Source areas of the Eastern Ebro Valley loess (NE Iberian Peninsula): Heavy mineral composition as a provenance indicator

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Abstract
Loess deposits of the Eastern Ebro Valley (NE Iberian Peninsula) occupy an area of ~2000 km² mostly on the east of the Ebro depression and locally in the Móra Basin. We studied 38 samples of loess and 47 samples of their possible source areas, in order to determine the location of the origin of the loess materials, based on their particle size distribution and composition of heavy minerals. Our results clearly differentiate two depositional loess basins disconnected by the reliefs of the Pre-Coastal Range with different mineralogical signatures: the Ebro Basin and the Móra Basin. The most relevant variables to discern source areas were the percentage of heavy minerals, the concentrations of zircon, staurolite, rutile and inosilicates, and the presence of gypsum. In the Ebro Basin, the largest set of loess deposits is located around a depocenter in the Batea region and along several lateral areas, with materials that come from the central alluvial plain of the Middle Ebro River (25 to >100 km travel distance), in addition to deflated materials from Miocene interfluvial substrates. On the other hand, loess deposits of the Móra Basin come from more proximal source areas (<15 km), originating from the alluvial materials of the Lower Ebro River that contain the contributions of its tributaries, the Cinca and Segre rivers. Taking into account the homogeneity in the mineralogical composition of complex loess sequences, we deduced a general WNW–ESE wind direction, which would have remained constant over the last two cold periods (MIS-2 and MIS-6). Along this axis, the Pàndols–Cavalls Range, with a height difference of 200–350 m, acted as a wind barrier separating the loess of Ebro Basin from the loess of Móra Basin.

KEYWORDS
Ebro Valley, heavy mineral analysis, paleowind direction, Quaternary aeolian deposits, tracer minerals, travel distance

1 | INTRODUCTION

Loess is an aeolian deposit that covers large areas of the Earth’s surface (Muhs, 2013). It has been the subject of research by European scientists from the first half of the 19th century onwards (Smalley & Rogers, 1996). Besides the agronomic interest of soils developed on loess (Smalley et al., 2001), these deposits have a high scientific value from the paleoenvironmental point of view, since they represent very useful terrestrial sedimentary records for reconstructing Quaternary paleoclimates and the evolution of paleoenvironmental scenarios linked to them (Marković et al., 2015; Nie et al., 2015).

Loess covers are very well represented in Northern and Central Europe, where they form significant, thick, continuous sedimentary assemblages. Among them, the extended Northern European Loess Belt is related to the activity of the Fennoscandian and British Isles Ice Sheets. Other extensive deposits, in the middle basins of large...
rivers (Rhine, Danube, Rhône, Po), have their sources in the Alpine glacial system. In addition to the ice sheets and Alpine system, fluvially transported material coming from low mountain ranges also plays a major role in loess formation. Besides the glacier and fluvial-fed systems, other less well-developed loess areas appear at the periphery of aeolian sandsheets and dunefields, particularly along the Atlantic coasts (coastal systems) and also inland, in association with easily erodible detrital rocks such as sandstones, where strong winds are generated by relief (continental systems) (Bertran et al., 2016; 2021; Haase et al., 2007; Lehmkühl et al., 2021). In contrast, in Southern Europe (i.e. in the Iberian Peninsula, Southern France and along the Adriatic coast), loess deposits have a smaller extent and are mostly discontinuous (Costantini et al., 2018; Coudé-Gaussen, 1990). In these areas, smaller glacial systems (Pyrenees, Iberian System, Dinarides) are believed to be responsible for feeding rivers (Bertran et al., 2021; Wolf et al., 2019). The mass of available materials that form these loess deposits is largely due to glacial abrasion processes. It was much more active in areas covered by large masses of ice, which later became the sources of particulate materials (Bertran et al., 2021).

Recent studies propose several genetic models based on the climatic and geomorphological characteristics of loess source areas (Lehmkuhl et al., 2021), especially the abrasion capacity of glacial systems and the type of connection with subsequent fluvial and aeolian transport processes (Bertran et al., 2021; Li et al., 2020). All these models consider combined transport systems that start in glacial environments as the main mechanisms to produce fine particles of sand and silt, which would later be taken up and reworked by river drainage networks. They would finally be transported by winds from alluvial areas towards annexed landscapes at a regional or continental scale. This interpretation has led to the idea that the alluvial plains of large rivers are amongst the most appropriate systems for acting as potential source areas of wind-transported particles (Lehmkuhl et al., 2016; Muhs, 2007; Smalley et al., 2009).

