Dear Reviewer:

Thank you for your letter and comments concerning our manuscript EGUSPHERE-2024-2065 entitled "Dust storms transport proteinaceous matter from the Gobi Desert to Northern China". Those comments are all valuable and very helpful improving our paper. As suggested by Reviewer 1, the terms "dust storm" and "dust event" were used interchangeably throughout the manuscript. To maintain consistency, the revised manuscript exclusively uses the term "Asian dust events." Consequently, the title has been updated to "Asian Dust Transport of Proteinaceous Matter from the Gobi Desert to Northern China." We have incorporated your suggestions and revised the manuscript according to the points as follows.

Anonymous Referee #1

The manuscript addresses an interesting and emerging issue regarding the role of dust storms in aerosol transport, its effect on particulate matter composition, and its role in critical environmental issues such as air quality and ecosystem productivity. Specifically, the authors assessed the contribution of Gobi dust to proteinaceous combined amino acids (CAAs) in PM2.5 across four urban regions in Northern China during dust events. The authors collected data from multiple sites (Beijing, Tianjin, Shijiazhuang, Taiyuan), which allows for a regional comparison of CAAs in PM2.5. Their approach involved analyzing the concentrations and δ 15N isotopic signatures of CAAs from both the Gobi desert and local urban sources. Additionally, they quantified the Gobi dust's input to CAAs in PM2.5 and evaluated the dry deposition fluxes of protein-N to explore the biogeochemical impacts. Overall, the rationale for this study is well stated, the experiment is well described, and the work points out important issues that are highly relevant. However, the manuscript needs some improvement.

Major Comments

Q1: The comparison of protein characteristics in PM2.5 from urban environments with those in dust sources lacks a detailed interpretation of its broader implications, being limited only to surface-level comparison. The study could benefit from a more in-depth discussion of the potential biological or chemical mechanisms by which dust-borne CAAs affect the local and regional nitrogen cycle. This could help link the findings to broader environmental implications.

Answer: Thank you for your suggestion. A more discussion of the potential biological or chemical mechanisms by which dust-borne CAAs affect the local and regional nitrogen cycle were added in the manuscript. Please refer to Section 4.2, 4.3 and 4.4 in discussion.

Three biological and chemical mechanisms may explain the variation in protein characteristics in $PM_{2.5}$ during the dust period, as discussed in the revised manuscript.

First, the correlation of total CAAs concentrations in PM_{2.5} with the local meteorological conditions

(including temperature, humidity or wind speed) and PM₁₀ concentration (Table S4) showed that the increase concentration of CAAs in PM2.5 during the dust period was not influenced by local meteorological conditions, but were directedly linked to the long-transported Gobi dust source. Then, we used compound-specific nitrogen isotope analyses of individual CAAs to estimate the contribution of protein from dust sources and its dry deposition flux. The results indicated a decreasing trend in the dry deposition of protein-N along the transport pathway. This suggests that the increased airborne protein concentration in downwind areas during dust events may be depend on the dry deposition of dust particles, with protein either coagulating or condensing onto them. Proteinaceous matter in the atmosphere has been shown to serve as a bioavailable nitrogen source for plants and microorganisms (Ho et al., 2015; Samy et al., 2013; Zhang and Anastasio, 2003). Thus, protein-N from the Gobi Desert may act as a nutrient source for marine ecosystems, significantly contributing to ecological processes in China's marginal seas, such as promoting phytoplankton fertilization and stimulating nitrogen fixation (Duarte et al., 2006; Ho et al., 2015). Our results also indicate that the protein-N inputs from the Gobi Desert are comparable to, or even exceed, the dry deposition of WSON in these regions. The recurring dust events and the increased Asian-supplied protein-N during spring, coinciding with the algae bloom season, may have widespread effects on the productivity of downwind ecosystems, influenced by the East Asian dust belt.

Second, during long-range transport, hydrophilic protein particles mix with mineral particles to form various internal mixtures. These mixed particles can absorb water efficiently, thereby increasing CCN activity and altering optical properties, which is important for understanding global climate and hydrological cycles (Adachi et al., 2020).

