## Spatial source contribution and interannual variation in deposition of dust aerosols over the Chinese Loess Plateau

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November 27, 2023

### Abstract

The Chinese Loess Plateau (CLP) in northern China serves one of the most prominent loess records in the world. The CLP is an extensive record of changes in past aeolian dust activity in East Asia; however, the interpretation of the loess records is hampered by ambiguity regarding the origin of loess-forming dust and an incomplete understanding of the circulation forcing dust accumulation. In this study, we used a novel modeling approach combining a dust emission model FLEXDUST with simulated back trajectories from FLEXPART to trace the dust back to where it was emitted. Over 21 years (1999-2019), we modeled back trajectories for fine ( $\tilde{2}$  2mu) and super-coarse ( $\tilde{2}$  20mu) dust particles at six CLP sites during the peak dust storm season from March to May. The source receptor relationship from FLEXPART is combined with the dust emission inventory from FLEXDUST to create site-dependent high-resolution maps of the source contribution of deposited dust. The nearby dust-emission areas dominate the source contribution at all sites. Wet deposition is important for dust deposition at all sites, regardless of dust size. Non-negligible amounts of dust from distant emission regions could be wet deposited on the CLP following high-level tropospheric transport, with the super-coarse dust preferentially from emission areas upwind of sloping topography. On an interannual scale, the phase of the Arctic Oscillation (AO) in winter was found to have a strong impact on the deposition rate on the CLP, while the strength of the East Asian Winter Monsoon was less influential.

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16	•	The source contribution of emitted dust aerosols to the Chinese Loess Plateau
17		(CLP) is mapped using backward trajectory modeling.
18	•	Distant dust emission regions in Central Asia have a non-negligible impact on
19		the CLP, with wet-deposited dust showing a clearer connection.
20	•	Strong dust deposition years over the CLP are associated with the negative

phase of the Arctic Oscillation.

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### 22 Abstract

The Chinese Loess Plateau (CLP) in northern China serves one of the most promi-23 nent loess records in the world. The CLP is an extensive record of changes in past 24 aeolian dust activity in East Asia; however, the interpretation of the loess records is hampered by ambiguity regarding the origin of loess-forming dust and an incomplete understanding of the circulation forcing dust accumulation. In this study, we 27 used a novel modeling approach combining a dust emission model FLEXDUST with 28 simulated back trajectories from FLEXPART to trace the dust back to where it was 29 emitted. Over 21 years (1999-2019), we modeled back trajectories for fine (~ 2  $\mu$ m) 30 and super-coarse ( $\sim 20 \ \mu m$ ) dust particles at six CLP sites during the peak dust 31 storm season from March to May. The source receptor relationship from FLEX-32 PART is combined with the dust emission inventory from FLEXDUST to create 33 site-dependent high-resolution maps of the source contribution of deposited dust. 34 The nearby dust-emission areas dominate the source contribution at all sites. Wet 35 deposition is important for dust deposition at all sites, regardless of dust size. Non-36 negligible amounts of dust from distant emission regions could be wet deposited 37 on the CLP following high-level tropospheric transport, with the super-coarse dust 38 preferentially from emission areas upwind of sloping topography. On an interan-39 nual scale, the phase of the Arctic Oscillation (AO) in winter was found to have 40 a strong impact on the deposition rate on the CLP, while the strength of the East 41 Asian Winter Monsoon was less influential. 42

### 43 Plain Language Summary

The dust deposits on the Chinese Loess Plateau (CLP) in northern China pro-44 vide an extensive record of changes in past dust activity in East Asia. In this work, 45 we combine an atmosphere transport model with a dust emission model to trace the 46 dust backward to where it was emitted. Choosing six locations at the CLP where we 47 trace the dust back to emissions over a 21-year period during the most active dust 48 months from March until May, for which we investigated how different parts of the 49 CLP were influenced by the different dust emission areas, transport mechanisms, 50 and wind circulation patterns. We find that dust is mainly brought from the emis-51 sions areas located near the CLP, still dust from far-away emissions areas in Cen-52 tral Asia has the potential to reach the CLP in smaller amounts. Furthermore, our 53 results show that we removal of dust by precipitation is important for dust deposi-54 tion on the CLP, a mechanism that is often overlooked when explaining variations 55 in past dust activity. On a year-to-year basis the dust deposition rate over the CLP 56 was found to be connected to the high-latitude northern hemisphere's climate oscil-57 lation (Arctic Oscillation) rather than the regional East Asian winter monsoon. 58

### 59 1 Introduction

Mineral dust is deeply embedded within the Earth system, influencing the 60 planet's energy balance, cloud characteristics, atmospheric chemistry, and biogeo-61 chemistry (Shao et al., 2011; Kok et al., 2023). Dust is mainly emitted from dry 62 and sparsely vegetated arid and semi-arid regions. Emission is driven by surface 63 winds that lift sand grains from the soil bed, where the impact upon reentry ejects 64 and fragments smaller soil particles, which then get transported. The dust-emitting 65 capacity of the soil is strongly influenced by environmental factors such as soil mois-66 ture, vegetation, soil texture, and the structure of the top soil. On geological time 67 scales, dust depositional records show increases in dust activity during glacial periods (Porter, 2001; Maher, 2016). However, as human expansion has increased the in-69 tensification of land use in recent years, we are now seeing a profound impact of hu-70 man activity on dust activity, e.g. the drying of Aral Sea and dust bowl in the 1930s 71

North America. The antropogenic signal is also evident in the depositional records,
which show that in the last 150 years there has been an increase in the dust burden
globally of about 50% (Hooper & Marx, 2018). However, we do not know to what
extent this is due to a forcing caused by increased human activity and changes in
land use in semi-arid regions or to modifications in precipitation and wind patterns
in a changing climate (Kok et al., 2023).

Changes in dust activity are important because of the climate effect of dust. 78 Fine dust aerosols exhorts cooling effect due to its ability to reflect incoming sun-79 light, whereas coarse dust aerosols cause a warming effect as they interact with the outgoing long-wave radiation. Furthermore, different dust-emitting regions have dif-81 ferent dust mineralogy, which affects its ability to absorb radiation (Dubovik et al., 82 2002). Atmospheric dust particles also affect clouds by acting as nuclei for conden-83 sation, contributing to droplet formation. Additionally, dust particles act as nuclei 84 to initiate glaciation of clouds above the temperatures of homogeneous freezing. Ice 85 clouds generally have a warming effect, whereas liquid clouds have a cooling effect 86 (Storelymo, 2017). The deposition of dust on snow accelerates snow melt (Wittmann et al., 2017; Sarangi et al., 2020), while dust input can increase ecosystem produc-88 tivity in regions lacking nutrients, by providing iron or phosphorus (Martin, 1990; 89 Yu et al., 2015). These processes impact the climate system, but the complexities 90 introduced by the poor constraints of, for example, dust particle size (A. Adebiyi et 91 al., 2023; Ryder et al., 2018), shape (Huang et al., 2020), mineralological composi-92 tion (Li et al., 2021) and dust-cloud interactions (Sagoo & Storelvmo, 2017) of air-93 borne dust particles, make the overall climatic effect of the dust uncertain (Kok et 94 al., 2023).

One of the most prominent dust records on Earth for studying environmental drivers of changes in the dust cycle is the Chinese Loess Plateau (CLP), located in the north central part of China (Figure 1). Here windblown dust has been accumulating since at least the late Oligocene-early Miocene (c. 25-22 Ma) (Guo et al., 2002; Qiang et al., 2011), providing a long-term, nearly continuous record of past responses of the East Asian Dust Cycle (EADC) to environmental and climatic changes (e.g. Stevens et al., 2007; Lu et al., 2010; Maher, 2016; Y. Sun et al., 2020).

The sediment sources, dust-emitting regions, transportation, and deposition 103 processes of the eolian deposits on the CLP are debated (e.g. W. Peng et al., 2023; H. Zhang et al., 2022; Shang et al., 2016; Bird et al., 2015; D. Sun et al., 2008). 105 However, most agree that the dust was generally sourced from the dry Asian inte-106 rior and proximal deserts via energetic, surface-level northwesterly winds driven by 107 the East Asian Winter Monsoon (EAWM) system (Z. Ding et al., 1999). In addition 108 to the EAWM, the prevailing mid-latitude westerlies in the middle and upper tro-109 posphere play an important role in transporting dust to the CLP. The EAWM and 110 the westerlies are suggested to explain the bimodal particle size distribution widely observed in the CLP records. The coarse particles are suggested to be transported 112 by the strong surface winds associated with the EAWM and the fine particles are 113 suggested to represent the background dust component of the CLP transported by 114 the high-level westerly air flow (Miao et al., 2004; D. Sun, 2004; D. Sun et al., 2008). 115 Furthermore, both the particle size distribution and the dust accumulation of the 116 loess records exhibit a northwest-southeast gradient (Z. Ding et al., 1999), which fur-117 ther supports the northwesterly and westerly wind driven transport pathways. 118

Recent studies using single grain provenance indicators on the CLP dust records shed more light on dust emission areas and transport pathways, and their variation in the geologic past (Bird et al., 2015; Shang et al., 2016; Nie et al., 2018; H. Zhang et al., 2021, 2022; Bohm et al., 2023; W. Peng et al., 2023). However, the interpretation of the CLP provenance records is hampered by the complexity of the source-tosink system and by the fact that dust provenance signals reflect not only the trans-

porting wind systems, but also the various processes that affect dust availability in 125

the source regions. Furthermore, single grain provenance ultimately tells us what 126 the source rocks and sediments are, which might be different from the dust-emitting

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regions. 128

Several provenance studies have reported spatial provenance differences across 129 the CLP for both Neogene Red Clay and Quaternary loss (e.g. Bird et al., 2015; 130 Shang et al., 2016; H. Zhang et al., 2021; W. Peng et al., 2023). However, the cause 131 of the spatial differences in provenance is not well established. The Yellow River 132 has been argued to cause these spatial differences by bringing material from the 133 Northern Tibetan Plateau closer to the CLP before the final aeolian transport step 134 (Stevens et al., 2013; Nie et al., 2015). Contrasting eolian dust transport pathways 135 for different parts of the CLP have also been proposed to add to the spatial dif-136 ference (Shang et al., 2016), with the northern and northeastern parts of the CLP 137 dominated by dust from the northwestern deserts via the EAWM, and the south-138 ern and southwestern parts of the CLP dominated by dust from the Northern Ti-1 20 betan Plateau and even the remote Taklamakan desert via the westerlies (Shang et al., 2016). 141

Applications of the provenance method to further unravel the dust cycle over 142 the CLP is also hampered by the fact that most single grain provenance approaches 143 are based on a relatively large particle size (>  $20 \ \mu m$ ). Our understanding of the transport of such large dust particles in the atmosphere is still limited and highly debated (A. Adebiyi et al., 2023). Super-coarse dust particles (sensu A. Adebiyi et 146 al. (2023),  $> 20\mu m$ ) have commonly been assumed to be transported only a short 147 distance (1-100 km) from the source region because of the high gravitational settling 148 velocity (Tsoar & Pye, 1987), which contradicts CLP provenance inferences of dust 149 derived from remote deserts, such as the Taklamakan. Furthermore, recent observa-150 tions have documented the possibility of long-distance transportation (> 1000 km) of 151 giant (  $> 62.5\mu m$ ) dust particles (Van Der Does et al., 2018; Varga et al., 2021). A better understanding of large dust particle transportation to the CLP will help the 153 interpretation of dust provenance proxies, and thus improve our understanding of 154 the EADC on geological time scales. 155

Dust models have been widely applied to understand the EADC, however, 156 there have been only limited cases where dust models have been applied to understand the formation of the CLP dust records in particular (Shi & Liu, 2011). More-158 over, previous modeling studies of past and present EADC have typically applied 159 global or regional climate models (Shi & Liu, 2011; S. L. Gong et al., 2006), which 160 mainly consider particles within the fine to coarse range (  $< 10\mu$ m) (Zhao et al., 161 2022). These models are also unable to determine the exact sources of emitted dust 162 for different locations at the CLP, which would greatly enhance the provenance in-163 terpretations of the CLP dust records. To address these issues, we used the Lagrangian particle dispersion model FLEXPART. We run FLEXPART backward in a 165 receptor-oriented viewpoint, which, paired with a dust emission field, allows us to es-166 tablish high-resolution source contribution maps. We apply FLEXPART to establish 167 source receptor relationships between six loess sites across the CLP and examine the 168 interannual variations in deposition at these over a 20-year period from 1999-2019. 169 In this study, both fine (1.7-2.5  $\mu$ m) and super-coarse (15-20  $\mu$ m) dust particles are 170 simulated to better understand whether the deposition of fine particles occurs under 171 the same climate conditions as that of coarse particles and whether the source re-172 gions are the same. We chose to study the interannual variation in the dust deposi-173 tion rate over the CLP because we are interested in understanding the processes re-174 sponsible for the changes in loess MAR, with implications for identifying the factors 175 that could be driving the changes that have occurred on longer time scales (F. Peng 176 et al., 2022). 177

### <sup>178</sup> 2 Model and Data

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### 2.1 FLEXDUST and FLEXPART

FLEXDUST is an offline dust emission model developed to be used in pair 180 with the Lagrangian dispersion model FLEXPART to study the long-range trans-181 port and deposition of windblown dust (Groot Zwaaftink et al., 2016). The combination FLEXDUST-FLEXPART has previously been applied to study the sources, 183 transport, and deposition of high-latitude dust (Groot Zwaaftink et al., 2017; Kylling 184 et al., 2018; Wittmann et al., 2017). The dust emitted in FLEXDUST is assumed 185 to have a volume size distribution between 0.2 and 20  $\mu m$  according to the brittle fragmentation theory described in Kok (2011). Tang et al. (2023) identified sev-187 eral weaknesses in the way dust-emitting areas in East Asia were represented in 188 the original FLEXDUST version. Therefore, we use the updated version of FLEX-DUST by Tang et al. (2023), which includes several changes to better represent present day dust emission areas in East Asia. To avoid confusion with the stan-191 dard version of FLEXDUST, we refer to the updated Tang et al. (2023) version of 192 FLEXDUST as FLEXDUST-EA. Changes in FLEXDUST-EA compared to FLEX-193 DUST include: (1) the parameterization of vertical dust flux described by Kok et 194 al. (2014) with a modified parameter for the calculation of the threshold friction ve-195 locity according to Shao and Lu (2000), (2) the soil texture dataset in the ISRIC 196 SoilGrids dataset described by de Sousa et al. (2020), (3) new improved topographical erodibility scaling, and (4) herbaceous and crop land cover types in the GLC-NMO land cover dataset are considered to have a potential to emit dust in addition 199 to bare ground and sparse vegetation. For a complete description of the changes in 200 FLEXDUST-EA compared to the standard version, see Tang et al. (2023). 201