Although there are some precedents (Solé-Sabaris et al., 1957), loess deposits of the Iberian Peninsula were not described and analysed in depth until a few decades ago. The most important loess assemblies correspond to the large Cenozoic basins of the Ebro River (Boixadera et al., 2015; Iriondo & Kröhling, 2004; Poch et al., 2021) and the Tagus River (Calvo et al., 2016; García-Giménez et al., 2012; Wolf et al., 2019). In the Ebro and Tagus loess areas, the average grain size ranges from coarse silt to fine sand. They comprise accumulations 2–8 m thick, with discontinuous and irregular outcrops exhibiting high carbonate contents (35–60%). Extension and location of the deposits are influenced by aerodynamic factors related to regional orography and katabatic wind adaptation. Other sets of loess deposits have been described at scattered locations on the Iberian Peninsula (La Mancha, Girona), but they are much smaller entities than the previous ones (Mücher et al., 1990).

The relationships between fluvial network, regional wind systems and loess accumulations will help us to know the dust provenance areas, main transport direction, travel distance and orographic effect of surrounding landscapes. Hence, the paleogeographic and paleoenvironmental reconstruction of the Ebro Valley would allow a better understanding of climatic and geomorphological dynamics (fluvial, alluvial, aeolian, weathering) operating in the region during the last phases of the Quaternary.

To establish the loess source areas, various techniques have been developed based on the recognition of specific or tracing characteristics in both sediments and source areas. The techniques consisted of the identification of heavy minerals (Chmielowska & Salata, 2020; Muhs et al., 2018; Peng et al., 2016; Thamó-Bozsó et al., 2014; Wolf et al., 2019), geochemical composition (Bosq et al., 2020; Schatz et al., 2015; Waroszewski et al., 2021) and U–Pb isotope analysis of zircons (Fenn et al., 2022; Uyúri et al., 2016; Wolf et al., 2021). One of the most useful and effective methods is the comparison of sets of heavy minerals present in the loess sand fraction and their potential source materials: fluvial terraces and regional lithological substrates (Chmielowska & Salata, 2020; Peng et al., 2016; Thamó-Bozsó et al., 2014; Wolf et al., 2021). As ancillary techniques, particle grain size analysis of the loess and original materials, and analyses of the general chemical composition, are also used. The main criteria employed in the interpretation of the possible origin of aeolian deposits are: (a) the average size of the grains is related to the mode of transport—saltation, suspension at low or high altitude and the distance of transport, and understanding that coarse fractions correspond to nearby (local) contributions; and (b) the mineralogical nature of particles, in which changes in composition can be interpreted as changes in wind direction or in the source of deflated materials (Wolf et al., 2019). Recent studies on loess origin (Bosq et al., 2020; Waroszewski et al., 2021; Wolf et al., 2019) have found that the alluvial plains of large rivers and the sedimentary substrate and soils of interfluvial areas are the preferential zones for the mobilization of loess particles (Römer et al., 2016; Smalley et al., 2009).

Previous studies on loess of the Eastern Ebro Valley (Boixadera et al., 2015; Plata et al., 2021) show coarse sizes of loess particles indicating proximal provenance. Our research aimed to identify source areas of the aeolian materials from Zaragoza to the east, the prevailing wind direction during loess deposition, possible transport distances and the role of geographical features in the separation of depositional areas in the Ebro Valley.

The working hypothesis was based on accepting three assumptions: (a) source areas are relatively close when grain sizes are large (Pye, 1995); (b) the mineralogical signature of loess is inherited from that of the wind-transported sediments and thus can be determined with techniques of separation and identification of heavy minerals (Thamó-Bozsó et al., 2014); and (c) alluvial plains of large rivers may be a potential source of particles (Lehmkuhl et al., 2016; Smalley et al., 2009). Techniques of cartography and field description, particle size analysis, mineralogical separation and identification were combined to carry out our research.