Third, aerosol proteins increase might affect the generation of secondary aerosols (Li et al., 2020). Under high relative humidity condition, water might condense onto protein aerosols due to their good hygroscopicity, resulting in forming a film of water on the aerosol surface. This water film is easy to adsorb gaseous pollutants in the atmosphere, including SO₂, NOx, O₃ and VOCs. The oxidation rate of these gaseous pollutants in the liquid-solid heterogeneous system is much faster than that in gaseous phase. Thus, water film on the protein surface may accelerate the oxidation of the absorbed gaseous pollutants in the atmosphere.

Thank you very much for your suggestions. These revisions have provided us with a more accurate understanding of the factors and mechanisms influencing protein concentrations in the atmospheres of downwind cities during dust transport, which has enhanced the environmental implications of this study.

Q2: The sampling period (March 24–31, 2018) represents a short window of time. While the authors acknowledge the occurrence of dust storms during this period, it is unclear if these results are representative of long-term trends or typical dust events. A broader period could strengthen the conclusions, or could the authors provide additional context on whether this period represents typical dust activity for that year or region.

Answer: Thank you for your suggestion. Globally, the primary sources of mineral dust are the arid regions of North Africa, the Arabian Peninsula, Central Asia, and Northeast Asia (Filonchyk, 2022;

Zhou et al., 2019). East Asia is the second-largest dust source region worldwide, with annual dust emissions estimated at 214 Tg yr⁻¹ (Tian et al., 2020). Furthermore, the westerlies in the Northern Hemisphere's middle latitudes can transport Asian dust from upwind areas to distant downwind regions, potentially completing a global cycle and exerting far-reaching impacts (Xie et al., 2023; Zhou et al., 2019). Asian dust particles primarily originate from the arid and semi-arid areas of northwestern China and Mongolia, including the Taklamakan Desert, the Gobi Desert, the Badan Jaran Desert, the Tengger Desert, and others (Shao and Dong, 2006). Recent research indicates that the Gobi Desert, rather than the Taklamakan Desert, is the primary contributor to dust concentrations in East Asia in spring (Chen et al., 2017; Tang et al., 2018). Spring is considered the peak season for sand and dust storms in Northeast Asia, as positive surface pressure anomalies over the Tamil Peninsula intensify cold air outbreaks across the desert regions of northwestern China and Mongolia (Yang et al., 2008). A particularly intense and widespread dust event occurred between March 26–29, 2018, in the North China Plain, which was the most significant dust storm in recent years in China (Zhou et al., 2019). This dust event affected nearly two-thirds of China and parts of the Northwest Pacific (Tian et al., 2020). Therefore, PM_{2.5} sampling at four representative sites (Beijing, Tianjin, Shijiazhuang, and Taiyuan) located in the downwind areas of the Gobi Desert during the 26–29 March 2018 dust event provides a typical representation of Asin dust activity in northern China.

The content has been incorporated into the introduction section of the revised manuscript. Line 83~99.

Q3: The use of satellite imagery and back-trajectory analysis to confirm Gobi dust as the source is an important strength of the paper. However, additional validation through ground-based measurements or comparison with other dust episodes would confirm that the identified CAA increases were directly attributable to Gobi dust.

Answer: Thank you for your suggestion. Sorry for our unclear description. In this study, the conclusion that the identified CAA concentrations in the North China Plain increase during the dust period and are directly attributable to Gobi dust is based on ground-based observations. Satellite imagery and back-trajectory are auxiliary tools in this study. Surface soil, vegetation, and PM2.5 samples were collected from both the North China Plain and the Gobi Desert source area. The concentrations and δ^{15} N values of individual CAAs in these samples were measured. By comparing the concentrations, percentage compositions, and nitrogen isotopes of individual CAAs between dust and non-dust periods, we found that CAAs transported by Gobi dust were rich in alanine, glycine, and glutamic acid. The concentrations and percentages of these three CAAs in PM2.5 from Northern China notably increased during dust periods. From the non-dust to dust periods, glycine and leucine in urban PM2.5 exhibited negative shifts in their δ^{15} N values, confirming that Gobi dust is a significant source of CAAs in PM2.5 in Northern China. Thus, this study represents a field-based investigation. We analyzed the variation in the percentage composition and nitrogen isotopes of individual CAAs in PM2.5 samples from the non-dust to dust periods, which confirmed the contribution of Gobi dust to the increased CAA concentrations in downwind areas.

