles: local plants

(12.6±10.3%), road dust (32.7±19.8%), and anthropogenic emissions (35.9±10.0%), along with an external contribution from Gobi dust sources (18.7±14.1%). The Gobi dust contribution estimated by MIXSIAR showed agreement with our two-endmember model estimates (17 ± 25% ~ 23 ± 10%). After normalizing MIXSIAR results to exclude Gobi dust contributions, natural sources (local plant + road dust) showed only a modest increase from 47.8% (non-dust) to 55.8% (dust periods), representing an 8% enhancement.

During this dust sampling campaign, only the Taiyuan site exhibited significant wind speed increases, while other sampling locations showed no remarkable meteorological variations. Future research should incorporate more extended observational periods of dust events, with particular emphasis on downwind areas experiencing significant meteorological changes. Such extended investigations will enable more accurate assessment of how long-range transported dust sources influence biogeochemical cycles in downwind ecosystems.

Besides that, uncertainties associated in the two-endmember mixing model analysis, which derived from the $\delta^{15}N$ variabilities of combined Gly and Leu in emission sources (mean ± SD $\delta^{15}N$ values) were added in our revised manuscript. Please refer to line 607~608. The contributions of dust sources to Beijing, Tianjin, Shijiazhuang, and Taiyuan during this dust event were calculated for glycine as 94 ± 17%, 78 ± 7%, 36 ± 1%, and 17 ± 25% respectively. For leucine, the contributions were 98 ± 23%, 83 ± 11%, 44 ± 12%, and 23 ± 10%, respectively.

Stable Isotope Analysis in R

The Bayesian mixing models in R model (MixSIAR) has been widely used to determine diet composition, population structure, and animal movement, because it can use the isotope values (biotracer data) to estimate the proportions of source contributions to a mixture, and incorporate the uncertainties associated with source isotope values (Stock, and Semmens, 2016, Song et al., 2021). In our study, the MixSIAR model with $\delta^{15}N$ values of both glycine and leucine was used to incorporate source apportionment of CAAs in Taiyuan during the non-dust period and dust period, respectively. If quantifying dust source contributions using nitrogen isotopic values from individual amino acids (e.g., glycine or leucine alone) by mixing local and Gobi dust sources would introduce larger uncertainty. This limitation stems from the fact that $\delta^{15}N$ values of glycine or leucine in Gobi dust show no statistically significant differences compared to those of urban road soils across all study sites ($p > 0.05$; Figure S5). Therefore, both $\delta^{15}N$ values of both glycine and leucine from local and the Gobi dust sources were used to minimal uncertainty. Details on the assumed values for each potential end-member are of great importance. As discussed in section 4.1, local dominant plants, surface road dust and anthropogenic industrial activities were the major sources of CAAs in $PM_{2.5}$ during the non-dust period. Therefore, three major sources, including local dominant plants, surface road dust and anthropogenic sources were used to simulate the relative contributions of major urban protein sources during non-dust periods in Taiyuan (Table S1). Four distinct source categories were used to calculate their respective contribution to urban proteinaceous matter in $PM_{2.5}$: dust from Gobi Desert, local dominant plants, road dust and anthropogenic activitiy sources in Taiyuan. In our estimations, uncertainties associated in the source analysis were derived from the $\delta^{15}N$ variabilities of major combined Gly and Leu sources (mean ± SD $\delta^{15}N$ values of each source were input; Table S1). The result of MixSIAR model was exhibited in Table S5.

All changes can be tracked in the revised manuscript. Thank you very much again.

Yours sincerely,
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References

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