## 2 | STUDY AREA

The Ebro River Valley, located in the NE of the Iberian Peninsula with a drained area of about 85 000 km², is one of the largest basins in the Western Mediterranean. The triangular shape of the river basin corresponds almost exactly to the former Cenozoic foreland basin located between the Pyrenees, the Iberian Range and the Catalan Coastal Ranges. It initially acted as a marine basin open to the Atlantic Ocean between the Paleocene and the Eocene. Between the Upper Eocene and the Upper Miocene, it became an endorheic continental basin. The timespan of onset of exorheism and, therefore, of the erosive emptying
of this depression towards the Mediterranean Sea ranges from the Middle Miocene (Arche et al., 2010; García-Castellanos et al., 2003) to the Pliocene (Babault et al., 2006). Sediments of the centre of the Ebro Valley are basically Miocene evaporitic sequences in the western zone (evaporites around Zaragoza), whereas they are detrital series of Oligocene and Miocene sandstones, marls, lutites, lacustrine limestones and more rarely evaporites in the eastern zone between Sástago and Mequinenza (Solà & Costa, 1997) (Figures 1 and 2). The Middle Ebro (Ollero, 1996) drains the innermost area of the Iberian Range and the Western Pyrenees, where metamorphic, quartzitic and carbonated materials predominate. In the Eastern Pyrenean axial zone, extensive granodiorite batholiths (Maladeta and Andorra) crop out, surrounded by metamorphic slate assemblages. They are drained by the valleys of the Cinca and Segre rivers (northern tributaries of the Ebro River in the Central and Eastern Pyrenees).

The bottom of the Ebro Valley (Figure 2) has well-developed Quaternary river terrace levels with a maximum width of 10 km. Downstream of Sástago, the course begins to have a noticeable increase in meanders, 400–600 m wide, and it incises into the valley, forming canyons. The Cinca and Segre rivers have a 2–3 km-wide terrace system. The Móra Basin (Figures 1 and 2) is a small graben located in the Catalan Coastal Ranges, near the contact with the Ebro Basin delimited by normal faults active during the Neogene (Teixell, 1988).

The graben contains Oligocene, Miocene and Plio-Pleistocene detrital sediments.

The climate is Continental Mediterranean, with little rainfall and extreme temperatures that make the central area of the basin (Los Monegros) the northernmost arid area in Europe (Valero-Garcés et al., 2005). Annual rainfall can reach 1500 mm in the Pyrenees but drops sharply to <400 mm in the centre of the valley, due to the shadow effect of the Iberian Range. Annual evapotranspiration can exceed 1200 mm. The winds are dry and strong with a WNW–ESE direction, locally known as ‘cierzo’ (Gutiérrez et al., 2013; Mensua & Ibáñez, 1975; Puicercús et al., 1997).

The climate underwent major changes in the region during the Quaternary, with the alternation of cold and warm climates (González-Sampériz et al., 2008). On a global scale, precipitations \( P \) were lower during the glacial periods because of lower sea temperatures. However, evapotranspiration \( E \) was also weaker on the continent. Over most of Spain, climate modelling suggests higher soil moisture during the glacial period of the Last Glacial Maximum because of the much higher \( P-E \) budget (Scheff et al., 2017; Strandberg et al., 2011). These changing environments led to a succession of complex morphogenetic systems. In cooling periods, glacial and periglacial processes dominated in mountain areas and a combination of alluvial aeolian environments were developed at lower

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**Figure 1** Geological map of the Ebro Valley. The location map (a) shows the Ebro Valley in green and the study area in yellow. The main map (b) is derived from Pardo et al. (2004) and the geological map of Spain 1:1 000 000 from Rodríguez et al. (2015). Coordinate system UTM 31 N. [Color figure can be viewed at wileyonlinelibrary.com]
altitudes (Rodríguez-Ochoa et al., 2019). On the other hand, warmer periods enhanced the activity of fluvial and alluvial systems, linked to the disappearance of glacial and periglacial environments.

The loess of the Eastern Ebro Valley, the largest loess area in Iberia, comprises two groups of aeolian assemblages (Boixadera et al., 2015; Plata et al., 2021). The most important, referred to in this work as Ebro Basin loess, occupies a large area in the southeast of the Ebro Valley, very close to the contact with the reliefs of the Catalan Coastal Range. The Ebro Basin loess is located mainly south of the Ebro River, bounded on the north by the towns of Chiprana, Mequinenza and Ascó; and to the south by Calaceite and Gandesa (Figure 2). It occupies a 1500–2000 km² area on carbonate platforms and E–SE sides of valleys, at altitudes between 150 and 450 m a.s.l. The centre of the deposition area is around the village of Batea. The other group is known as the Morà Basin loess and crops out in a small area of about 150–200 km² east of the Ebro Basin loess, clearly separated by the positive reliefs of the Catalan Pre-Coastal Range, also called the Catalana Prelitoral Range (represented by the Pàndols and Cavalls ranges, 705 m a.s.l.).