Additionally, we reviewed literature on ground-based dust observations in the North China Plain during our sampling period. Tian et al. (2020) also confirmed that an intensive dust event occurred in the North China Plain from 26–29 March 2018 originating from western Inner Mongolia. Line 281~282.

Q4: The authors use a fixed dry deposition velocity (Vd) based on previous studies. However, Vd is typically influenced by factors such as particle size, wind speed, and hygroscopicity. The use of a fixed value introduces uncertainty in the estimation of deposition fluxes. A site-specific Vd estimate would improve the accuracy of their calculations.

Answer: Thank you for your suggestion. Indeed, the use of a fixed value introduces uncertainty in the estimation of deposition fluxes. However, since we did not obtain data on the size distribution of CAAs, site-specific Vd cannot be obtained. In future research, we will further study the particle size distribution characteristics of atmospheric proteins to more accurately calculate the atmospheric protein-N deposition flux. In the revised manuscript, we have provided the uncertainty in protein deposition flux caused by the uncertainty of dry deposition velocity of protein-N (Figure 8).

Primary biological aerosols have been found to be distributed from nanometers up to about a tenth of a millimeter, with their size distribution influenced by their sources (Fröhlich-Nowoisky et al., 2016). Therefore, in this study, the deposition velocities of protein-N were assumed to be the same as those used to estimate water-soluble nitrogen dry deposition (0.012 m s⁻¹), given that protein-N is a significant component of water-soluble organic nitrogen (WSON) in aerosols and WSON has also been detected in both coarse and fine fractions (Zamora et al., 2011; Zhang and Anastasio, 2003). The uncertainty in the value for the dry deposition velocity can lead to the uncertainty in dry flux estimates. For particles in the size range where gravitational setting is the controlling factor, Vd values obtained by model and field experiment were consistent (Spokes et al., 2000). (Duce et al., 1991) reported that under wind speeds ranging from 0 to 13 m s⁻¹ and relative humidity between 0% and 100%, the deposition velocity for submicrometer aerosol particles is 0.1 cm s-1 \pm a factor of 3, while the deposition velocity for supermicrometer crustal particles is 1 cm s-1, also with an uncertainty factor of 3. During the dust period, the wind speeds in Beijing, Tianjin, Shijiazhuang, and Taiyuan were $2.0 \sim 2.4 \text{ m s}^{-1}$, $4.1 \sim 6.1 \text{ m s}^{-1}$, $1.7 \sim 2.6 \text{ m s}^{-1}$ and $4.6 \sim 7.0 \text{ m s}^{-1}$, respectively, and the relative humidity were 25.8~28.4%, 24.3~28.0%, 24.3~29.1%, and 37.0~38.5%, respectively. The variations in wind speed and relative humidity across the four sampling cities were relative minor, and their ranges fell within those reported by (Duce et al., 1991).

Based on this, the uncertainty for the deposition velocity of aerosol protein-N in this study was set to a factor of 3. Line 198~216. Using the formula for error propagation for multiplication:

$$\left(\frac{\Delta F dry}{F dry}\right)^2 = \left(\frac{\Delta C}{C}\right)^2 + \left(\frac{\Delta V d}{V d}\right)^2$$

Since the concentration C is measured with negligible uncertainty (0.1) compared to Vd, the term $\Delta C/C$ can be considered zero. Therefore, the equation simplifies to:

$$\frac{\Delta F dry}{F dry} = \frac{\Delta V d}{V d}$$

Given $\triangle Vd/Vd = 3$:

$$\frac{\Delta F dry}{F dry} = 3$$

Consequently, Fdry will also have the same factor of uncertainty:

$$F_{dry,min} = C \cdot \frac{Vd}{3}$$

Each deposition flux value of protein-N with the uncertainty range at the four sampling cities was provided in the revised manuscript (Figure 8).