Dust transport and deposition are calculated using FLEXPART version 10.4 202 (Pisso et al., 2019). FLEXPART is an open source model and has been applied to 203 study the transport of a wide range of atmospheric tracers, such as, mineral dust, 204 black carbon, and volcanic ash (Groot Zwaaftink et al., 2022, 2016; Choi et al., 2020; 205 Eckhardt et al., 2008). FLEXPART calculates the trajectories of a large number 206 of computational particles that are used to describe the transport and deposition 207 of tracers in the atmosphere. This version of FLEXPART includes the improved 208 wet deposition scheme based on cloud information from the ECWMF input fields (Grythe et al., 2017). In addition, FLEXPART accounts for gravitational settling, dry deposition, and in- and below-cloud scavenging of simulated dust aerosols. Dry 211 deposition is treated using a resistance scheme (Stohl et al., 2005) and in-cloud wet 212 deposition distinguishes between scavenging in the liquid and ice phase (Grythe et 213 al., 2017). 214

FLEXPART can be run both in forward (source-oriented) and backward (receptor-215 oriented) mode. The model gives the same results when running in forward or back-216 ward mode, except for some minor numerical differences (Seibert & Frank, 2004), 217 and therefore the direction can be set for computational efficiency. Since we are in-218 terested in mapping the spatiotemporal contribution of different dust-emitting re-219 gions to a receptor, we run FLEXPART in backward mode. In the backward con-220 figuration, computational particles are released at a predefined receptor location, 221 and then each particle is traced backward in time. The distribution of the residence time of the computational particles in each grid cell of the output grid is used to 223 calculate the emission sensitivity (ES). ES gives a relationship between a poten-224 tial source in a grid cell and deposition at the receptor. ES is calculated separately 225 for each dust particle size bin and deposition process. We include two size bins in 226 our FLEXPART simulations, corresponding to fine dust (mean diameter  $2\mu m$ ) and 227 super-coarse dust (mean diameter  $17.6\mu m$ ) following the terminology of A. Adebiyi 228 et al. (2023). These two sizes were selected to represent the clay and silt fractions that are typically observed in the Chinese loess. In addition to size, efficiencies for



Figure 1. Digital elevation model (Shuttle Radar Topography Mission) and the locations of the Chinese Loess Plateau, the study sites and major dust emitting areas in Central-East Asia. The distribution of loess, loess-like silt, and the undifferentiated sediments are from Börker et al. (2018), and the deserts and wind 477 eroded land in China from 1:200,000 Desert Distribution Dataset provided by the Environmental and Ecological Science Data Center 478 for West China, National Natural Science Foundation of China (http://westdc.westgis.ac.cn; last access 27 July 2020).

each of the wet removal processes can be assigned differently depending on the kind 231 of dust-aerosol represented. The parameters we used to represent the two types of 232 airborne dust particles in our FLEXPART simulations are listed in Table 1. Dry 233 deposition ES is calculated by releasing particles from the receptor at the surface level between 0-30 meters and then calculating the particle backtrajectories. Wet 235 deposition ES is calculated by releasing particles throughout the atmospheric col-236 umn to determine whether a particle is wet scavenged. Then only backtrajectories of 237 the particles that undergo wet scavenging are calculated. Consequently, to achieve 238 a statistically reliable outcome, a greater number of particles are necessary for wet 239 deposition simulations. ES averaged over a selected output layer height can be mul-240 tiplied by the dust emission inventory (units kg  $m^{-3}s^{-1}$ ) created by FLEXDUST, to 241 generate a map that quantifies each contribution of the source element to the depo-242 sition at the receptor (Eckhardt et al., 2017). Summing the source contribution (SC) 243 of each source element yields the deposition rate at the receptor. 244

### 245 2.2 FLEXPART backtrajectory set up and workflow



**Figure 2.** Flow chart showing the workflow for the FLEXPART-FLEXDUST backward trajectory modeling analysis.

Backward FLEXPART simulations were performed during the main dust storm season between March 1st and May 31st over a 21-year period from 1999 until 2019 for six locations across the CLP shown in Figure 1. The selected sites encompassed both well-established loess sites with historical loess and red-clay deposits (Baode, Luochuan, Lingtai, and Lantian ), as well as modern dust observation sites (Shapotou and SACOL). This diverse geographical selection of sites allows for a comprehensive assessment of dust deposition susceptibility across different parts of the CLP to different dust emissions regions and circulation patterns.

FLEXPART was configured the following way, every particle release contained 254 either 50,000 or 200,000 particles, depending on whether dry or wet deposition is 255 simulated. Particles within one release are evenly distributed over 3-hourly inter-256 vals. The extent of the backtrajectories was limited to a maximum particle age of 257 10 days. ES was calculated with 0.1 degree resolution, with an output grid spanning 258 50-128° longitude, 25-65° latitude, and stored as 3-hourly averages. The FLEXDUST-259 EA dust emission flux is multiplied by the averaged ES of 0-100 meters to derive the deposition source contribution. The parameters of the two particle size bins that we include are listed in Table 1. We have summarized the stages of the workflow in 262 Figure 2, commencing with the retrieval and preparation of the forcing data. Then 263 the calculations of dust emissions, dry and wet dust deposition ES are done concur-264 rently. Finally, the FLEXDUST dust emission inventory is combined with FLEX-265 PART ES during post-processing to produce the source contribution maps and the 266 deposition rate for each of the receptor locations. 267

	Description	Fine	Super-coarse
PCRAIN_AERO	Efficiency of below cloud scavenging by rain $(1=100\%, 0=\text{off})$	1.00	1.00
PCSNOW_AERO	Efficency of below cloud scavenging by snow $(1=100\%, 0=off)$	1.00	1.00
PCC_AERO	CCN efficiency (in cloud scavenging)	0.15	0.30
PIN_AERO	IN efficiency (in cloud scavenging)	0.02	0.10
PDENSITY	Particle density (dry deposition)	$2500.0{\rm kg}{\rm m}^{-3}$	$2500.0{\rm kg}{\rm m}^{-3}$
PDQUER	Mean particle diameter (dry deposition)	$2.057~\mu\mathrm{m}$	17.32 $\mu \mathrm{m}$
PDSIGMA	Normalized particle diameter deviation	1.21	1.15

 Table 1.
 FLEXPART species parameters for the fine and super-coarse dust particle size bins.

We used the most recent ECMWF Reanalysis v5 (ERA5) atmospheric forcing 268 dataset in our FLEXPART and FLEXDUST simulations, the dataset is described in 269 Hersbach et al. (2020). We used a regional domain cutout of ERA5 extending from 270 10°E to 160°E and 10°N to 80°N, with a horizontal resolution of 30 km (T639) and 271 137 vertical levels from the surface up to 0.01 hPa (80 km). ERA5 data were pre-272 pared and downloaded from the Copernicus Climate Change Serviced Climate Data 273 Store using the Flex extract version 7.0.4 Python package (Tipka et al., 2020). All 274 ERA5 data are available for free from the Copernicus Climate Data store. 275

### 276 2.3 Model evaluation

Long-term field observations of dust deposition on the CLP suitable for model
evaluation do not exist. Hence, we have to depend on aerosol reanalysis to assess
the performance of FLEXPART and FLEXDUST-EA. Specifically, we use ModernEra Retrospective Analysis for Research and Applications, version 2 (MERRA-2)
aerosol reanalysis produced by NASA's Global Modeling and Assimilation Office

(Gelaro et al., 2017). This dataset has previously been shown reliable at simulating 282 the EADC (H. Liu et al., 2018; W. Yao et al., 2020). Similarly to ERA5, MERRA-2 283 assimilates essential atmospheric variables such as wind, precipitation, and surface pressure. However, unlike ERA5, MERRA-2 additionally incorporates aerosol optical depth (AOD) retrievals sourced from various ground-based and spaceborne instruments, including the Moderate Resolution Imaging Spectroradiometer (MODIS), the 287 Multiangle Imaging Spectroradiometer (MISR), and the Aerosol Robotic Network 288 (AERONET). Assimilation should bring aerosol loading and AOD in MERRA-2 289 closer to observations. AOD represents the total aerosol loading but does not pro-290 vide information on aerosol composition, and therefore the model determines the 291 contribution of dust aerosols to the total AOD. MERRA-2 represent dust aerosols 202 using five size bins (0.1-1.0, 1.0-1.8, 1.8-3.0, 3.0-6.0, and 6.0-10.0  $[\mu m]$ ). Dust emis-293 sion and deposition are not assimilated in MERRA-2; and are in this regard simi-294 lar to FLEXDUST relying on the model physics. The wind patterns of MERRA-2 295 should be similar to those observed in ERA5, however, due to its lower spatial reso-296 lution of 0.675 °longitude and 0.5 °latitude, it is not able to capture local-scale winds 297 as accurately as ERA5. This means that MERRA-2 does not necessarily outperform 298 any other dust model (Gelaro et al., 2017; Zhao et al., 2022). For example, Tang et al. (2023) compared FLEXPART for the mega dust storm in spring 2021 with the two most widely used aerosol reanalysis products MERRA-2 and CAMS. The study 301 revealed that FLEXPART exhibited consistency with MERRA-2 with respect to the 302 strength and timing of the event. However, the overall spatio-temporal patterns of 303 total concentration were more in line with CAMS. We still chose to use MERRA-2 304 as our benchmark because it has a continuous data record dating back to 1980 and 305 covering the entire modeling period of this work. 306

### 307 3 Results and Discussion

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### 3.1 The spatial distribution and interannual variation of the East Asian spring dust cycle

Our setup using FLEXPART coupled with FLEXDUST-EA was developed 310 to represent East Asian sources and Tang et al. (2023) showed that it can realisti-311 cally simulate the evolution of single dust events, compared to both observations 312 and reanalysis. Here, we evaluated FLEXDUST-EA by comparing it with MERRA-313 2 over a longer time period. Zamora et al. (2022) have also shown a comparison 314 with MERRA-2 products focusing on the Arctic region, where they found mainly 315 differences based on a lack of high-latitude dust sources in MERRA-2. The spa-316 tial distribution of dust emissions in FLEXDUST-EA aligns closely with that of MERRA-2, as illustrated in Figure 3. Both FLEXDUST-EA and MERRA-2 identify 318 key dust emission hot spots in East Asia, including the Taklamakan desert, north-319 western deserts, and the Gobi desert, which is consistent with previous studies (Kok 320 et al., 2021; Shi & Liu, 2011; Shao & Dong, 2006; J. Sun et al., 2001). Moreover, 321 FLEXDUST-EA shows a strong correlation with MERRA-2 regarding the interan-322 nual variation in spring dust emission across all major source regions. Both models 323 show notably lower dust emissions during the springs of 2005 and 2019 and higher emissions during 2001 and 2004. In addition, the period between 1999 and 2009 has 325 greater variability in dust emissions compared to 2010 to 2019 in both FLEXDUST-326 EA and MERRA-2. 327

<sup>328</sup> Next, we evaluate how FLEXPART represents the interannual variation of dust <sup>329</sup> deposition at our sites by comparing the springtime dust deposition simulated by <sup>330</sup> FLEXPART with that of the MERRA-2 grid box closest to our sites. Figure 4a <sup>331</sup> shows the interannual variation of the total spring deposition simulated by FLEX-<sup>332</sup> PART versus MERRA-2, comparing the fine dust size bin (mean diameter 2  $\mu$ m) <sup>333</sup> in FLEXPART with the 1.0-1.8  $\mu$ m size bin in MERRA-2. The models correlate



Figure 3. Normalized mean spring dust emissions (a) FLEXDUST and (b) MERRA-2 between 1999 and 2019. The following dust-emitting regions are indicated: Taklamakan (blue, A), Qaidam Basin (purple, D), sources northwest of CLP (green, E,F,H), Mongolia (orange, G,I), Gurbantunggut Desert (brown, B) and Central Asia (pink), the letters correspond to dust emission regions named in Figure 1. (c) Normalized timeseries of spring dust emissions FLEXDUST (solid line) and MERRA-2 (dotted line), numbers in the upper right corners show the correlation coefficient. (d) The relative dust emission strength of each source region.



Figure 4. (a) Inter-annual variation of total spring deposition simulated by FLEXPART (blue) and MERRA-2 (orange) of the fine dust particle size bin (FLEXPART mean diameter 2.057  $\mu$ m,size bin 2 in MERRA-2, 1.0-1.8  $\mu$ m) for each of the sites at the CLP. The envelope indicates one standard deviation. (b) The ratio of wet deposition to total deposition at each receptor site. The blue bar is FLEXPART and orange bar is MERRA2.(c) Relative distribution of dust deposition between the receptor sites in FLEXPART and MERRA-2.

well for Luochuan, Lantian, and Shapotou, but FLEXPART and MERRA-2 are 334 not correlated at SACOL and Lingtai. SACOL and Lingtai are located close to the 335 mountains in a region with complex topography, which is not well resolved by typical model grids. Additionally, accurately representing precipitation in mountainous 337 regions poses a significant challenge, and different reanalysis datasets may have sub-338 stantial variation in their precipitation rates (J. Yao et al., 2020). Moreover, SACOL 339 and Lingtai experience a high proportion of wet deposition, as shown in Figure 4b, 340 making the differences in precipitation events between ERA5 and MERRA-2 a po-341 tential source of reduced correlation. There is a greater correlation in the dry depo-342 sition rate between MERRA-2 and FLEXPART for all sites (see Figure S1) than in 343 wet deposition. In addition, the wet deposition has a larger interannual variation in 344 FLEXPART compared to MERRA-2. Consequently, wet deposition amplify the differences between FLEXPART and MERRA-2 to total deposition. Finally, modeling 346 dust (wet) deposition is an overarching problem in current dust models, with models 347 showing large differences in the relative importance of wet to dry deposition (Zhao 348 et al., 2022; Huneeus et al., 2011). 349

FLEXPART and MERRA-2 also show differences in the spatial distribution 350 of dust deposition throughout the CLP (Figure 4c). MERRA-2 has a smaller spa-351 tial difference in dust deposition throughout the CLP, while in FLEXPART the two 352 sites close to the source regions make up more than 50% of the combined deposi-353 tion of our sites. Our set-up with backward trajectories limited to 10 days and the 354 comparison of relatively larger sized particles in FLEXPART to smaller particles in 355 MERRA-2 can both contribute to this model inconsistency, but it is also related to 356 the longer lifetime in MERRA-2 compared to FLEXPART (Tang et al., 2023). 357

Although conducting a detailed investigation of the differences in the repre-358 sentation of the EADC in FLEXPART and MERRA-2 is outside the scope of this 359 study. Still, our initial assessment has revealed that the interannual variation in dust 360 emissions in FLEXDUST-EA shown in Figure 3 aligns well with that of MERRA-2, particularly for the important source regions in East Asia. However, total dust de-362 position over the CLP in FLEXPART is not consistent with MERRA-2 at all sites, 363 mainly due to differences in the occurrence of wet deposition events. Evaluating the 364 representation of dust deposition in both MERRA-2 and FLEXPART poses chal-365 lenges due to the limited constraints available on dust deposition in East Asia and 366 the rudimentary nature of dust deposition parameterization in models (X. Zhang et 367 al., 2019).