From a paleogeographic point of view, the loess of the study area have been classified in the domain of the Western Mediterranean loess (Lehmkuhl et al., 2021) and considered as small continental systems in Cenozoic basins (Bertran et al., 2021). Their dominant grain size fraction is very fine sand followed by coarse silt, so they have been classified as sandy loess in the USDA textural classification (Bertran et al., 2021; Coudé-Gaussen, 1990). Their average mineral composition is dominated by quartz (40–45%) and calcite (28–45%). Organic matter is very scarce (average 0.35%). Deposits are usually 2–5 m thick, although in very favourable positions they can reach 10 m. Their high lateral discontinuity makes them very difficult to map at regional scales. Most of the deposits correspond to very young sequences, dated between 17 and 34 ka with OSL techniques, that have been related to cooling phases of the MIS-2 stage. Occasionally, older complex sequences have been identified in Mas de l’Alerany and Chiprana profiles, with ages between 130 and 144 ka with the post-IRSL method, and thus they would be related to stage MIS-6 (Boixadera et al., 2015; Dominik et al., 2021; Plata et al., 2021).

3 | METHODOLOGY

Identifying the provenance of the aeolian materials that form the loess deposits of the Eastern Ebro Valley has been based on the statistically tested similarity of (1) the grain size, (2) the mineralogical signature and (3) the chemical properties between the source area and the loess deposits. In the case of grain size, the degree of particle size sorting and fractioning caused by the wind deflation and transport plays a major role (Bertran et al., 2021; Bosq et al., 2018; Crouvi et al., 2008). The heavy mineral analyses compare the presence and frequency of representative mineral species in sources and deposits (Chmielowska & Salata, 2020; Römer et al., 2016; Song et al., 2014; Thamó-Bozsó et al., 2014; Wolf et al., 2019). Calcium carbonate and
gypsum contents have also been selected as common chemical components. Their presence in loess deposits can be attributed to a potential contribution of Miocene interfluvial lithologies. Gypsum in loess can be related to the evaporitic sequences of Miocene formations near Zaragoza as a major source (Poch et al., 2021).

We analysed the grain size distribution of 47 samples of the potential source areas: 42 from fluvial terraces and 5 from Miocene substrates (Supplementary Table S1). The fluvial sediments were collected along 350 linear km of Ebro, Cinca and Segre river terraces (Figure 2). These terraces are 8–20 m higher than the present-day talweg, and have ages around 47 ± 4 ka, obtained by correlation from the dated Cinca and Segre lower terrace levels (Lewis et al., 2009, 2017; Stange et al., 2013); therefore, they would correspond to the floodplain levels contemporary with the formation of the loess deposits (Boixadera et al., 2015; Plata et al., 2021). The Miocene substrates consist of sandstones, limestones and marls, representative of the main lithostratigraphic units located east of Zaragoza (Figure 2). Potential Miocene sand sources correspond to sandstone paleochannels with a much reduced surface outcropping.

Regarding the loess deposits, 195 samples were collected (Supplementary Table S1). Among them, 175 correspond to systematic samplings of 10 vertical profiles with depth intervals from 20 to 50 cm. An additional 20 samples were taken at 16 single points to obtain a more complete spatial distribution. Figure 3 shows examples of the sampled terraces and loess sediments.

The grain size analyses were carried out with a Coulter 230 laser diffractometer, which has a resolution of diameters between 0.04 and 2000 μm in the University of Barcelona’s Laboratory of Sedimentology. The samples were previously treated with H₂O₂ to remove organic matter and with sodium polyphosphate, which acts as a dispersant, for 6 h. The percentages of sand, silt and clay (USDA classification) were plotted in the USDA textural triangle.

The identification of heavy mineral composition and the analyses of chemical properties (calcium carbonate and gypsum contents) were carried out on 18 alluvial terrace samples (10 from the Ebro River and 8 from the Cinca and Segre tributaries). Only two samples of the Miocene substrate contained sufficient heavy mineral content for their analysis. Besides, 38 samples were collected from loess deposits (22 from the different profiles and 16 from the single points). A summary of the samples is presented in Supplementary Table S1.