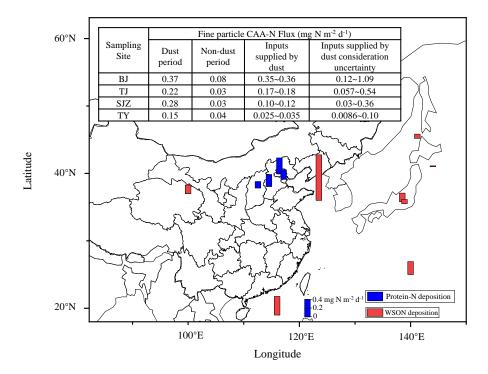


Figure 8. The dry deposition flux of protein-N at BJ, TJ, SJZ and TY during the dust period (blue bar) and published reports of the dry deposition flux of WSON measured in different atmospheric scenarios (red bar). The length of the bars represents the dry deposition flux of WSON and protein-N. The WSON deposition data were sourced from previous studies (Shi et al., 2010; Ho et al., 2015; Matsumoto et al., 2014; Tsagkaraki et al., 2021; Nakamura et al., 2006; Zhang et al., 2011). The method of calculating deposition fluxes of protein-N with the uncertainty range at the four sampling sites was provided in the supplementary manuscript (Text 1).

Q5: The paper focuses heavily on the Gobi dust contribution but does not extensively explore the potential influence of local urban sources of CAAs. It would be helpful to discuss how industrial, vehicular, or other anthropogenic emissions contribute to the CAAs in $PM_{2.5}$. This would provide a clearer differentiation between dust and urban source contributions. Further justification or consideration of urban $\delta 15N$ variability is needed.

Answer: Thank you for your suggestion. We have added more discussion focused on the local urban sources contributing to the CAAs in aerosols in the revised manuscript. A new section, "4.1 Local Urban Sources of CAAs in PM_{2.5}," has been added to discuss these sources. The concentration of CAAs measured during the non-dust period was compared with previous studies. The δ^{15} N values of individual CAAs in PM_{2.5} from four cities were compared to their respective values in local common plants, soil, and burning sources across the four urban sites to further identify the local

sources of CAAs in PM_{2.5}. Additionally, we compared our results of protein source apportionment with previous reports, finding no discrepancies. The results confirmed that local dominant plants, surface road dust, and anthropogenic industrial activities involving combustion processes were the major sources of CAAs in PM_{2.5} across Beijing, Tianjin, Shijiazhuang, and Taiyuan during the nondust period.

Please refer to Line 454~484.

Minor Comments

Q6: It is suggested that the sampling locations be visually represented on a map and be incorporated into Fig 8 as a subplot. This will provide a clearer spatial understanding of the study's geographic scope.

Answer: Thank you for your suggestion. The reviewer 2 also made a similar suggestion. A map showing the locations of the collected samples in the Gobi Desert and each city has been added in Supplementary Material (Figure S1).

Q7: The terms dust storm and dust event are used interchangeably throughout the manuscript, but they can be interpreted as two different transport mechanisms in the dust transport field. The author should consistently use one or the

Answer: Thank you for your suggestion. The term "dust storm" has been revised to "Asian dust event" throughout the revised manuscript.

Reviewer 2

Q8: The main limitation of the work in my opinion is about sample representativeness. Line 110: how many soil and plant samples from Gobe were analyzed? Was there any difference in the results from soil samples at different depths within the 0-10 cm? What area of the desert were the samples collected from? Only one latitude and longitude in indicated in the text. How representative are the samples? Similarly, how many local samp. A table summarizing, number of different samples, average and standard deviation of the values determined in these samples, and a map showing location of the collected samples in Gobe and each city are needed (in SI). How confident can we be because of this limitation in the estimated fraction of proteinaceous PM2.5 originating from Gobe Desert?

Answer: Thank you for your suggestion. A table summarizing, number of different samples, average and standard deviation of the values determined in these samples (Table S1) and a map showing location of the collected samples in Gobi and each city (Figure S1) have been included in the supplementary materials. Surface soil samples from the Gobi Desert were collected from an area ranging between 43.46°N to 43.60°N and 112.00°E to 112.05°E, covering approximately 45 km², as shown in Figure S1. The Gobi Desert is the major source of sand for dust storms in Asia during the spring (An et al., 2013). Therefore, five sampling sites along the transport pathway of the dust event that occurred from March 26 to 29, 2018 (Figure 2), were selected to represent the Erenhot Gobi Desert. Each site was free of anthropogenic interference. Surface sand samples, which are most likely to be aerosolized, were collected from the tops of dunes using a plastic spatula and

stored in sealed plastic bags until transported to the laboratory. At each location, surface soil was collected from five randomly selected sampling points within a radius of approximately 20 cm. These five sub-samples were then combined to create one representative sample. Line 129~137. Apologies for the confusion. By "0–10 cm," we refer to the surface soil samples that were collected. During dust events, the surface sand samples from the tops of dunes are the most likely to be aerosolized. Therefore, at all sampling locations, only surface soil was collected, following the method outlined by (An et al., 2013). Therefore, "0–10 cm," was changed to "the surface soil" in revised manuscript.