369

# 3.2 Springtime dust transport to the Chinese Loess Plateau: wet versus dry deposition

Figure 5 shows the 20-year averaged spring source contribution maps for each site and both dust particle sizes. The source contribution is normalized by the mean contribution, enabling a relative comparison of the influence of different sources, independent of the deposition rate at the site. The pie chart represents the relative contribution of the source regions as defined in Figure 3 to the total deposition at the site.

Across all CLP locations, Figure 5 shows that the predominant source region that accounts for the majority of dust deposition is the deserts northwest of the CLP. Depending on the site, this source contributes between 48% and 78% of the fine dust and between 47% and 88% of the super-coarse dust. In particular, the source contribution of super-coarse dust in Figure 5 shows a higher contribution from nearby sources compared to fine dust. Fine dust shows a gradual transition from the primary dust sources to less influential ones as one moves away from the receptor location on the source contribution map. The relative source contributions



Figure 5. Averaged spring dust source contribution (1999-2019) of dry- and wet deposition scaled by the mean contribution. The pie chart shows the contribution from each of the source regions as defined in Figure 3.

of fine dust at our CLP locations shown in Figure 5 are consistently similar. For example, at Baode, Luochuan, Lingtai, and Lantian, the order of importance of the different source regions remains consistent: (1) Northwest deserts, (2) Taklamakan, (3) Mongolia, (4) Central Asia, (5) Qaidam Basin, and (6) Gurbantunggut desert. Even at SACOL and Shapotou, which have more distinct source contributions, the main difference lies in an increased contribution from the northwestern dust sources.

In contrast, the source composition of super-coarse dust displays more dis-391 tinct patterns at each location. For instance, super-coarse dust from Shapotou originates primarily from the nearby desert to the northwest of the CLP, with only a 393 minor contribution from Taklamakan. Baode experiences a substantial influx of 394 super-coarse dust from Mongolia, a source nearly absent at other sites. Another no-395 table aspect of super-coarse dust is the trend of increasing influence of dust from 396 the Qaidam Basin across the southeast transect of the CLP. Although the Qaidam 397 Basin contributes modestly to emissions, it becomes the second most influential 308 source at SACOL, Luochuan, Lingtai, and third at Lantian. Central Asia and the Gurbantunggut desert, although not primary sources of dust to the CLP, however, are not negligible at certain sites. For instance, Central Asia contributes to 6-7% of 401 the super-coarse dust deposited at Lingtai and Lantian, emphasizing how even larger 402 dust particles from distant dust emission regions can impact the CLP. 403

The difference between the dry and wet deposition source contribution is shown 404 in Figure 5. The source contributions of wet and dry deposition of fine dust are 405 mostly similar except for dry deposited dust being more influenced by the north-406 western sources, whereas wet deposition has a stronger influence from the more dis-407 tant sources. On the other hand, the source contribution of wet deposited super-408 coarse dust noticeably diverges from the sources of dry deposited dust. The wet 409 deposition source contribution displays a more noisy pattern, indicating less fre-410 quent occurrences of wet deposition for super-coarse dust. The larger variability of 411 the wet-deposited super-coarse dust translates into a more erratic spatial pattern 412 in the wet deposition source contribution compared to the relatively robust pattern 413 observed for dry deposition. 414

The source contribution maps presented in Figure 5 represent the cumulative 415 effects of emissions, transport, and deposition. To pick out the general dust trans-416 port routes to the CLP, we take the centroid trajectory of all the particles released within each 3-hourly particle release and combine them into one dust transporting 418 trajectory by calculating the center of mass of all the trajectories over the whole pe-419 riod weighted by the deposition rate modeled at the time of release. The dust trans-420 port trajectories for all sites and for dry and wet deposition are shown in Figure 6. 421 In the case of fine dust, the trajectories demonstrate minimal variation between re-422 ceptor locations, consistent with relatively uniform source contribution maps across 423 the different sites. Conversely, for super-coarse dust, the transport patterns of drydeposited dust closely resemble those of dry-deposited fine dust, primarily exhibit-425 ing a prevailing northwesterly transport (Figure 6 a, b). An exception is observed 426 for SACOL, displaying a more westerly transport, in alignment with the substantial 427 contribution from the Qaidam Basin. Wet deposition of super-coarse dust exhibits 428 significant variability in transport trajectories between receptors, contributing to the 429 site-dependent nature of super-coarse dust source contributions. A common trend 430 across all sites is that wet-deposited dust is generally influenced by southerly air-431 masses, resulting in overall westerly transport. Given that the source of moisture 432 originates from the south, this pattern is reasonable. Furthermore, the trajectories 433 show an increase in altitude as they approach the receptor, indicating that wet-434 deposited dust typically experiences uplift due to convection initiated by the typical 435 cold air advection observed during a dust event. 436



Figure 6. Weighted average centroid dust loading trajectories by the deposition rate at the receptor at the time of arrival. (a) and (b) are dry deposition dust loading trajectories for fine (mean diameter 2.057  $\mu$ m) and super-coarse dust (mean diameter 17.32  $\mu$ m) particles. (c) and (d) wet deposition dust loading trajectories for the fine and super-coarse dust respectively. The mean height is along the trajectory shown by the coloring and the red dots along the trajectories are equally spaced 12hours apart. The shading underneath show the mean dust emission rate.

Wind speed only changes the mass emitted for dust up to 20  $\mu$ m, but does not 437 change the size distribution at emissions (Kok, 2011). Therefore, the variations in 438 source contribution between fine and super-coarse dust primarily stem from differ-439 ences in their lifetimes. In FLEXPART, we changed two parameters that control 440 dust particle lifetime: the wet scavenging efficiency (assumed to be size-dependent) 441 and the particle size that governs the dry deposition velocity. The impact of differ-442 ent gravitational settling can be seen from the difference in the source contribution 443 between dry-deposited fine and super-coarse dust. To keep the super-coarse dust in 444 the air sufficiently long enough to reach the CLP, either the wind speed has to be 445 sufficiently strong such that the dust arrives before it falls out or the dust has to experience vertical lifting to counter the gravitational settling. 447

Topography has been proposed to increase vertical dust transport. Heisel et 448 al. (2021) utilized high-resolution large-eddy simulations to demonstrate that gen-449 tly sloping terrain can substantially amplify vertical dust transport. The source 450 contribution maps of wet-deposited super-coarse dust showed a distinct band of 451 higher contribution along the southern edge of the Taklamakan. Additionally, it is 452 important to observe that this band of intensified source contribution is less promi-453 nent in the source contribution maps for dry deposition, suggesting that this phe-454 nomenon cannot be solely attributed to heightened emissions along the southern 455 boundary. Moreover, similar to the Taklamakan, the Qaidam Basin, a source sur-456 rounded by sloping terrain, is also a substantial source of wet-deposited super-coarse 457 dust. Hence, potentially suggesting that when super-coarse dust ascends to higher 458 atmospheric levels, it becomes scavenged within the cloud and falls out with the precipitation. Particles of this size readily activate as Cloud Condensation Nuclei 460 (CCN) and rapidly grow to raindrop size. Consequently, a substantial amount of 461 coarse dust could potentially accelerate precipitation onset, mitigating the tendency 462

of continental air with high concentrations of CCN to produce numerous small cloud
droplets (Posselt & Lohmann, 2008).

In summary, there are two prominent pathways for dust transport. The first pathway exhibits a prevailing northwesterly direction, gathering dust primarily from the northwestern desert regions. This pattern is strongly associated with the dry deposition of dust on the CLP. Conversely, the second dust transport pathway, linked to wet deposition, follows a more westerly trajectory. Along this route, topographical features drive dust to higher atmospheric levels, where it is then scavenged within the clouds and subsequently washed out over the CLP.

### 3.3 Drivers of interannual variations in EADC

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Indices	Definition	Reference
Arctic Oscillation (AO)	Leading EOF of the 1000hPa geopotential height anomalies poleward of 20°N	Thompson and Wallace (1998)
East Asian Winter Mon- soon intesity index (EAWMI)	Difference in area averaged 300hPa zonal winds $U_{300}(27.5^{\circ}-37.5^{\circ}\text{N}, 110^{\circ}-170^{\circ}\text{E})-U_{300}(50^{\circ}-60^{\circ}\text{N}, 80-140^{\circ}\text{E})$	Jhun and Lee (2004)
Monsoon Index (MO)	Difference in MSLP between two grid points (105°E, 52.5°N) near Irkutsk and (145°E, 43.75°N) near Nemuro.	Sakai and Kawamura (2009)
Siberian High intensity in- dex (SHI)	Area averaged MSLP (40°-65°N, 80°-120°E)	Wang and Chen (2010)
East Asian Temperature Gradient (EATG)	Gradient in zonally averaged temperature between 30°- 120°E, and taken the averaged gradient between 40° and 50°N	J. Liu et al. (2020)

 Table 2.
 Definition of the climatic indices used in the correlation analysis.

In the following, we utilize the FLEXPART simulations to examine the factors 473 driving the interannual variations in the spring EADC over the past 21 years from 474 1999 to 2019. We establish correlations between dust emission and deposition and 475 various climatic variables representing different aspects of the East Asian circula-476 tion. The indices that we used, along with their definitions, are summarized in Table 477 2. All indices are calculated from monthly ERA5 data for both winter (December-February) and spring (March-May). Decadal trends are removed from the time se-479 ries before calculating the correlations to avoid the influence of low-frequency cli-480 mate variations. 481

The correlations of the winter and spring circulation with spring dust emissions 482 are shown in Figure 7b,c. Figure 7b shows that the circulation of the preceding win-483 ter has a significant impact on the frequency and intensity of dust storms in spring. 484 Emissions from the northwestern deserts are anticorrelated with winter AO. As ev-485 ident in Figure 5 in the previous section, this is the most significant dust source for 486 the CLP. Interestingly, winter AO does not significantly affect emissions from other 487 source regions. Instead, Taklamakan emissions are negatively correlated with the 488 winter monsoon and are most strongly correlated with the Siberian High Intensity 180 Index (SHI), suggesting that the location and strength of the SH are important factors that regulate dust emissions in the Taklamakan desert. Qaidam emissions have 491 correlations similar to those of Taklamakan, albeit weaker. Emissions from Central 492 Asia and Gurbantunggut are not strongly related to the winter circulation of East 493 Asia. For the spring circulation shown in Figure 7c, dust emissions in Mongolia and 494



Figure 7. (a) Correlations between winter (December-January-February, DJF) and spring (March-April-May, DJF)circulation indices and modeled spring dry- and wet deposition of supercoarse dust, for the whole domain and contributions from individual source regions. (b-c) Correlation with FLEXDUST-EA dust emissions in the source regions and with winter and spring circulation respectively. The size of the circle indicate the strength of the correlation.

northwestern deserts are more strongly correlated with the magnitude of the East
Asian Temperature Gradient (EATG), that is, a large negative temperature gradient favors larger emissions. Taklamakan emissions do not show a significant correlation with any particular spring circulation index. Qaidam is similar again to Taklamakan except that it is more positively correlated with AO. The intensity of the SH
in spring is significantly anti-correlated with emissions from Central Asia and Gurbantunggut.

Figure 7a shows the correlations between source contributions of super-coarse dust, delineated by contributions from each dust emission region to each site. Looking at the winter circulation, we observe significant negative correlations between the AO and dry deposition at Shapotou, SACOL, Baode, and Luochuan. Additionally, AO is negatively correlated with wet deposition at Lantian and SACOL. The correlations with AO are most pronounced for source contributions from Mongolia and the northwestern deserts, although they vary by site.

Interestingly, wet deposition at SACOL demonstrates significant correlations 509 with all winter circulation indices, including AO and MO, both of which are linked 510 to the EATG. Additionally, the SHI correlates with both AO and MO, even though 511 AO and MO do not exhibit a significant correlation with each other (see Supplement 512 Figure S3). Considering these insights, we infer that, for wet deposition at SACOL, 513 the strength of EATG in winter plays a critical role in creating favorable conditions 514 for wet deposition in spring, mainly sourced from the northwestern deserts. How-515 ever, for dry deposition at SACOL, AO is the only significantly correlated index, 516 aligning with AO's correlation with emissions from the northwestern sources, the 517 primary dust source for SACOL. 518

The strength of the EAWM is correlated with dust from Qaidam and Gurban-519 tunggut to the SACOL site. The spring EAWM circulation features are correlated 520 with an increased contribution from the Gurbantunggut desert to Lantian, Lingtai, 521 and Luochuan. The deposition of Central Asian dust at the sites is generally not 522 correlated with any of the circulation indicators, except at Baode site, which shows a 523 significant negative correlation between the contribution of dry deposited dust from 524 Central Asia and the SHI, where the SH can act as a barrier for long-range dust 525 transport. 526

The correlations presented above are for the super-coarse dust size bin. Fine 527 dust correlations are provided in the Supplement (Figure S4). The correlations be-528 tween the winter AO circulation and spring deposition remain consistent across fine 529 and super-coarse dust. However, in terms of spring circulation, the two particle sizes 530 are less consistent. Dry-deposited fine dust displays notably higher correlations with EATG and EAWMI, especially for dust transport from Taklamakan compared to 532 super-coarse dust. For the north western deserts fine and super-coarse dust exhibit 533 similar correlations with EATG. The correlations between circulation patterns and 534 wet deposition for fine dust are highly variable and strongly dependent on the site, 535 and this does not change from what is shown in Figure 5. This underscores that wet 536 deposition is the result of an intricate dynamic between the source, receptor, and 537 circulation modes. 538

To interpret the relationship between atmospheric circulation patterns and 539 dust deposition over the CLP, as revealed in the previous correlation analysis shown 540 in 7, we performed a composite analysis of years with weak and strong deposition 541 at sites, to identify site-dependent circulations features that typically cause an in-542 crease in dust deposition. Figure 8 shows the composite differences of 850hPa winds 543 and mean sea level pressure (MSLP), hatched areas show the regions where the dif-544 ferences between the strong and weak dust deposition years are significant. In the 545 case of dry deposition of super-coarse dust, all sites manifest AO-like composite dif-546



Figure 8. Composite difference of mean sea level pressure and 850hPa wind strong minus weak deposition years for super coarse dust for both winter (December-January-February, DJF) and spring (March-April-May, MAM) for all the locations. The left half shows composite difference based on dry deposition rate, while the right half is the composite difference based on total deposition rate. The hatched area indicates where the anomalies are significant (p < 0.05)

ferences during the winter preceding a strong deposition year. These anomalies show characteristic positive MSLP anomalies over the Arctic, typical of negative AO-like conditions (Thompson & Wallace, 1998; He et al., 2017). This AO-like pattern extends into the spring for most sites. However, for total deposition at Shapotou and Luochuan, the AO pattern is masked by other influences, as wet deposition is more event-dependent and less influenced by the overarching circulation state. A longer time series of simulations would be able to better distinguish the more random fluctuations due to wet deposition from the impact of large-scale circulation.