The heavy mineral analysis consisted of a pretreatment of 60 g of sample to destroy the organic matter and the particulate aggregates, as in the case of grain size analysis. To avoid coatings and aggregates, they were further subjected to an ultrasonic bath with a solution of nitric acid. The fraction size 50–250 μm was selected for the determination of heavy minerals, since it coincides with the most representative loess size in the region, and because this grain size facilitates their determination through the microscope (Morton & Hallsworth, 1994).

The identification of heavy minerals was conducted on the 50–250 μm fraction, after being attacked with 50% HCl to remove...

FIGURE 3 Examples of the sampling points. See Figure 2 for locations. (a) Ebro terrace in Sástago (T10); (b) mas de Caspolí loess profile (MDC); (c) Vilalba dels arcs loess profile (VILL); (d) loess samples of P1. Regarding the scales, b and c have lengths of 4 and 2 m, respectively, the hammer has a size of 25 cm and the shovel approximately 20 cm. [Color figure can be viewed at wileyonlinelibrary.com]
carbonates and washed repeatedly with deionized water to extract remaining reagents. About 5–10 g of the fraction underwent a double gravimetric separation of heavy minerals with dense liquids. Two decantation phases were performed: the first 10 min with bromoform (CHBr₃, density 2.89 g/cm³) and the following 20 min with diiodomethane or methylene iodide (CH₂I₂, density 3.32 g/cm³). The separation process was undertaken using a procedure similar to that described by Mange & Mauer (1992) and Fernández (2016).

Light minerals float in bromoform, whilst heavy minerals (HM) sink. Very heavy minerals (VHM), heavier than 3.32 g/cm³, were also determined as those sinking into the diiodomethane. This gravity method has been used successfully in previous mineral studies (Fenn et al., 2022; Gonçalves et al., 2017; Lin et al., 1992; Morton, 1985; Takehara et al., 2018). VHM include zircon, augite, diopside, staurolite, rutile, barite, garnet group, epidote and kyanite, these being the most representative. To simplify the description, pyroxenes and amphiboles were grouped as inosilicates. The diiodomethane method normally selects about 60% of the mineral species separated with bromoform, and discriminates rock fragments and micas, thus clearly facilitating mineral identification and counting (Mange & Mauer, 1992).

The VHM fraction was mounted on thin sections and fixed with resin. The identification of VHM species was performed with a petrographic polarized light microscope following the manuals of Mange & Mauer (1992) and Andò et al. (2012). In parallel, identification of minerals with a binocular microscope was conducted following the descriptions of Fernández (2016) and the website Sandatlas (2021). The number of identified specimens (grains) was counted following a total counting method as indicated by Mange & Mauer (1992) and Barsó (2006). The counting surface used was 1 cm × 1 cm plots for the source areas and 0.5 cm × 0.5 cm plots for the loess deposits, since the latter presented thinner grains and were consequently more concentrated, with a minimum of 150 transparent specimens per sample.

Results were calculated as abundance percentages of (a) heavy minerals (%HM), (b) very heavy minerals (%VHM), (c) transparent minerals in VHM fraction (%Trans) and (d) 17 VHM species identified in the mineral set.

Calcium carbonate was determined by Bernard calcimetry, with an attack of 50% HCl on 0.5 g of sample of the 50–250 µm fraction following the process described in Porta et al. (1986). Gypsum content was obtained by the gravimetric method from the dehydration of gypsum by heating samples in an oven at 105°C (Artieda, 2013; Artieda et al., 2006) and turbidimetric determination using BaCl₂.

The statistical analyses were performed on 22 variables: percentage of heavy minerals (%HM), percentage of very heavy minerals (%VHM), percentage of transparent minerals (%Trans), percentages of 17 mineral species and the arithmetic mean and mode of the grain size distribution. The results of the statistical analyses helped to check the validity of the spatially homogeneous regions of loess depositional and particle provenance areas, as well as the consistency of the groups and to know which variables best explained each group of data.

The statistical treatment consisted of a multivariate principal component analysis (PCA) and a Pearson correlation matrix of the variables, which were previously normalized with one standard deviation. Pearson correlation tests with a p-value <0.05 were considered significant. Subsequently, a Kruskall–Wallis univariate analysis (H-test) was carried out on the seven most outstanding variables of the PCA, to know whether the different samples were equidistributed and therefore whether they belonged to the same population. If the differences were significant according to Kruskall–Wallis, we applied Dunn’s test to perform pairwise comparisons between each independent group, in order to know which groups were significantly different. The statistical analyses were carried out using the statistical program PAST v.1.89 (Hammer et al., 2001).