Five surface soil and twelve dominate plant samples were collected from the Gobi Desert (Table S1).

Q9: Line 1 of abstract: Particulate matter transported in dust storms can influence biogeochemical cycles of many elements and not just nitrogen so I suggest removing the reference to nitrogen in this introductory sentence.

Answer: Thank you for your suggestion. "nitrogen" was deleted in this sentence.

Q10: Line 40, I'm not sure how presence of primary particles from proteinaceous material can affect new particle formation. Can you please clarify?

Answer: Thank you for your suggestion. In a previous study, (Li et al., 2022) suggested that aerosol proteins might affect the generation of secondary aerosols. They proposed that under haze condition, the relative humidity increases and then high humidity facilitates water condensation onto protein aerosols due to their good hygroscopicity, resulting in forming a film of water on the aerosol surface. This water film is easy to adsorb gaseous pollutants in the atmosphere, including SO₂, NO_X, NH₃, O₃ and VOCs. It is well known that the oxidation rate of gaseous pollutants in the liquid-solid heterogeneous system is much faster than that of gaseous phase. The reaction of O₃ in wet particles generates strong oxidizing substances such as hydroxyl free radical (·OH), which accelerated the oxidation of the absorbed SO₂ and NO_X into SO₄²⁻ and NO₃⁻. During these processes, some parts of the protein core may be oxidized or nitrified protein through nitrification reaction (Liu et al., 2017; Shiraiwa et al., 2012).

Q11: Line 75, define GLY

Answer: Thank you for your suggestion. GLY (glycine) was defined in the revised manuscript.

Q12: Line 84: "...representative urban centers..."

Answer: Thank you for your suggestion. It has been revised as your suggestion.

Q13: Figure S1: The figure lacks geographical references (i.e., borders, city markers with legends, etc) to guide the reader to the relative location of dust sources in Gobe and receptor sites. Also, please add the color scale.

Answer: Thank you for your suggestion. Geographical references including borders, city markers with legends and the color scale have been added.

Q14: Line 106: remove "1 from"

Q15: Section 2.2: details on extraction efficiency of the developed methods need to be discussed Answer: Thank you for your suggestion. The extraction efficiency of the method was added in the supplement material.

Answer: Thank you for your suggestion. To evaluate the extraction efficiency, analytical method was applied to the samples were spiked with the amino acid standard mixtures ($100\mu l\ lnmol\ \mu l^{-1}$). The average recovery ratios of the individual amino acids were shown in Table S2. The recoveries for the majority of CAAs ranged from 80.7% (tyrosine) to 106.5% (glycine). The precisions of the investigated AAs were better than 10%. Line $170\sim173$.

Table S2. The recoveries and precision of CAA analysis.

Amino acids	Recovery (%)	Precision (%)
Glycine (Gly)	106.5	1.1
Alanine (Ala)	92.6	3.6
Aspartic acid (Asp)	95.0	8.6
Glutamic acid (Glu)	105.6	8.8
γ-amino butyric acid (Gaba)	95.7	7.7
Serine (Ser)	92.7	5.7
Proline (Pro)	93.2	6.7
Threonine (Thr)	90.0	6.1
Valine (Val)	95.5	1.3
Lysine (Lys)	82.5	6
Leucine (Leu)	92.0	0.8
Isoleucine (Ile)	93.1	2
Arginine (Arg)	85.4	9.4
Phenylalanine (Phe)	93.7	5.4
Ornithine (Orn)	91.5	9.9
Tyrosine (Tyr)	80.7	0.3
Histidine (His)	95.1	1.7
Methionine (Met)	86.6	2.7

Q16: Line 159: This sentence is not clear to me. Based on the previous sentence, I thought concentration of asparagine and glutamine cannot be determined, but total concentration of asparagine+ aspartic acid and glutamine+ glutamic acid can be. Is that not the case?