Figure 9 shows the composite anomalies of the geopotential height and winds at 500hPa for super-coarse dust. During the winter preceding a strong deposition year, it indicates an area of reduced geopotential height that for Lantian, Lingtai, Luochuan and Baode is located to the north west of the site, while for Shapatou and SACOL negative geopotential height anomaly is located more to the north east. This suggests that increases in dust deposition as a result of dry deposition at these sites are closely associated with the location of this geopotential height anomaly and the corresponding changes in the wind patterns.

During the negative phase of AO, there is usually an increase in the frequency 563 and intensity of cold air outbreaks (COAB) in East Asia (He et al., 2017; Yang et 564 al., 2020). H. Liu et al. (2018) conducted a detailed analysis investigating the rela-565 tionship between spring dust emissions and the preceding winter AO using MERRA-2 reanalysis. Using regression analysis, they revealed a correspondence between winter 500 hPa geopotential height and the leading empirical orthogonal function of 568 spring dust emissions, similar to the composite anomalies we found shown in Figure 569 9. H. Liu et al. (2018) suggested that a negative AO results in anomalous cold con-570 ditions in central Siberia, which persist into the ensuing spring, amplified by snow-571 albedo and cloud feedbacks. The resultant colder temperatures in the inner regions 572 of East Asia create a more negative temperature gradient in the region, intensify-573 ing the East Asian Trough (EAT). This in turn creates a favorable environment for dust emissions. Their explanation of how winter AO influences spring surface tem-575 perature aligns with our correlation analysis, where a negative AO corresponds to a 576 more pronounced temperature gradient over East Asia, elevating dust transport to 577 the CLP from the northwestern sources. 578

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### 3.4 Implications for interpreting Chinese Loess Plateau dust records

Our results reveal a clear link between the depositional mechanism and the dust-emitting regions. This observation may have major implications on how we in-581 terpret the provenance of the CLP dust records (Quaternary loess and Neogene red 582 clay) also on geological time scales. The CLP dust provenance studies often con-583 sider changes in emission and transport mechanisms, but commonly overlook the fi-584 nal step of the EADC, the deposition. Our results on the deposition of both fine and 585 super-coarse particles indicate that wet deposition contains more dust from west-586 ern emission areas than dry deposition (Figure 6), suggesting the shift in CLP dust 587 provenance may not only be related to changes in dust transport pathways but also the deposition mode. The CLP dust records are associated with coarser grain sizes 589 during glacials compared to interglacials. These differences have been explained by 590 higher dust influx and reduced soil development coupled with stronger wind speeds 591 during glacial periods (Maher, 2016; Kohfeld & Harrison, 2003). However, our re-592 sults imply that increased wet deposition of fine/clay-sized dust during the wetter 593 interglacials could provide an additional explanation for the observed grain-size dif-594 ferences between glacials and interglacials. Similarly, the finer grain size of late Neogene red clay deposits compared to Quaternary loess may be associated with an in-596 crease in the wet deposition of finer particles during intensified EASM conditions 597 (Ren et al., 2020; Z. L. Ding et al., 1999). 598



Figure 9. Circulation composite anomalies of 500hPa wind strength (colored, unit m/s), wind direction (vectors, unit m/s) and geopotential height (contours, unit dam, distance between contours 2 dam, solid contour are positive anomalies while dotted contours are negative anomalies) for winter (December-January-February, DJF) and spring (March-April-May, DJF) of strong - weak dry and total deposition years. The hatched area indicates where the anomalies are significant (p < 0.05)

The results of our model, which covers the full dust cycle (i.e., emission, trans-599 port, and deposition), may also have important implications on the observed spatial 600 differences in source contribution across the CLP. Previous single-grain provenance 601 studies (targeting silt-sized particles) have documented Neogene and Quaternary dust source differences between the northeastern (e.g., Baode site) and the south-603 western (e.g., Lantian) CLP (Shang et al., 2016; H. Zhang et al., 2021; W. Peng et 604 al., 2023; Bird et al., 2015). These observations were supported by backward tra-605 jectory modeling showing more dominant atmospheric transport pathways from the 606 Taklamakan desert and Northern Tibetan Plateau to the southwestern CLP than to 607 the northeastern CLP (Shang et al., 2016). The results of our model, taking into ac-608 count the full EADC, further corroborate these conclusions (Figure 5). In addition, spatial differences in dust provenance have been found to be particularly prominent 610 in coarse dust particles and wet deposited dust (Figure 5). This offers a plausible 611 eolian mechanism for explaining the spatial provenance contrast of the red clay and 612 loess deposits, and its temporal variations, in addition to the well-established hy-613 pothesis emphasizing the importance of large fluvial systems (namely the Yellow 614 River) in providing dust particles closer to the CLP, and therefore in creating the 615 observed spatial provenance gradient (Stevens et al., 2013; Bird et al., 2015; Nie et 616 al., 2015; W. Peng et al., 2023; H. Zhang et al., 2021; Sun et al., 2022). For instance, 617 W. Peng et al. (2023) found an increase in Pliocene spatial provenance contrast in 618 the Red Clay, and argued that this was partly caused by the intensified Pliocene 619 EASM. This argument is well in line with our results, since the intensified EASM 620 can enhance the wet deposition flux over the CLP. Therefore, an increase in wet de-621 position can act to amplify the spatial provenance differences created primarily by 622 the fluvial input of material. 623

Our modeling results with super-coarse/silt-sized dust particles indicate that 624 even large dust particles deposited on the CLP may be sourced from distant dust-625 emitting sources (e.g., Taklamakan) although they are not the dominant contribu-626 tor to the deposition on the CLP. Larger dust particles (super-coarse or larger) de-627 posited far from the dust emission region have also been reported in recent observa-628 tional studies (Van Der Does et al., 2018; Ryder et al., 2018; A. A. Adebiyi & Kok, 2020). The fact that we also find reason to believe that super-coarse dust deposited 630 over the CLP can be from the distant dust-emitting regions provides additional sup-631 port to the validity of the single-grain provenance proxy studies using mainly the 632 large mineral particles in detecting the source origin of emitted dust. Strengthen-633 ing claims and insights on loess formation mechanisms and climatic implications 634 made based on the coarse dust fractions in the loess deposits. We note that spheri-635 cal particles are assumed in our model simulations, thus might be considered a lower 636 bound of the long-distance transport of super-coarse dust to the CLP. Further modeling studies using a-spherical particles with lower settling velocities and potentially 638 longer transport distance than spherical particles may help to better understand the 639 long-distance transport of large dust particles. 640

The interannual variation of the spring dust deposition rate over the CLP has 641 been found to be more closely associated with AO than with the strength of the 642 EAWM in both winter and spring (Figure 7 and S4). This is distinct from the com-643 mon understanding within the paleoclimate communities which emphasizes the im-644 portance of EAWM in regulating the dust cycle over the CLP (e.g. An et al., 2014). 645 However, consistent with studies of the present-day interannual variation of the EADC, 646 which generally support the strong control of AO instead of EAWM on the variation 647 of EADC (Mao et al., 2011; H. Liu et al., 2018), with the negative AO being favor-648 able for stronger dust storms in East Asia. Although the results are based on the annual time scale, they may have strong implications for understanding long-term 650 changes in the EADC and the interpretation of the red clay and loess record, espe-651 cially when it is difficult to use the strength of EAWM to explain the dust records 652

and its variations. For instance, provenance research on the dust records deposited 653 during the warmhouse Eocene indicates the absence (or significantly weaker) SH 654 coupled with possible long-term negative phase of AO -like conditions (Bohm et al. 655 2023X, in revision). Moreover, it has been noticed that the cyclicity of the oxygen isotopes of speleothem records over East Asia, an indicator of East Asian summer 657 monsoon, is dominated by precession cycle (20 ka) on orbital time scale, which is in 658 contrast to the loess records, which reveal a dominance of the glacial-interglacial cy-659 cle (100 ka) (Cheng et al., 2021). The larger influence of AO-like conditions (over 660 EAWM) on the variation of dust deposition over the CLP during the glacial-inter-661 glacial cycle, may offer a plausible explanation of the "apparent" discrepancy be-662 tween loess and speleothem records from East Asia. This explanation is consistent with the fact that high-latitude forcing is the dominant influence on Loess records (Stevens et al., 2018). However, on the basis of this work, it is not possible to con-665 clude whether this is generally the case because the climatic conditions during the 666 glacial stages are very different from those of today. Although, this could be an-667 swered by driving this modeling setup using data from paleoclimate simulations in the future. 669

### 670 4 Conclusion

To our knowledge, this is the first modeling study to provide a detailed picture 671 of how proximate and distant dust emission areas contribute to dust accumulation 672 on the CLP. We provide evidence for the importance of considering wet deposition 673 when interpreting the loess records, showing that wet-deposited dust differs in both 674 the typical transport trajectory and the source of the emitted dust. Even super-675 coarse dust could be wet deposited at the CLP where sloping topography upwind 676 of dust emission area can act to increase the vertical dust transport substantially, making it possible for even large dust particles to be scavenged inside clouds. This 678 could be an important mechanism for bringing potentially non-negligible amounts 679 on of coarse dust particles from more distant sources like the Taklamakan desert and 680 Quaidam basin. Dry-deposited dust is mainly sourced from the nearby dust-emitting 681 areas to the north-west of the CLP, this transport pathway is favorable under nega-682 tive AO-like conditions. That the AO is an important driver of the EADC is in line 683 with our understanding of the EADC under present day climate conditions (H. Liu 68/ et al., 2018; Mao et al., 2011; D.-Y. Gong et al., 2006). This study provides a direct link between AO and dust accumulation on the CLP. Our link between AO and dust accumulation on the CLP was established based on current climate conditions, 687 which were different in the past, but it does warrant a stronger emphasis on high-688 latitude forcings when explaining the variability in dust accumulation rate of the 689 loess record rather than the common partitioning of enhanced summer monsoon or 690 winter monsoon conditions, as also suggested by Roe (2009). 691

FLEXPART has also been shown to be a very useful tool for understanding 692 the dust cycle, as it can pinpoint the exact dust source contribution of a receptor 693 site. The FLEXPART ES is independent of the dust emission inventory, so FLEX-694 PART can be used to easily test the effect and impact of different emission schemes 695 and hypothesis of changes in dust emission regions in the past. A promising future application of this setup would be to combine FLEXPART and state-of-the-art pale-697 oclimate model reconstructions to give insight into how environmental changes have influenced the source region and dust transport mechanism at the CLP. This would 699 be invaluable in formation to further aid in interpretation of the CLP provenance 700 proxies and help to understand how the EADC might change in the future. 701

- 702 Acronyms
- 703 CAOB Cold air outbreaks
- **EAWM** East Asian Winter Monsoon
- 705 CLP Chinese Loess Plateau
- 706 AO Arctic Oscillation
- 707 LPDM Lagrangian particle dispersion model
- 708 MAR Mass accumulation rate
- **EOF** Empirical orthogonal functions
- 710 MSLP Mean sea level pressure
- 711 DJF December January February
- 712 MAM March April May
- **EADC** East Asian dust cycle
- **ES** Emission sensitivity
- **515 SHI** Siberian High Intensity index
- **EATG** East Asian Tempereature Gradient
- 717 MO Monsoon index

### 718 Acknowledgments

This work was supported by the Research Council of Finland (grant no 316799). 719 The simulations were performed on resources provided by Sigma2 - the National In-720 frastructure for High Performance Computing and Data Storage in Norway, project 721 no. nn2806k. The MERRA-2 data used in this study have been provided by the 722 Global Modeling and Assimilation Office (GMAO) at NASA Goddard Space Flight 723 Center and are available from NASA GES DISC (https://disc.gsfc.nasa.gov/ 724 datasets?project=MERRA-2 last access October 2021). ERA5 data used in this 725 study were downloaded using the Copernicus Climate Change Service (C3S) Climate 726 Data Store. Neither the European Commission nor ECMWF is responsible for any 727 use that may be made of the information it contains. 728

The FLEXDUST-EA source code is available at https://github.com/huitang 729 -earth/FLEXDUST/tree/FES2022\_Tangetal\_updated\_KOK14, and the FLEXPART 730 codes are available at https://github.com/Ovewh/flexpart/tree/release-10. 731 The workflows for setting up the FLEXPART simulations are available on github: 732 https://github.com/MasterOnDust/FLEXPART-script/releases/tag/ ) The work-733 flow for post-processing the raw simulation output and combining the FLEXDUST 734 emission inventory with FLEXPART ES is available on GitHub (https://github .com/MasterOnDust/FP\_preprosses\_workflow/tree/AGU-Haugvaldstad-et-al 736 -2023). The workflow to generate the figures shown in this manuscript is available 737 in the following GitHub repository:https://github.com/MasterOnDust/AGU\_JGR 738 \_CLP\_SOURCE\_WORKFLOW/tree/AGU-Haugvaldstad-et-al-2023 . The data files re-739 quired to generate the figures shown in the manuscript are available from Zenodo 740 Haugvaldstad et al. (2023) https://doi.org/10.5281/zenodo.10114436. The 741 full 3hourly FLEXPART source contribution, emission sensitivity and FLEXDUST 742 dust emission inventory are archived on the Norwegian Research Data Archive; see 743 (DOI: https://doi.org/10.11582/2023.00134, https://doi.org/10.11582/ 744 2023.00135), Haugvaldstad (2023a, 2023b). 745

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### Spatial source contribution and interannual variation in 1 deposition of dust aerosols over the Chinese Loess 2 Plateau

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16	•	The source contribution of emitted dust aerosols to the Chinese Loess Plateau
17		(CLP) is mapped using backward trajectory modeling.
18	•	Distant dust emission regions in Central Asia have a non-negligible impact on
19		the CLP, with wet-deposited dust showing a clearer connection.
20	•	Strong dust deposition years over the CLP are associated with the negative

phase of the Arctic Oscillation.