4 | RESULTS

4.1 | Exploratory analyses of the source and loess areas

The samples from source areas and loess deposits were subdivided into tentative groups considering their geomorphological setting. The potential source areas consist of fluvial terraces and interfluvos of Miocene materials, subdivided as follows. (1) Terraces of the Middle Ebro River (ME), between Zaragoza and the confluence of the Cinca and Segre river systems. Metamorphic materials from the dismantling of the Iberian System and the Western Pyrenees are the main clastic components of these terraces. (2) Terraces from the Cinca and Segre rivers (C+S), as the main tributaries of the Ebro River in this region. They include materials from the Central Pyrenees, such as slates and granitoids. (3) Terraces from the Lower Ebro River (LE), from the confluence of the Cinca and Segre rivers to the Móra Basin outlet, with mixed materials of Middle Ebro and Cinca–Segre system. (4) Interfluvos of the central area of the Ebro Basin (MI), of Miocene epoch, with materials rich in carbonates and gypsum.

The loess deposits are mostly found on platforms, slopes and valleys around the Ebro River, from Chiprana to Ascó-Gandesa locations and within the Móra Basin. They were subdivided into five areas (Figure 2). (1) Central zone (C), corresponding to the area with the largest accumulation, extension and continuity of loess deposits, located around Batea and on a straight line with the Upper Ebro River to Sástago. (2) Northern area (NC), including deposits bordering the Ebro River itself, together with some samples located on the left bank, also very close to the river, grouping the area south and southeast of Mequinenza. (3) Southern area (SC), located between the valley of the Matarranya and the rivers surrounding Calaceite. (4) Eastern area (EC), including the marginal deposits located east of the central area, between Ascó and Gandesa and very close to the right bank of the Ebro River. (5) Móra Basin (MB), located in a small depression behind the orographic barrier of the Pre-Coastal Range (Pàndols and Cavalls ranges), around the Tivissa Platform.

Taking into account the results of the mineralogical analyses (Supplementary Table S2), the PCA shows that the first two axes explain around 41% of the variance (Supplementary Table S3). The first axis explains 22% of the variability, with the percentages of staurolite (0.37), zircon (0.33), rutile (0.31), transparent minerals (−0.34) and inosilicates (−0.39) as the most important variables. The variables with highest weights in the second axis are the grain size mode (0.43) and mean (0.41), and the percentages of heavy minerals (0.37) and very heavy minerals (0.34).

Although the cumulative variance of the first two axes is not very high, some groupings are observed that are consistent with previous
tentative groups (Figure 4). The first axis divides both the sections of the terraces and the loess areas. In the case of the terraces, it locates the ME to the right part of the graph, with higher percentages of zircon, staurolite and rutile, and the C + S and LE on the left part, with higher percentages of transparent minerals and inosilicates. The C loess samples have similar characteristics to the ME, but differ from the MB and EC samples, which can be grouped with C + S plus LE. The second axis separates the loess samples from the terraces (lower and upper side of the graph, respectively), the latter having higher mean/mode grain sizes and higher percentages of heavy minerals than those of loess (Figure 4). The rest of the samples (NC and SC) are more scattered and can be considered as intermediate stages.

The correlation matrix of the most important mineralogical variables (Supplementary Table S4) shows that, besides the expected high correlation between the grain size mean and mode ($R = 0.92$), the percentage of heavy minerals is positively correlated with the very heavy minerals ($R = 0.64$). Moreover, the percentage of transparent minerals is positively correlated with the inosilicates ($R = 0.65$) and negatively correlated with zircon, rutile and staurolite ($R = -0.70, -0.60$ and $-0.48$, respectively).

The Kruskall–Wallis test showed significant differences ($p < 0.05$) between the samples of sediment sources for all the variables analysed, specifically between the Middle Ebro and the Cinca–Segre system, and between the loess from the central zone and the loess of Môra Basin (Table 1).

4.2 | Grain size

The average particle size of the fine earth (<2000 μm) on the source areas is similar, although the mode of the grain size of Cinca and Segre systems is higher than ME and LE. The average diameter of loess grains was clearly smaller than that from the terraces. Loess from C (right bank of the Ebro River) and MB are the coarsest ones, while the other marginal areas (NC, SC, EC) decrease slightly, but only significantly in the NC. The mode results show similar behaviour to the mean (Table 2).