Answer: Sorry for our mistake. The concentration of aspartic acid represents total concentration of asparagine+ aspartic acid and the concentration of glutamic acid represents glutamine+ glutamic acid. It was changed to "Since asparagine and glutamine are converted to aspartic acid and glutamic acid in the hydrolysis process, respectively, the concentration and $\delta^{15}N$ value of combined aspartic acid represents the sum of aspartic acid and asparagine. The concentration and $\delta^{15}N$ value of combined glutamic acid represents the sum of glutamic acid and glutamine." Line 186-187.

Q17: Line 169: As you mention, deposition velocity for particles is size dependent. What ranges of Vd is expected for the larger sizes of fine aerosols that are the focus of this paper? How much uncertainty would this bring to the estimates of deposition fluxes calculated for the different cities?

Answer: Thank you for your suggestion. The uncertainty in the value for the dry deposition velocity can lead to the uncertainty in dry flux estimates. For particles in the size range where gravitational setting is the controlling factor, Vd values obtained by model and field experiment were consistent (Spokes et al., 2000). (Duce et al., 1991) reported that under wind speeds ranging from 0 to 13 m s⁻¹ and relative humidity between 0% and 100%, the deposition velocity for submicrometer aerosol particles is 0.1 cm s⁻¹ ± a factor of 3, while the deposition velocity for supermicrometer crustal particles is 1 cm s⁻¹, also with an uncertainty factor of 3. During the dust period, the wind speeds in Beijing, Tianjin, Shijiazhuang, and Taiyuan were 2.0~2.4 m s⁻¹, 4.1~6.1 m s⁻¹, 1.7~2.6 m s⁻¹ and 4.6~7.0 m s⁻¹, respectively, and the relative humidity were 25.8~28.4%, 24.3~28.0%, 24.3~29.1%, and 37.0~38.5%, respectively. The variations in wind speed and relative humidity across the four sampling cities were relative minor, and their ranges fell within those reported by (Duce et al., 1991). Based on this, the uncertainty for the deposition velocity of aerosol protein-N in this study was set to a factor of 3. Therefore, Vd ranged from 0.004 m/s to 0.036 m/s. Since Fdry=C·Vd, the formula was used for error propagation for multiplication:

$$\left(\frac{\Delta F dry}{F dry}\right)^2 = \left(\frac{\Delta C}{C}\right)^2 + \left(\frac{\Delta V d}{V d}\right)^2$$

Since the concentration C is measured with negligible uncertainty (0.1) compared to Vd, the term $\Delta C/C$ can be considered zero. Therefore, the equation simplifies to:

$$\frac{\Delta F dry}{F dry} = \frac{\Delta V d}{V d}$$

Given $\triangle Vd/Vd = 3$:

$$\frac{\Delta F dry}{F dry} = 3$$

Consequently, Fdry will also have the same factor of uncertainty:

$$F_{dry,min} = C \cdot \frac{Vd}{3}$$
$$F_{dry,max} = C \cdot 3 \cdot Vd$$

Each deposition flux value of protein-N with the uncertainty range at the four sampling cities was provided in the revised manuscript (Figure 8). Line 198~216.

Q18: Figure 1. Are the indicate date stamps indicating midnight or noon? Please clarify in the caption. Either way, it doesn't look like the peak in PM10 in Shijiazhuang occurred 11:00 to 18:00.

Answer: Thank you for your suggestion. The timestamps indicate 21:00. This was added in the caption of figure 1. With the consideration of the timestamps is 21:00, the peak in PM10 in Shijiazhuang occurred 11:00 to 18:00.

Q19: Line 239: I believe the reference here should be to Figure 2

Answer: Sorry for our mistake. It has been revised as your suggestion.

Q20: Line 261: consider changing 'increment' to 'increase'

Answer: Thank you for your suggestion. It was revised to "increase".

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

Yours sincerely,

Ren-Guo Zhu, Hua-Yun Xiao, Meiju Yin, Hao Xiao, Zhongkui Zhou, Yuanyuan Pan, Guo Wei, Cheng Liu

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