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### 22 Abstract

The Chinese Loess Plateau (CLP) in northern China serves one of the most promi-23 nent loess records in the world. The CLP is an extensive record of changes in past 24 aeolian dust activity in East Asia; however, the interpretation of the loess records is hampered by ambiguity regarding the origin of loess-forming dust and an incomplete understanding of the circulation forcing dust accumulation. In this study, we 27 used a novel modeling approach combining a dust emission model FLEXDUST with 28 simulated back trajectories from FLEXPART to trace the dust back to where it was 29 emitted. Over 21 years (1999-2019), we modeled back trajectories for fine (~ 2  $\mu$ m) 30 and super-coarse ( $\sim 20 \ \mu m$ ) dust particles at six CLP sites during the peak dust 31 storm season from March to May. The source receptor relationship from FLEX-32 PART is combined with the dust emission inventory from FLEXDUST to create 33 site-dependent high-resolution maps of the source contribution of deposited dust. 34 The nearby dust-emission areas dominate the source contribution at all sites. Wet 35 deposition is important for dust deposition at all sites, regardless of dust size. Non-36 negligible amounts of dust from distant emission regions could be wet deposited 37 on the CLP following high-level tropospheric transport, with the super-coarse dust 38 preferentially from emission areas upwind of sloping topography. On an interan-39 nual scale, the phase of the Arctic Oscillation (AO) in winter was found to have 40 a strong impact on the deposition rate on the CLP, while the strength of the East 41 Asian Winter Monsoon was less influential. 42

### 43 Plain Language Summary

The dust deposits on the Chinese Loess Plateau (CLP) in northern China pro-44 vide an extensive record of changes in past dust activity in East Asia. In this work, 45 we combine an atmosphere transport model with a dust emission model to trace the 46 dust backward to where it was emitted. Choosing six locations at the CLP where we 47 trace the dust back to emissions over a 21-year period during the most active dust 48 months from March until May, for which we investigated how different parts of the 49 CLP were influenced by the different dust emission areas, transport mechanisms, 50 and wind circulation patterns. We find that dust is mainly brought from the emis-51 sions areas located near the CLP, still dust from far-away emissions areas in Cen-52 tral Asia has the potential to reach the CLP in smaller amounts. Furthermore, our 53 results show that we removal of dust by precipitation is important for dust deposi-54 tion on the CLP, a mechanism that is often overlooked when explaining variations 55 in past dust activity. On a year-to-year basis the dust deposition rate over the CLP 56 was found to be connected to the high-latitude northern hemisphere's climate oscil-57 lation (Arctic Oscillation) rather than the regional East Asian winter monsoon. 58

### 59 1 Introduction

Mineral dust is deeply embedded within the Earth system, influencing the 60 planet's energy balance, cloud characteristics, atmospheric chemistry, and biogeo-61 chemistry (Shao et al., 2011; Kok et al., 2023). Dust is mainly emitted from dry 62 and sparsely vegetated arid and semi-arid regions. Emission is driven by surface 63 winds that lift sand grains from the soil bed, where the impact upon reentry ejects 64 and fragments smaller soil particles, which then get transported. The dust-emitting 65 capacity of the soil is strongly influenced by environmental factors such as soil mois-66 ture, vegetation, soil texture, and the structure of the top soil. On geological time 67 scales, dust depositional records show increases in dust activity during glacial periods (Porter, 2001; Maher, 2016). However, as human expansion has increased the in-69 tensification of land use in recent years, we are now seeing a profound impact of hu-70 man activity on dust activity, e.g. the drying of Aral Sea and dust bowl in the 1930s 71

North America. The antropogenic signal is also evident in the depositional records,
which show that in the last 150 years there has been an increase in the dust burden
globally of about 50% (Hooper & Marx, 2018). However, we do not know to what
extent this is due to a forcing caused by increased human activity and changes in
land use in semi-arid regions or to modifications in precipitation and wind patterns
in a changing climate (Kok et al., 2023).

Changes in dust activity are important because of the climate effect of dust. 78 Fine dust aerosols exhorts cooling effect due to its ability to reflect incoming sun-79 light, whereas coarse dust aerosols cause a warming effect as they interact with the outgoing long-wave radiation. Furthermore, different dust-emitting regions have dif-81 ferent dust mineralogy, which affects its ability to absorb radiation (Dubovik et al., 82 2002). Atmospheric dust particles also affect clouds by acting as nuclei for conden-83 sation, contributing to droplet formation. Additionally, dust particles act as nuclei 84 to initiate glaciation of clouds above the temperatures of homogeneous freezing. Ice 85 clouds generally have a warming effect, whereas liquid clouds have a cooling effect 86 (Storelymo, 2017). The deposition of dust on snow accelerates snow melt (Wittmann et al., 2017; Sarangi et al., 2020), while dust input can increase ecosystem produc-88 tivity in regions lacking nutrients, by providing iron or phosphorus (Martin, 1990; 89 Yu et al., 2015). These processes impact the climate system, but the complexities 90 introduced by the poor constraints of, for example, dust particle size (A. Adebiyi et 91 al., 2023; Ryder et al., 2018), shape (Huang et al., 2020), mineralological composi-92 tion (Li et al., 2021) and dust-cloud interactions (Sagoo & Storelvmo, 2017) of air-93 borne dust particles, make the overall climatic effect of the dust uncertain (Kok et 94 al., 2023).

One of the most prominent dust records on Earth for studying environmental drivers of changes in the dust cycle is the Chinese Loess Plateau (CLP), located in the north central part of China (Figure 1). Here windblown dust has been accumulating since at least the late Oligocene-early Miocene (c. 25-22 Ma) (Guo et al., 2002; Qiang et al., 2011), providing a long-term, nearly continuous record of past responses of the East Asian Dust Cycle (EADC) to environmental and climatic changes (e.g. Stevens et al., 2007; Lu et al., 2010; Maher, 2016; Y. Sun et al., 2020).

The sediment sources, dust-emitting regions, transportation, and deposition 103 processes of the eolian deposits on the CLP are debated (e.g. W. Peng et al., 2023; H. Zhang et al., 2022; Shang et al., 2016; Bird et al., 2015; D. Sun et al., 2008). 105 However, most agree that the dust was generally sourced from the dry Asian inte-106 rior and proximal deserts via energetic, surface-level northwesterly winds driven by 107 the East Asian Winter Monsoon (EAWM) system (Z. Ding et al., 1999). In addition 108 to the EAWM, the prevailing mid-latitude westerlies in the middle and upper tro-109 posphere play an important role in transporting dust to the CLP. The EAWM and 110 the westerlies are suggested to explain the bimodal particle size distribution widely observed in the CLP records. The coarse particles are suggested to be transported 112 by the strong surface winds associated with the EAWM and the fine particles are 113 suggested to represent the background dust component of the CLP transported by 114 the high-level westerly air flow (Miao et al., 2004; D. Sun, 2004; D. Sun et al., 2008). 115 Furthermore, both the particle size distribution and the dust accumulation of the 116 loess records exhibit a northwest-southeast gradient (Z. Ding et al., 1999), which fur-117 ther supports the northwesterly and westerly wind driven transport pathways. 118

Recent studies using single grain provenance indicators on the CLP dust records shed more light on dust emission areas and transport pathways, and their variation in the geologic past (Bird et al., 2015; Shang et al., 2016; Nie et al., 2018; H. Zhang et al., 2021, 2022; Bohm et al., 2023; W. Peng et al., 2023). However, the interpretation of the CLP provenance records is hampered by the complexity of the source-tosink system and by the fact that dust provenance signals reflect not only the trans-
porting wind systems, but also the various processes that affect dust availability in 125

the source regions. Furthermore, single grain provenance ultimately tells us what 126 the source rocks and sediments are, which might be different from the dust-emitting

127

regions. 128

Several provenance studies have reported spatial provenance differences across 129 the CLP for both Neogene Red Clay and Quaternary loss (e.g. Bird et al., 2015; 130 Shang et al., 2016; H. Zhang et al., 2021; W. Peng et al., 2023). However, the cause 131 of the spatial differences in provenance is not well established. The Yellow River 132 has been argued to cause these spatial differences by bringing material from the 133 Northern Tibetan Plateau closer to the CLP before the final aeolian transport step 134 (Stevens et al., 2013; Nie et al., 2015). Contrasting eolian dust transport pathways 135 for different parts of the CLP have also been proposed to add to the spatial dif-136 ference (Shang et al., 2016), with the northern and northeastern parts of the CLP 137 dominated by dust from the northwestern deserts via the EAWM, and the south-138 ern and southwestern parts of the CLP dominated by dust from the Northern Ti-1 20 betan Plateau and even the remote Taklamakan desert via the westerlies (Shang et al., 2016). 141

Applications of the provenance method to further unravel the dust cycle over 142 the CLP is also hampered by the fact that most single grain provenance approaches 143 are based on a relatively large particle size (>  $20 \ \mu m$ ). Our understanding of the transport of such large dust particles in the atmosphere is still limited and highly debated (A. Adebiyi et al., 2023). Super-coarse dust particles (sensu A. Adebiyi et 146 al. (2023),  $> 20\mu m$ ) have commonly been assumed to be transported only a short 147 distance (1-100 km) from the source region because of the high gravitational settling 148 velocity (Tsoar & Pye, 1987), which contradicts CLP provenance inferences of dust 149 derived from remote deserts, such as the Taklamakan. Furthermore, recent observa-150 tions have documented the possibility of long-distance transportation (> 1000 km) of 151 giant (  $> 62.5\mu m$ ) dust particles (Van Der Does et al., 2018; Varga et al., 2021). A better understanding of large dust particle transportation to the CLP will help the 153 interpretation of dust provenance proxies, and thus improve our understanding of 154 the EADC on geological time scales. 155

Dust models have been widely applied to understand the EADC, however, 156 there have been only limited cases where dust models have been applied to understand the formation of the CLP dust records in particular (Shi & Liu, 2011). More-158 over, previous modeling studies of past and present EADC have typically applied 159 global or regional climate models (Shi & Liu, 2011; S. L. Gong et al., 2006), which 160 mainly consider particles within the fine to coarse range (  $< 10\mu$ m) (Zhao et al., 161 2022). These models are also unable to determine the exact sources of emitted dust 162 for different locations at the CLP, which would greatly enhance the provenance in-163 terpretations of the CLP dust records. To address these issues, we used the Lagrangian particle dispersion model FLEXPART. We run FLEXPART backward in a 165 receptor-oriented viewpoint, which, paired with a dust emission field, allows us to es-166 tablish high-resolution source contribution maps. We apply FLEXPART to establish 167 source receptor relationships between six loess sites across the CLP and examine the 168 interannual variations in deposition at these over a 20-year period from 1999-2019. 169 In this study, both fine (1.7-2.5  $\mu$ m) and super-coarse (15-20  $\mu$ m) dust particles are 170 simulated to better understand whether the deposition of fine particles occurs under 171 the same climate conditions as that of coarse particles and whether the source re-172 gions are the same. We chose to study the interannual variation in the dust deposi-173 tion rate over the CLP because we are interested in understanding the processes re-174 sponsible for the changes in loess MAR, with implications for identifying the factors 175 that could be driving the changes that have occurred on longer time scales (F. Peng 176 et al., 2022). 177

## <sup>178</sup> 2 Model and Data

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## 2.1 FLEXDUST and FLEXPART

FLEXDUST is an offline dust emission model developed to be used in pair 180 with the Lagrangian dispersion model FLEXPART to study the long-range trans-181 port and deposition of windblown dust (Groot Zwaaftink et al., 2016). The combination FLEXDUST-FLEXPART has previously been applied to study the sources, 183 transport, and deposition of high-latitude dust (Groot Zwaaftink et al., 2017; Kylling 184 et al., 2018; Wittmann et al., 2017). The dust emitted in FLEXDUST is assumed 185 to have a volume size distribution between 0.2 and 20  $\mu m$  according to the brittle fragmentation theory described in Kok (2011). Tang et al. (2023) identified sev-187 eral weaknesses in the way dust-emitting areas in East Asia were represented in 188 the original FLEXDUST version. Therefore, we use the updated version of FLEX-DUST by Tang et al. (2023), which includes several changes to better represent present day dust emission areas in East Asia. To avoid confusion with the stan-191 dard version of FLEXDUST, we refer to the updated Tang et al. (2023) version of 192 FLEXDUST as FLEXDUST-EA. Changes in FLEXDUST-EA compared to FLEX-193 DUST include: (1) the parameterization of vertical dust flux described by Kok et 194 al. (2014) with a modified parameter for the calculation of the threshold friction ve-195 locity according to Shao and Lu (2000), (2) the soil texture dataset in the ISRIC 196 SoilGrids dataset described by de Sousa et al. (2020), (3) new improved topographical erodibility scaling, and (4) herbaceous and crop land cover types in the GLC-NMO land cover dataset are considered to have a potential to emit dust in addition 199 to bare ground and sparse vegetation. For a complete description of the changes in 200 FLEXDUST-EA compared to the standard version, see Tang et al. (2023). 201

Dust transport and deposition are calculated using FLEXPART version 10.4 202 (Pisso et al., 2019). FLEXPART is an open source model and has been applied to 203 study the transport of a wide range of atmospheric tracers, such as, mineral dust, 204 black carbon, and volcanic ash (Groot Zwaaftink et al., 2022, 2016; Choi et al., 2020; 205 Eckhardt et al., 2008). FLEXPART calculates the trajectories of a large number 206 of computational particles that are used to describe the transport and deposition 207 of tracers in the atmosphere. This version of FLEXPART includes the improved 208 wet deposition scheme based on cloud information from the ECWMF input fields (Grythe et al., 2017). In addition, FLEXPART accounts for gravitational settling, dry deposition, and in- and below-cloud scavenging of simulated dust aerosols. Dry 211 deposition is treated using a resistance scheme (Stohl et al., 2005) and in-cloud wet 212 deposition distinguishes between scavenging in the liquid and ice phase (Grythe et 213 al., 2017). 214

FLEXPART can be run both in forward (source-oriented) and backward (receptor-215 oriented) mode. The model gives the same results when running in forward or back-216 ward mode, except for some minor numerical differences (Seibert & Frank, 2004), 217 and therefore the direction can be set for computational efficiency. Since we are in-218 terested in mapping the spatiotemporal contribution of different dust-emitting re-219 gions to a receptor, we run FLEXPART in backward mode. In the backward con-220 figuration, computational particles are released at a predefined receptor location, 221 and then each particle is traced backward in time. The distribution of the residence time of the computational particles in each grid cell of the output grid is used to 223 calculate the emission sensitivity (ES). ES gives a relationship between a poten-224 tial source in a grid cell and deposition at the receptor. ES is calculated separately 225 for each dust particle size bin and deposition process. We include two size bins in 226 our FLEXPART simulations, corresponding to fine dust (mean diameter  $2\mu m$ ) and 227 super-coarse dust (mean diameter  $17.6\mu m$ ) following the terminology of A. Adebiyi 228 et al. (2023). These two sizes were selected to represent the clay and silt fractions that are typically observed in the Chinese loess. In addition to size, efficiencies for