The texture triangle (Figure 5) shows that the sand fraction of the terraces ranges from 60% to 90% and in loess from 20% to 70%. The sand contents of C and MB deposits and C + S terraces have intermediate values between 50% and 70%. The coarse texture of the loess allows us to classify them as sandy loess (37–67% sand; Bertran et al., 2021) and as wind-borne sands in the few cases where sand content exceeds 67%. The Ebro Basin loess decrease in particle size from the central zone to the surrounding areas. The grain size of the terraces is larger than that of the loess; therefore, the hypothesis of the terraces being the source of the loess should be considered.

4.3 | Total heavy mineral percentage (%HM)

Figure 6 and Table 1 show the distributions of heavy minerals in the 50–250 μm fraction separated by bromoform, sampled from source areas and loess deposits. There are significant differences depending on the river reach considered (ME, C + S and LE): the percentage in the ME was 2.36 ± 0.46%, while in the Cinca and Segre tributaries it was fourfold (C + S, 10.96 ± 2.76%). As a result of this enrichment, the LE increased its content to 5.20 ± 2.77%, although differences between ME and LE were not significant according to Tukey’s Q test.

In general, Miocene sediments from the interfluves (MI) contained a much lower percentage of heavy minerals than the fluvial systems, with an average of 0.19 ± 0.46% (except in sandstone formations, where they were 0.88 ± 0.04%). Therefore, the Miocene sediments acting as source areas would tend to reduce the percentage of heavy minerals.
TABLE 1  Percentage and standard deviation of heavy mineral analyses. Values followed by the same letter/colour are not significantly different ($p < 0.05$) within each group (source areas or loess deposits). Miocene samples are not included in the Kruskal–Wallis test. $S$ is the number of samples. Original data in Supplementary Table S2.

| Ref. | $S$ | %HM ($>2.89 \text{ g/cm}^3$) | %VHM ($>3.2 \text{ g/cm}^3$) | %Transparent | %Zircon | %Staurolite | %Rutile | %Inosilicates |
|------|-----|-----------------------------|-----------------------------|--------------|---------|-------------|---------|---------------|
| Source areas | | | | | | | | |
| ME   | 5   | 2.36 ± 0.46 | a                | 0.22 ± 0.12 | 16.09 ± 4.76 | a    | 19.69 ± 4.13 | a    | 10.17 ± 3.95 | a  | 8.71 ± 1.56 | a  | 23.43 ± 9.34 | a  |
| C+S  | 8   | 10.96 ± 2.76 | b                | 0.55 ± 0.28 | 31.93 ± 4.87 | b    | 4.61 ± 3.46 | b    | 1.05 ± 0.93 | b  | 4.36 ± 1.89 | b  | 55.43 ± 8.84 | b  |
| LE   | 5   | 5.20 ± 2.77  | ab               | 0.38 ± 0.19 | 40.09 ± 14.38 | ab   | 7.23 ± 2.87 | ab   | 2.75 ± 0.72 | ab | 4.44 ± 2.01 | b  | 54.19 ± 8.76 | b  |
| MI   | 5   | 0.88 ± 0.04  |                 | 0.07 ± 0.06 | 18.52 ± 9.80 |       | 12.16 ± 5.26 |       | 10.51 ± 6.73 |     | 3.24 ± 0.21 |     | 30.21 ± 12.97 |     |
| Loess deposits | | | | | | | | |
| C    | 13  | 0.92 ± 0.54  | a                | 0.16 ± 0.15 | 15.45 ± 2.68 | a    | 29.84 ± 4.09 | a    | 4.81 ± 2.79 | a  | 7.23 ± 1.68 | a  | 30.38 ± 5.41 | a  |
| NC   | 11  | 1.75 ± 1.54  | ab               | 0.15 ± 0.13 | 26.82 ± 7.56 | b    | 15.20 ± 6.13 | b    | 2.40 ± 1.82 | b  | 5.33 ± 2.33 | b  | 42.71 ± 9.55 | b  |
| SC   | 3   | 1.27 ± 0.20  | ab               | 0.08 ± 0.08 | 19.62 ± 3.17 | ab   | 15.15 ± 5.49 | b    | 1.69 ± 1.89 | b  | 8.45 ± 3.28 | a  | 43.62 ± 8.18 | ab |
| EC   | 3   | 0.55 ± 0.17  | a                | 0.10 ± 0.06 | 38.06 ± 28.72 | b    | 7.62 ± 4.22 | b    | 1.52 ± 1.51 | b  | 2.35 ± 2.33 | b  | 62.33 ± 20.41 | b  |
| MB   | 8   | 2.52 ± 1.89  | b                | 0.17 ± 0.18 | 32.72 ± 9.03 | b    | 9.33 ± 3.37 | b    | 1.71 ± 1.04 | b  | 4.23 ± 2.26 | b  | 53.62 ± 11.42 | b  |