Figure 1. Digital elevation model (Shuttle Radar Topography Mission) and the locations of the Chinese Loess Plateau, the study sites and major dust emitting areas in Central-East Asia. The distribution of loess, loess-like silt, and the undifferentiated sediments are from Börker et al. (2018), and the deserts and wind 477 eroded land in China from 1:200,000 Desert Distribution Dataset provided by the Environmental and Ecological Science Data Center 478 for West China, National Natural Science Foundation of China (http://westdc.westgis.ac.cn; last access 27 July 2020).

each of the wet removal processes can be assigned differently depending on the kind 231 of dust-aerosol represented. The parameters we used to represent the two types of 232 airborne dust particles in our FLEXPART simulations are listed in Table 1. Dry 233 deposition ES is calculated by releasing particles from the receptor at the surface level between 0-30 meters and then calculating the particle backtrajectories. Wet 235 deposition ES is calculated by releasing particles throughout the atmospheric col-236 umn to determine whether a particle is wet scavenged. Then only backtrajectories of 237 the particles that undergo wet scavenging are calculated. Consequently, to achieve 238 a statistically reliable outcome, a greater number of particles are necessary for wet 239 deposition simulations. ES averaged over a selected output layer height can be mul-240 tiplied by the dust emission inventory (units kg  $m^{-3}s^{-1}$ ) created by FLEXDUST, to 241 generate a map that quantifies each contribution of the source element to the depo-242 sition at the receptor (Eckhardt et al., 2017). Summing the source contribution (SC) 243 of each source element yields the deposition rate at the receptor. 244

#### 245 2.2 FLEXPART backtrajectory set up and workflow



**Figure 2.** Flow chart showing the workflow for the FLEXPART-FLEXDUST backward trajectory modeling analysis.

Backward FLEXPART simulations were performed during the main dust storm season between March 1st and May 31st over a 21-year period from 1999 until 2019 for six locations across the CLP shown in Figure 1. The selected sites encompassed both well-established loess sites with historical loess and red-clay deposits (Baode, Luochuan, Lingtai, and Lantian ), as well as modern dust observation sites (Shapotou and SACOL). This diverse geographical selection of sites allows for a comprehensive assessment of dust deposition susceptibility across different parts of the CLP to different dust emissions regions and circulation patterns.

FLEXPART was configured the following way, every particle release contained 254 either 50,000 or 200,000 particles, depending on whether dry or wet deposition is 255 simulated. Particles within one release are evenly distributed over 3-hourly inter-256 vals. The extent of the backtrajectories was limited to a maximum particle age of 257 10 days. ES was calculated with 0.1 degree resolution, with an output grid spanning 258 50-128° longitude, 25-65° latitude, and stored as 3-hourly averages. The FLEXDUST-259 EA dust emission flux is multiplied by the averaged ES of 0-100 meters to derive the deposition source contribution. The parameters of the two particle size bins that we include are listed in Table 1. We have summarized the stages of the workflow in 262 Figure 2, commencing with the retrieval and preparation of the forcing data. Then 263 the calculations of dust emissions, dry and wet dust deposition ES are done concur-264 rently. Finally, the FLEXDUST dust emission inventory is combined with FLEX-265 PART ES during post-processing to produce the source contribution maps and the 266 deposition rate for each of the receptor locations. 267

	Description	Fine	Super-coarse	
PCRAIN_AERO	Efficiency of below cloud scavenging by rain $(1=100\%, 0=\text{off})$	1.00	1.00	
PCSNOW_AERO	Efficency of below cloud scavenging by snow $(1=100\%, 0=off)$	1.00	1.00	
PCC_AERO	CCN efficiency (in cloud scavenging)	0.15	0.30	
PIN_AERO	IN efficiency (in cloud scavenging)	0.02	0.10	
PDENSITY	Particle density (dry deposition)	$2500.0{\rm kg}{\rm m}^{-3}$	$2500.0{\rm kg}{\rm m}^{-3}$	
PDQUER	Mean particle diameter (dry deposition)	$2.057~\mu\mathrm{m}$	17.32 $\mu \mathrm{m}$	
PDSIGMA	Normalized particle diameter deviation	1.21	1.15	

 Table 1.
 FLEXPART species parameters for the fine and super-coarse dust particle size bins.

We used the most recent ECMWF Reanalysis v5 (ERA5) atmospheric forcing 268 dataset in our FLEXPART and FLEXDUST simulations, the dataset is described in 269 Hersbach et al. (2020). We used a regional domain cutout of ERA5 extending from 270 10°E to 160°E and 10°N to 80°N, with a horizontal resolution of 30 km (T639) and 271 137 vertical levels from the surface up to 0.01 hPa (80 km). ERA5 data were pre-272 pared and downloaded from the Copernicus Climate Change Serviced Climate Data 273 Store using the Flex extract version 7.0.4 Python package (Tipka et al., 2020). All 274 ERA5 data are available for free from the Copernicus Climate Data store. 275

#### 276 2.3 Model evaluation

Long-term field observations of dust deposition on the CLP suitable for model
evaluation do not exist. Hence, we have to depend on aerosol reanalysis to assess
the performance of FLEXPART and FLEXDUST-EA. Specifically, we use ModernEra Retrospective Analysis for Research and Applications, version 2 (MERRA-2)
aerosol reanalysis produced by NASA's Global Modeling and Assimilation Office

(Gelaro et al., 2017). This dataset has previously been shown reliable at simulating 282 the EADC (H. Liu et al., 2018; W. Yao et al., 2020). Similarly to ERA5, MERRA-2 283 assimilates essential atmospheric variables such as wind, precipitation, and surface pressure. However, unlike ERA5, MERRA-2 additionally incorporates aerosol optical depth (AOD) retrievals sourced from various ground-based and spaceborne instruments, including the Moderate Resolution Imaging Spectroradiometer (MODIS), the 287 Multiangle Imaging Spectroradiometer (MISR), and the Aerosol Robotic Network 288 (AERONET). Assimilation should bring aerosol loading and AOD in MERRA-2 289 closer to observations. AOD represents the total aerosol loading but does not pro-290 vide information on aerosol composition, and therefore the model determines the 291 contribution of dust aerosols to the total AOD. MERRA-2 represent dust aerosols 202 using five size bins (0.1-1.0, 1.0-1.8, 1.8-3.0, 3.0-6.0, and 6.0-10.0  $[\mu m]$ ). Dust emis-293 sion and deposition are not assimilated in MERRA-2; and are in this regard simi-294 lar to FLEXDUST relying on the model physics. The wind patterns of MERRA-2 295 should be similar to those observed in ERA5, however, due to its lower spatial reso-296 lution of 0.675 °longitude and 0.5 °latitude, it is not able to capture local-scale winds 297 as accurately as ERA5. This means that MERRA-2 does not necessarily outperform 298 any other dust model (Gelaro et al., 2017; Zhao et al., 2022). For example, Tang et al. (2023) compared FLEXPART for the mega dust storm in spring 2021 with the two most widely used aerosol reanalysis products MERRA-2 and CAMS. The study 301 revealed that FLEXPART exhibited consistency with MERRA-2 with respect to the 302 strength and timing of the event. However, the overall spatio-temporal patterns of 303 total concentration were more in line with CAMS. We still chose to use MERRA-2 304 as our benchmark because it has a continuous data record dating back to 1980 and 305 covering the entire modeling period of this work. 306

#### 307 3 Results and Discussion

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### 3.1 The spatial distribution and interannual variation of the East Asian spring dust cycle

Our setup using FLEXPART coupled with FLEXDUST-EA was developed 310 to represent East Asian sources and Tang et al. (2023) showed that it can realisti-311 cally simulate the evolution of single dust events, compared to both observations 312 and reanalysis. Here, we evaluated FLEXDUST-EA by comparing it with MERRA-313 2 over a longer time period. Zamora et al. (2022) have also shown a comparison 314 with MERRA-2 products focusing on the Arctic region, where they found mainly 315 differences based on a lack of high-latitude dust sources in MERRA-2. The spa-316 tial distribution of dust emissions in FLEXDUST-EA aligns closely with that of MERRA-2, as illustrated in Figure 3. Both FLEXDUST-EA and MERRA-2 identify 318 key dust emission hot spots in East Asia, including the Taklamakan desert, north-319 western deserts, and the Gobi desert, which is consistent with previous studies (Kok 320 et al., 2021; Shi & Liu, 2011; Shao & Dong, 2006; J. Sun et al., 2001). Moreover, 321 FLEXDUST-EA shows a strong correlation with MERRA-2 regarding the interan-322 nual variation in spring dust emission across all major source regions. Both models 323 show notably lower dust emissions during the springs of 2005 and 2019 and higher emissions during 2001 and 2004. In addition, the period between 1999 and 2009 has 325 greater variability in dust emissions compared to 2010 to 2019 in both FLEXDUST-326 EA and MERRA-2. 327

<sup>328</sup> Next, we evaluate how FLEXPART represents the interannual variation of dust <sup>329</sup> deposition at our sites by comparing the springtime dust deposition simulated by <sup>330</sup> FLEXPART with that of the MERRA-2 grid box closest to our sites. Figure 4a <sup>331</sup> shows the interannual variation of the total spring deposition simulated by FLEX-<sup>332</sup> PART versus MERRA-2, comparing the fine dust size bin (mean diameter 2  $\mu$ m) <sup>333</sup> in FLEXPART with the 1.0-1.8  $\mu$ m size bin in MERRA-2. The models correlate



Figure 3. Normalized mean spring dust emissions (a) FLEXDUST and (b) MERRA-2 between 1999 and 2019. The following dust-emitting regions are indicated: Taklamakan (blue, A), Qaidam Basin (purple, D), sources northwest of CLP (green, E,F,H), Mongolia (orange, G,I), Gurbantunggut Desert (brown, B) and Central Asia (pink), the letters correspond to dust emission regions named in Figure 1. (c) Normalized timeseries of spring dust emissions FLEXDUST (solid line) and MERRA-2 (dotted line), numbers in the upper right corners show the correlation coefficient. (d) The relative dust emission strength of each source region.



Figure 4. (a) Inter-annual variation of total spring deposition simulated by FLEXPART (blue) and MERRA-2 (orange) of the fine dust particle size bin (FLEXPART mean diameter 2.057  $\mu$ m,size bin 2 in MERRA-2, 1.0-1.8  $\mu$ m) for each of the sites at the CLP. The envelope indicates one standard deviation. (b) The ratio of wet deposition to total deposition at each receptor site. The blue bar is FLEXPART and orange bar is MERRA2.(c) Relative distribution of dust deposition between the receptor sites in FLEXPART and MERRA-2.

well for Luochuan, Lantian, and Shapotou, but FLEXPART and MERRA-2 are 334 not correlated at SACOL and Lingtai. SACOL and Lingtai are located close to the 335 mountains in a region with complex topography, which is not well resolved by typical model grids. Additionally, accurately representing precipitation in mountainous 337 regions poses a significant challenge, and different reanalysis datasets may have sub-338 stantial variation in their precipitation rates (J. Yao et al., 2020). Moreover, SACOL 339 and Lingtai experience a high proportion of wet deposition, as shown in Figure 4b, 340 making the differences in precipitation events between ERA5 and MERRA-2 a po-341 tential source of reduced correlation. There is a greater correlation in the dry depo-342 sition rate between MERRA-2 and FLEXPART for all sites (see Figure S1) than in 343 wet deposition. In addition, the wet deposition has a larger interannual variation in 344 FLEXPART compared to MERRA-2. Consequently, wet deposition amplify the differences between FLEXPART and MERRA-2 to total deposition. Finally, modeling 346 dust (wet) deposition is an overarching problem in current dust models, with models 347 showing large differences in the relative importance of wet to dry deposition (Zhao 348 et al., 2022; Huneeus et al., 2011). 349

FLEXPART and MERRA-2 also show differences in the spatial distribution 350 of dust deposition throughout the CLP (Figure 4c). MERRA-2 has a smaller spa-351 tial difference in dust deposition throughout the CLP, while in FLEXPART the two 352 sites close to the source regions make up more than 50% of the combined deposi-353 tion of our sites. Our set-up with backward trajectories limited to 10 days and the 354 comparison of relatively larger sized particles in FLEXPART to smaller particles in 355 MERRA-2 can both contribute to this model inconsistency, but it is also related to 356 the longer lifetime in MERRA-2 compared to FLEXPART (Tang et al., 2023). 357

Although conducting a detailed investigation of the differences in the repre-358 sentation of the EADC in FLEXPART and MERRA-2 is outside the scope of this 359 study. Still, our initial assessment has revealed that the interannual variation in dust 360 emissions in FLEXDUST-EA shown in Figure 3 aligns well with that of MERRA-2, particularly for the important source regions in East Asia. However, total dust de-362 position over the CLP in FLEXPART is not consistent with MERRA-2 at all sites, 363 mainly due to differences in the occurrence of wet deposition events. Evaluating the 364 representation of dust deposition in both MERRA-2 and FLEXPART poses chal-365 lenges due to the limited constraints available on dust deposition in East Asia and 366 the rudimentary nature of dust deposition parameterization in models (X. Zhang et 367 al., 2019).

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# 3.2 Springtime dust transport to the Chinese Loess Plateau: wet versus dry deposition

Figure 5 shows the 20-year averaged spring source contribution maps for each site and both dust particle sizes. The source contribution is normalized by the mean contribution, enabling a relative comparison of the influence of different sources, independent of the deposition rate at the site. The pie chart represents the relative contribution of the source regions as defined in Figure 3 to the total deposition at the site.

Across all CLP locations, Figure 5 shows that the predominant source region that accounts for the majority of dust deposition is the deserts northwest of the CLP. Depending on the site, this source contributes between 48% and 78% of the fine dust and between 47% and 88% of the super-coarse dust. In particular, the source contribution of super-coarse dust in Figure 5 shows a higher contribution from nearby sources compared to fine dust. Fine dust shows a gradual transition from the primary dust sources to less influential ones as one moves away from the receptor location on the source contribution map. The relative source contributions



Figure 5. Averaged spring dust source contribution (1999-2019) of dry- and wet deposition scaled by the mean contribution. The pie chart shows the contribution from each of the source regions as defined in Figure 3.

of fine dust at our CLP locations shown in Figure 5 are consistently similar. For example, at Baode, Luochuan, Lingtai, and Lantian, the order of importance of the different source regions remains consistent: (1) Northwest deserts, (2) Taklamakan, (3) Mongolia, (4) Central Asia, (5) Qaidam Basin, and (6) Gurbantunggut desert. Even at SACOL and Shapotou, which have more distinct source contributions, the main difference lies in an increased contribution from the northwestern dust sources.

In contrast, the source composition of super-coarse dust displays more dis-391 tinct patterns at each location. For instance, super-coarse dust from Shapotou originates primarily from the nearby desert to the northwest of the CLP, with only a 393 minor contribution from Taklamakan. Baode experiences a substantial influx of 394 super-coarse dust from Mongolia, a source nearly absent at other sites. Another no-395 table aspect of super-coarse dust is the trend of increasing influence of dust from 396 the Qaidam Basin across the southeast transect of the CLP. Although the Qaidam 397 Basin contributes modestly to emissions, it becomes the second most influential 308 source at SACOL, Luochuan, Lingtai, and third at Lantian. Central Asia and the Gurbantunggut desert, although not primary sources of dust to the CLP, however, are not negligible at certain sites. For instance, Central Asia contributes to 6-7% of 401 the super-coarse dust deposited at Lingtai and Lantian, emphasizing how even larger 402 dust particles from distant dust emission regions can impact the CLP. 403

The difference between the dry and wet deposition source contribution is shown 404 in Figure 5. The source contributions of wet and dry deposition of fine dust are 405 mostly similar except for dry deposited dust being more influenced by the north-406 western sources, whereas wet deposition has a stronger influence from the more dis-407 tant sources. On the other hand, the source contribution of wet deposited super-408 coarse dust noticeably diverges from the sources of dry deposited dust. The wet 409 deposition source contribution displays a more noisy pattern, indicating less fre-410 quent occurrences of wet deposition for super-coarse dust. The larger variability of 411 the wet-deposited super-coarse dust translates into a more erratic spatial pattern 412 in the wet deposition source contribution compared to the relatively robust pattern 413 observed for dry deposition. 414

The source contribution maps presented in Figure 5 represent the cumulative 415 effects of emissions, transport, and deposition. To pick out the general dust trans-416 port routes to the CLP, we take the centroid trajectory of all the particles released within each 3-hourly particle release and combine them into one dust transporting 418 trajectory by calculating the center of mass of all the trajectories over the whole pe-419 riod weighted by the deposition rate modeled at the time of release. The dust trans-420 port trajectories for all sites and for dry and wet deposition are shown in Figure 6. 421 In the case of fine dust, the trajectories demonstrate minimal variation between re-422 ceptor locations, consistent with relatively uniform source contribution maps across 423 the different sites. Conversely, for super-coarse dust, the transport patterns of drydeposited dust closely resemble those of dry-deposited fine dust, primarily exhibit-425 ing a prevailing northwesterly transport (Figure 6 a, b). An exception is observed 426 for SACOL, displaying a more westerly transport, in alignment with the substantial 427 contribution from the Qaidam Basin. Wet deposition of super-coarse dust exhibits 428 significant variability in transport trajectories between receptors, contributing to the 429 site-dependent nature of super-coarse dust source contributions. A common trend 430 across all sites is that wet-deposited dust is generally influenced by southerly air-431 masses, resulting in overall westerly transport. Given that the source of moisture 432 originates from the south, this pattern is reasonable. Furthermore, the trajectories 433 show an increase in altitude as they approach the receptor, indicating that wet-434 deposited dust typically experiences uplift due to convection initiated by the typical 435 cold air advection observed during a dust event. 436



Figure 6. Weighted average centroid dust loading trajectories by the deposition rate at the receptor at the time of arrival. (a) and (b) are dry deposition dust loading trajectories for fine (mean diameter 2.057  $\mu$ m) and super-coarse dust (mean diameter 17.32  $\mu$ m) particles. (c) and (d) wet deposition dust loading trajectories for the fine and super-coarse dust respectively. The mean height is along the trajectory shown by the coloring and the red dots along the trajectories are equally spaced 12hours apart. The shading underneath show the mean dust emission rate.

Wind speed only changes the mass emitted for dust up to 20  $\mu$ m, but does not 437 change the size distribution at emissions (Kok, 2011). Therefore, the variations in 438 source contribution between fine and super-coarse dust primarily stem from differ-439 ences in their lifetimes. In FLEXPART, we changed two parameters that control 440 dust particle lifetime: the wet scavenging efficiency (assumed to be size-dependent) 441 and the particle size that governs the dry deposition velocity. The impact of differ-442 ent gravitational settling can be seen from the difference in the source contribution 443 between dry-deposited fine and super-coarse dust. To keep the super-coarse dust in 444 the air sufficiently long enough to reach the CLP, either the wind speed has to be 445 sufficiently strong such that the dust arrives before it falls out or the dust has to experience vertical lifting to counter the gravitational settling. 447

Topography has been proposed to increase vertical dust transport. Heisel et 448 al. (2021) utilized high-resolution large-eddy simulations to demonstrate that gen-449 tly sloping terrain can substantially amplify vertical dust transport. The source 450 contribution maps of wet-deposited super-coarse dust showed a distinct band of 451 higher contribution along the southern edge of the Taklamakan. Additionally, it is 452 important to observe that this band of intensified source contribution is less promi-453 nent in the source contribution maps for dry deposition, suggesting that this phe-454 nomenon cannot be solely attributed to heightened emissions along the southern 455 boundary. Moreover, similar to the Taklamakan, the Qaidam Basin, a source sur-456 rounded by sloping terrain, is also a substantial source of wet-deposited super-coarse 457 dust. Hence, potentially suggesting that when super-coarse dust ascends to higher 458 atmospheric levels, it becomes scavenged within the cloud and falls out with the precipitation. Particles of this size readily activate as Cloud Condensation Nuclei 460 (CCN) and rapidly grow to raindrop size. Consequently, a substantial amount of 461 coarse dust could potentially accelerate precipitation onset, mitigating the tendency 462

of continental air with high concentrations of CCN to produce numerous small cloud
droplets (Posselt & Lohmann, 2008).

In summary, there are two prominent pathways for dust transport. The first pathway exhibits a prevailing northwesterly direction, gathering dust primarily from the northwestern desert regions. This pattern is strongly associated with the dry deposition of dust on the CLP. Conversely, the second dust transport pathway, linked to wet deposition, follows a more westerly trajectory. Along this route, topographical features drive dust to higher atmospheric levels, where it is then scavenged within the clouds and subsequently washed out over the CLP.

### 3.3 Drivers of interannual variations in EADC

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Indices	Definition	Reference
Arctic Oscillation (AO)	Leading EOF of the 1000hPa geopotential height anomalies poleward of 20°N	Thompson and Wallace (1998)
East Asian Winter Mon- soon intesity index (EAWMI)	Difference in area averaged 300hPa zonal winds $U_{300}(27.5^{\circ}-37.5^{\circ}\text{N}, 110^{\circ}-170^{\circ}\text{E})-U_{300}(50^{\circ}-60^{\circ}\text{N}, 80-140^{\circ}\text{E})$	Jhun and Lee (2004)
Monsoon Index (MO)	Difference in MSLP between two grid points (105°E, 52.5°N) near Irkutsk and (145°E, 43.75°N) near Nemuro.	Sakai and Kawamura (2009)
Siberian High intensity in- dex (SHI)	Area averaged MSLP (40°-65°N, 80°-120°E)	Wang and Chen (2010)
East Asian Temperature Gradient (EATG)	Gradient in zonally averaged temperature between 30°- 120°E, and taken the averaged gradient between 40° and 50°N	J. Liu et al. (2020)

 Table 2.
 Definition of the climatic indices used in the correlation analysis.

In the following, we utilize the FLEXPART simulations to examine the factors 473 driving the interannual variations in the spring EADC over the past 21 years from 474 1999 to 2019. We establish correlations between dust emission and deposition and 475 various climatic variables representing different aspects of the East Asian circula-476 tion. The indices that we used, along with their definitions, are summarized in Table 477 2. All indices are calculated from monthly ERA5 data for both winter (December-February) and spring (March-May). Decadal trends are removed from the time se-479 ries before calculating the correlations to avoid the influence of low-frequency cli-480 mate variations. 481

The correlations of the winter and spring circulation with spring dust emissions 482 are shown in Figure 7b,c. Figure 7b shows that the circulation of the preceding win-483 ter has a significant impact on the frequency and intensity of dust storms in spring. 484 Emissions from the northwestern deserts are anticorrelated with winter AO. As ev-485 ident in Figure 5 in the previous section, this is the most significant dust source for 486 the CLP. Interestingly, winter AO does not significantly affect emissions from other 487 source regions. Instead, Taklamakan emissions are negatively correlated with the 488 winter monsoon and are most strongly correlated with the Siberian High Intensity 180 Index (SHI), suggesting that the location and strength of the SH are important factors that regulate dust emissions in the Taklamakan desert. Qaidam emissions have 491 correlations similar to those of Taklamakan, albeit weaker. Emissions from Central 492 Asia and Gurbantunggut are not strongly related to the winter circulation of East 493 Asia. For the spring circulation shown in Figure 7c, dust emissions in Mongolia and 494



Figure 7. (a) Correlations between winter (December-January-February, DJF) and spring (March-April-May, DJF)circulation indices and modeled spring dry- and wet deposition of supercoarse dust, for the whole domain and contributions from individual source regions. (b-c) Correlation with FLEXDUST-EA dust emissions in the source regions and with winter and spring circulation respectively. The size of the circle indicate the strength of the correlation.

northwestern deserts are more strongly correlated with the magnitude of the East
Asian Temperature Gradient (EATG), that is, a large negative temperature gradient favors larger emissions. Taklamakan emissions do not show a significant correlation with any particular spring circulation index. Qaidam is similar again to Taklamakan except that it is more positively correlated with AO. The intensity of the SH
in spring is significantly anti-correlated with emissions from Central Asia and Gurbantunggut.

Figure 7a shows the correlations between source contributions of super-coarse dust, delineated by contributions from each dust emission region to each site. Looking at the winter circulation, we observe significant negative correlations between the AO and dry deposition at Shapotou, SACOL, Baode, and Luochuan. Additionally, AO is negatively correlated with wet deposition at Lantian and SACOL. The correlations with AO are most pronounced for source contributions from Mongolia and the northwestern deserts, although they vary by site.

Interestingly, wet deposition at SACOL demonstrates significant correlations 509 with all winter circulation indices, including AO and MO, both of which are linked 510 to the EATG. Additionally, the SHI correlates with both AO and MO, even though 511 AO and MO do not exhibit a significant correlation with each other (see Supplement 512 Figure S3). Considering these insights, we infer that, for wet deposition at SACOL, 513 the strength of EATG in winter plays a critical role in creating favorable conditions 514 for wet deposition in spring, mainly sourced from the northwestern deserts. How-515 ever, for dry deposition at SACOL, AO is the only significantly correlated index, 516 aligning with AO's correlation with emissions from the northwestern sources, the 517 primary dust source for SACOL. 518

The strength of the EAWM is correlated with dust from Qaidam and Gurban-519 tunggut to the SACOL site. The spring EAWM circulation features are correlated 520 with an increased contribution from the Gurbantunggut desert to Lantian, Lingtai, 521 and Luochuan. The deposition of Central Asian dust at the sites is generally not 522 correlated with any of the circulation indicators, except at Baode site, which shows a 523 significant negative correlation between the contribution of dry deposited dust from 524 Central Asia and the SHI, where the SH can act as a barrier for long-range dust 525 transport. 526

The correlations presented above are for the super-coarse dust size bin. Fine 527 dust correlations are provided in the Supplement (Figure S4). The correlations be-528 tween the winter AO circulation and spring deposition remain consistent across fine 529 and super-coarse dust. However, in terms of spring circulation, the two particle sizes 530 are less consistent. Dry-deposited fine dust displays notably higher correlations with EATG and EAWMI, especially for dust transport from Taklamakan compared to 532 super-coarse dust. For the north western deserts fine and super-coarse dust exhibit 533 similar correlations with EATG. The correlations between circulation patterns and 534 wet deposition for fine dust are highly variable and strongly dependent on the site, 535 and this does not change from what is shown in Figure 5. This underscores that wet 536 deposition is the result of an intricate dynamic between the source, receptor, and 537 circulation modes. 538

To interpret the relationship between atmospheric circulation patterns and 539 dust deposition over the CLP, as revealed in the previous correlation analysis shown 540 in 7, we performed a composite analysis of years with weak and strong deposition 541 at sites, to identify site-dependent circulations features that typically cause an in-542 crease in dust deposition. Figure 8 shows the composite differences of 850hPa winds 543 and mean sea level pressure (MSLP), hatched areas show the regions where the dif-544 ferences between the strong and weak dust deposition years are significant. In the 545 case of dry deposition of super-coarse dust, all sites manifest AO-like composite dif-546



Figure 8. Composite difference of mean sea level pressure and 850hPa wind strong minus weak deposition years for super coarse dust for both winter (December-January-February, DJF) and spring (March-April-May, MAM) for all the locations. The left half shows composite difference based on dry deposition rate, while the right half is the composite difference based on total deposition rate. The hatched area indicates where the anomalies are significant (p < 0.05)

ferences during the winter preceding a strong deposition year. These anomalies show characteristic positive MSLP anomalies over the Arctic, typical of negative AO-like conditions (Thompson & Wallace, 1998; He et al., 2017). This AO-like pattern extends into the spring for most sites. However, for total deposition at Shapotou and Luochuan, the AO pattern is masked by other influences, as wet deposition is more event-dependent and less influenced by the overarching circulation state. A longer time series of simulations would be able to better distinguish the more random fluctuations due to wet deposition from the impact of large-scale circulation.

Figure 9 shows the composite anomalies of the geopotential height and winds at 500hPa for super-coarse dust. During the winter preceding a strong deposition year, it indicates an area of reduced geopotential height that for Lantian, Lingtai, Luochuan and Baode is located to the north west of the site, while for Shapatou and SACOL negative geopotential height anomaly is located more to the north east. This suggests that increases in dust deposition as a result of dry deposition at these sites are closely associated with the location of this geopotential height anomaly and the corresponding changes in the wind patterns.

During the negative phase of AO, there is usually an increase in the frequency 563 and intensity of cold air outbreaks (COAB) in East Asia (He et al., 2017; Yang et 564 al., 2020). H. Liu et al. (2018) conducted a detailed analysis investigating the rela-565 tionship between spring dust emissions and the preceding winter AO using MERRA-2 reanalysis. Using regression analysis, they revealed a correspondence between winter 500 hPa geopotential height and the leading empirical orthogonal function of 568 spring dust emissions, similar to the composite anomalies we found shown in Figure 569 9. H. Liu et al. (2018) suggested that a negative AO results in anomalous cold con-570 ditions in central Siberia, which persist into the ensuing spring, amplified by snow-571 albedo and cloud feedbacks. The resultant colder temperatures in the inner regions 572 of East Asia create a more negative temperature gradient in the region, intensify-573 ing the East Asian Trough (EAT). This in turn creates a favorable environment for dust emissions. Their explanation of how winter AO influences spring surface tem-575 perature aligns with our correlation analysis, where a negative AO corresponds to a 576 more pronounced temperature gradient over East Asia, elevating dust transport to 577 the CLP from the northwestern sources. 578

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#### 3.4 Implications for interpreting Chinese Loess Plateau dust records

Our results reveal a clear link between the depositional mechanism and the dust-emitting regions. This observation may have major implications on how we in-581 terpret the provenance of the CLP dust records (Quaternary loess and Neogene red 582 clay) also on geological time scales. The CLP dust provenance studies often con-583 sider changes in emission and transport mechanisms, but commonly overlook the fi-584 nal step of the EADC, the deposition. Our results on the deposition of both fine and 585 super-coarse particles indicate that wet deposition contains more dust from west-586 ern emission areas than dry deposition (Figure 6), suggesting the shift in CLP dust 587 provenance may not only be related to changes in dust transport pathways but also the deposition mode. The CLP dust records are associated with coarser grain sizes 589 during glacials compared to interglacials. These differences have been explained by 590 higher dust influx and reduced soil development coupled with stronger wind speeds 591 during glacial periods (Maher, 2016; Kohfeld & Harrison, 2003). However, our re-592 sults imply that increased wet deposition of fine/clay-sized dust during the wetter 593 interglacials could provide an additional explanation for the observed grain-size dif-594 ferences between glacials and interglacials. Similarly, the finer grain size of late Neogene red clay deposits compared to Quaternary loess may be associated with an in-596 crease in the wet deposition of finer particles during intensified EASM conditions 597 (Ren et al., 2020; Z. L. Ding et al., 1999). 598



Figure 9. Circulation composite anomalies of 500hPa wind strength (colored, unit m/s), wind direction (vectors, unit m/s) and geopotential height (contours, unit dam, distance between contours 2 dam, solid contour are positive anomalies while dotted contours are negative anomalies) for winter (December-January-February, DJF) and spring (March-April-May, DJF) of strong - weak dry and total deposition years. The hatched area indicates where the anomalies are significant (p < 0.05)

The results of our model, which covers the full dust cycle (i.e., emission, trans-599 port, and deposition), may also have important implications on the observed spatial 600 differences in source contribution across the CLP. Previous single-grain provenance 601 studies (targeting silt-sized particles) have documented Neogene and Quaternary dust source differences between the northeastern (e.g., Baode site) and the south-603 western (e.g., Lantian) CLP (Shang et al., 2016; H. Zhang et al., 2021; W. Peng et 604 al., 2023; Bird et al., 2015). These observations were supported by backward tra-605 jectory modeling showing more dominant atmospheric transport pathways from the 606 Taklamakan desert and Northern Tibetan Plateau to the southwestern CLP than to 607 the northeastern CLP (Shang et al., 2016). The results of our model, taking into ac-608 count the full EADC, further corroborate these conclusions (Figure 5). In addition, spatial differences in dust provenance have been found to be particularly prominent 610 in coarse dust particles and wet deposited dust (Figure 5). This offers a plausible 611 eolian mechanism for explaining the spatial provenance contrast of the red clay and 612 loess deposits, and its temporal variations, in addition to the well-established hy-613 pothesis emphasizing the importance of large fluvial systems (namely the Yellow 614 River) in providing dust particles closer to the CLP, and therefore in creating the 615 observed spatial provenance gradient (Stevens et al., 2013; Bird et al., 2015; Nie et 616 al., 2015; W. Peng et al., 2023; H. Zhang et al., 2021; Sun et al., 2022). For instance, 617 W. Peng et al. (2023) found an increase in Pliocene spatial provenance contrast in 618 the Red Clay, and argued that this was partly caused by the intensified Pliocene 619 EASM. This argument is well in line with our results, since the intensified EASM 620 can enhance the wet deposition flux over the CLP. Therefore, an increase in wet de-621 position can act to amplify the spatial provenance differences created primarily by 622 the fluvial input of material. 623

Our modeling results with super-coarse/silt-sized dust particles indicate that 624 even large dust particles deposited on the CLP may be sourced from distant dust-625 emitting sources (e.g., Taklamakan) although they are not the dominant contribu-626 tor to the deposition on the CLP. Larger dust particles (super-coarse or larger) de-627 posited far from the dust emission region have also been reported in recent observa-628 tional studies (Van Der Does et al., 2018; Ryder et al., 2018; A. A. Adebiyi & Kok, 2020). The fact that we also find reason to believe that super-coarse dust deposited 630 over the CLP can be from the distant dust-emitting regions provides additional sup-631 port to the validity of the single-grain provenance proxy studies using mainly the 632 large mineral particles in detecting the source origin of emitted dust. Strengthen-633 ing claims and insights on loess formation mechanisms and climatic implications 634 made based on the coarse dust fractions in the loess deposits. We note that spheri-635 cal particles are assumed in our model simulations, thus might be considered a lower 636 bound of the long-distance transport of super-coarse dust to the CLP. Further modeling studies using a-spherical particles with lower settling velocities and potentially 638 longer transport distance than spherical particles may help to better understand the 639 long-distance transport of large dust particles. 640

The interannual variation of the spring dust deposition rate over the CLP has 641 been found to be more closely associated with AO than with the strength of the 642 EAWM in both winter and spring (Figure 7 and S4). This is distinct from the com-643 mon understanding within the paleoclimate communities which emphasizes the im-644 portance of EAWM in regulating the dust cycle over the CLP (e.g. An et al., 2014). 645 However, consistent with studies of the present-day interannual variation of the EADC, 646 which generally support the strong control of AO instead of EAWM on the variation 647 of EADC (Mao et al., 2011; H. Liu et al., 2018), with the negative AO being favor-648 able for stronger dust storms in East Asia. Although the results are based on the annual time scale, they may have strong implications for understanding long-term 650 changes in the EADC and the interpretation of the red clay and loess record, espe-651 cially when it is difficult to use the strength of EAWM to explain the dust records 652

and its variations. For instance, provenance research on the dust records deposited 653 during the warmhouse Eocene indicates the absence (or significantly weaker) SH 654 coupled with possible long-term negative phase of AO -like conditions (Bohm et al. 655 2023X, in revision). Moreover, it has been noticed that the cyclicity of the oxygen isotopes of speleothem records over East Asia, an indicator of East Asian summer 657 monsoon, is dominated by precession cycle (20 ka) on orbital time scale, which is in 658 contrast to the loess records, which reveal a dominance of the glacial-interglacial cy-659 cle (100 ka) (Cheng et al., 2021). The larger influence of AO-like conditions (over 660 EAWM) on the variation of dust deposition over the CLP during the glacial-inter-661 glacial cycle, may offer a plausible explanation of the "apparent" discrepancy be-662 tween loess and speleothem records from East Asia. This explanation is consistent with the fact that high-latitude forcing is the dominant influence on Loess records (Stevens et al., 2018). However, on the basis of this work, it is not possible to con-665 clude whether this is generally the case because the climatic conditions during the 666 glacial stages are very different from those of today. Although, this could be an-667 swered by driving this modeling setup using data from paleoclimate simulations in the future. 669

#### 670 4 Conclusion

To our knowledge, this is the first modeling study to provide a detailed picture 671 of how proximate and distant dust emission areas contribute to dust accumulation 672 on the CLP. We provide evidence for the importance of considering wet deposition 673 when interpreting the loess records, showing that wet-deposited dust differs in both 674 the typical transport trajectory and the source of the emitted dust. Even super-675 coarse dust could be wet deposited at the CLP where sloping topography upwind 676 of dust emission area can act to increase the vertical dust transport substantially, making it possible for even large dust particles to be scavenged inside clouds. This 678 could be an important mechanism for bringing potentially non-negligible amounts 679 on of coarse dust particles from more distant sources like the Taklamakan desert and 680 Quaidam basin. Dry-deposited dust is mainly sourced from the nearby dust-emitting 681 areas to the north-west of the CLP, this transport pathway is favorable under nega-682 tive AO-like conditions. That the AO is an important driver of the EADC is in line 683 with our understanding of the EADC under present day climate conditions (H. Liu 68/ et al., 2018; Mao et al., 2011; D.-Y. Gong et al., 2006). This study provides a direct link between AO and dust accumulation on the CLP. Our link between AO and dust accumulation on the CLP was established based on current climate conditions, 687 which were different in the past, but it does warrant a stronger emphasis on high-688 latitude forcings when explaining the variability in dust accumulation rate of the 689 loess record rather than the common partitioning of enhanced summer monsoon or 690 winter monsoon conditions, as also suggested by Roe (2009). 691

FLEXPART has also been shown to be a very useful tool for understanding 692 the dust cycle, as it can pinpoint the exact dust source contribution of a receptor 693 site. The FLEXPART ES is independent of the dust emission inventory, so FLEX-694 PART can be used to easily test the effect and impact of different emission schemes 695 and hypothesis of changes in dust emission regions in the past. A promising future application of this setup would be to combine FLEXPART and state-of-the-art pale-697 oclimate model reconstructions to give insight into how environmental changes have influenced the source region and dust transport mechanism at the CLP. This would 699 be invaluable in formation to further aid in interpretation of the CLP provenance 700 proxies and help to understand how the EADC might change in the future. 701

- 702 Acronyms
- 703 CAOB Cold air outbreaks
- **EAWM** East Asian Winter Monsoon
- 705 CLP Chinese Loess Plateau
- 706 AO Arctic Oscillation
- 707 LPDM Lagrangian particle dispersion model
- 708 MAR Mass accumulation rate
- **EOF** Empirical orthogonal functions
- 710 MSLP Mean sea level pressure
- 711 DJF December January February
- 712 MAM March April May
- **EADC** East Asian dust cycle
- **ES** Emission sensitivity
- **515 SHI** Siberian High Intensity index
- **EATG** East Asian Tempereature Gradient
- 717 MO Monsoon index

#### 718 Acknowledgments

This work was supported by the Research Council of Finland (grant no 316799). 719 The simulations were performed on resources provided by Sigma2 - the National In-720 frastructure for High Performance Computing and Data Storage in Norway, project 721 no. nn2806k. The MERRA-2 data used in this study have been provided by the 722 Global Modeling and Assimilation Office (GMAO) at NASA Goddard Space Flight 723 Center and are available from NASA GES DISC (https://disc.gsfc.nasa.gov/ 724 datasets?project=MERRA-2 last access October 2021). ERA5 data used in this 725 study were downloaded using the Copernicus Climate Change Service (C3S) Climate 726 Data Store. Neither the European Commission nor ECMWF is responsible for any 727 use that may be made of the information it contains. 728

The FLEXDUST-EA source code is available at https://github.com/huitang 729 -earth/FLEXDUST/tree/FES2022\_Tangetal\_updated\_KOK14, and the FLEXPART 730 codes are available at https://github.com/Ovewh/flexpart/tree/release-10. 731 The workflows for setting up the FLEXPART simulations are available on github: 732 https://github.com/MasterOnDust/FLEXPART-script/releases/tag/ ) The work-733 flow for post-processing the raw simulation output and combining the FLEXDUST 734 emission inventory with FLEXPART ES is available on GitHub (https://github .com/MasterOnDust/FP\_preprosses\_workflow/tree/AGU-Haugvaldstad-et-al 736 -2023). The workflow to generate the figures shown in this manuscript is available 737 in the following GitHub repository:https://github.com/MasterOnDust/AGU\_JGR 738 \_CLP\_SOURCE\_WORKFLOW/tree/AGU-Haugvaldstad-et-al-2023 . The data files re-739 quired to generate the figures shown in the manuscript are available from Zenodo 740 Haugvaldstad et al. (2023) https://doi.org/10.5281/zenodo.10114436. The 741 full 3hourly FLEXPART source contribution, emission sensitivity and FLEXDUST 742 dust emission inventory are archived on the Norwegian Research Data Archive; see 743 (DOI: https://doi.org/10.11582/2023.00134, https://doi.org/10.11582/ 744 2023.00135), Haugvaldstad (2023a, 2023b). 745

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# Supporting Information for Spatial source contribution and interannual variation in deposition of dust aerosols over the Chinese Loess Plateau

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# Contents of this file

1. Figures S1 to S8

**Introduction** This document contains additional figures that did not make it to the final manuscript. But they are included in the supplement as supporting information for the analysis presented in the manuscript.



Figure S1. Interannual variation of simulated dry (a) and wet (b) dust deposition of fine dust at each site, FLEXPART (blue line) MERRA-2 (orange line) (FLEXPART: mean radius 2.057  $\mu$ m, MERRA-2 dust size bin 2 1.0-1.8  $\mu$ m). The envelope indicate one standard deviation.



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**Figure S2.** Histogram of daily dry and wet deposited dust for both fine and super-coarse sized dust at the six sites located on the CLP. The spring is split into a late (blue) and early (orange) spring period (late spring after the 15th of April).



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**Figure S3.** Correlation of circulation indices with each other for winter(December-January-February, DJF) and spring (March-April-May, MAM). The significant correlations are shown in the figure.

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Figure S4. (A) Correlations between winter (December-January-February, DJF) and spring (March-April-May, MAM) circulation indices and spring dry- and wet deposition rate of fine dust modeled by FLEXPART, for the whole domain and contributions from individual source regions. (B-C) Correlation with dust emissions in the source regions and with winter and spring circulation respectively. The size of the circle indicate the strength of the correlation. November 22, 2023, 8:12am



Figure S5. Composite of anomalies based on the difference between strong and weak dust dry-deposition years for winter (December-January-February, DJF) and spring (March-April-May, MAM), of mean sea level pressure and 850hPa winds. Hatched areas as significant.

Dry deposition DJF - 2 m/s

90°N

N°00 SHAPOTOU 30°N 30°N Dry deposition MAM- 2 m/s Total deposition DJF - 2 m/s Total deposition MAM- 2 m/s Total deposition



**Figure S6.** Composite of anomalies based on the difference between strong and weak dust dry-deposition years for winter (December-January-February, DJF) and spring (March-April-May, MAM), of 500hPa wind speed (shading) and geopotential height (contours, unit dam, distance between contours 2 dam, solid contour are positive anomalies while dotted contours are negative anomalies). The distance be Hatched areas as significant.


**Figure S7.** Circulation composite anomalies of 850hPa winds (vectors, unit: m/s) and mean sea level pressure for strong (colored, unit: Pa) - weak winter monsoon years and negative - positive winter AO. (a) and (b) is the DJF anomalies and (c) - (d) is MAM anomalies in the following spring. Data from ERA5.





**Figure S8.** Circulation composite anomalies of 500hPa winds strength (colored, unit m/s), wind direction (vectors, unit m/s) and geopotential height (contours, unit dam, distance between contours 2 dam) for strong - weak winter monsoon years and negative - positive winter AO. (a) and (b) is the DJF anomalies and (c) - (d) is MAM anomalies in the following spring. Data from ERA5.