*In the case of Miocene, the five samples are used to obtain the percentage of heavy minerals (%HM and %VHM) but only two samples have enough minerals to perform the analysis.
In the loess deposits (Figure 6 and Table 1), two sets of well-differentiated values were distinguished: (a) the loess located in the Ebro Basin, where the average percentage of heavy minerals ranges from 0.55 ± 0.17% in the EC to 1.75 ± 1.54% in the NC and (b) the loess from the Móra Basin with higher values (2.52 ± 1.89%). The latter is significantly higher than the heavy mineral contents of C and EC, while those of NC and SC have intermediate values (p < 0.05).

The grading of the heavy mineral contents of the source and loess areas, and the fact that the wind deflation and transport will tend to decrease the amount of heavy minerals from the source areas, allow us to relate the loess from the Ebro Basin to the terraces of the ME, although slightly impoverished with respect to the latter, as the average went from 2.36% to values <1.75%. On the other hand, loess from the Móra Basin exceeded the %HM of the ME, and therefore it is more pertinent to relate them to LE terraces, changing the averages from 5.20% to 2.52%. The decrease experienced in both cases was of the order of half the original percentage.

Some points had specific values well above these averages, such as those of P1 sample (6.76%) in MB, or Almatret (5.39%) in NC, which could incorporate sands from the Siurana River in the first case, and the Cinca and Segre rivers in the second case.

### 4.4 Transparent very heavy minerals percentage (%Trans)

The percentages of transparent minerals (%Trans) in the fraction of very heavy minerals are shown in Table 1. Transparent minerals are mainly silicates of igneous and metamorphic origin. They are present in river sands, while opaque minerals are mostly iron oxides and hydroxides, which could essentially come from Miocene sediments. In the ME terraces, we find low percentages of transparent minerals, with an average of 16.09 ± 4.76%. The tributaries of the Cinca-Segre system, with significantly higher values (31.93 ± 4.87%), caused an
increase in LE that reached 40.09 ± 14.38%, probably with contributions from the Siurana River in MB.

The percentages of transparent minerals in the central zone (C) loess were slightly lower (15.45 ± 2.68%) than in the ME terraces. Nevertheless, a significant increase takes place towards the Northern zone (NC: 26.82 ± 7.56%) and also, but not significantly, towards the other external zones (SC: 19.62 ± 3.17%, EC: 38.06 ± 28.72%). In contrast, in MB the average was 32.72 ± 9.03%, a value more related to the C + S or LE averages. Some values well above the average in MB, such as P1 sample (53.35%), could be directly related to contributions from the Siurana River.

4.5 | Very heavy mineral composition (VHM)

The most representative very heavy minerals studied in the source areas and loess deposits were zircon, staurolite, rutile and the inosilicates group (Table 1, Supplementary Table S1). The other minerals analysed (garnet, epidote, kyanite, monazite, barite, spinel, corundum, anatase, zoisite, titanite) were unrepresentative or did not show a differentiated spatial pattern to make them useful as tracer minerals.

4.5.1 | Source areas

Figure 7 and Table 1 show the percentages of the different mineral species (VHM) identified in the terraces and interfluves. Zircons from the ME were very abundant, significantly higher than in the C + S system. Downstream from the confluence, the samples from the LE reach show a sharp decrease in zircon percentages due to the dilution caused by the C + S system. The spatial distribution of zircon percentages in the study area is presented in Figure 8. The net differences in zircon concentrations raise the possibility of using it as a tracer mineral.

The behaviour of staurolite and rutile is the same as that of zircons, although not as marked. On the contrary, the proportion of the group of inosilicates in the LE doubles the values of the upper reaches. This increase is due to the enrichment of concentrations produced by the Cinca and Segre rivers. Miocene formations would be considered a secondary source of loess sediments. Generally, they had a rather low content of VHM. They contained some minerals from the Iberian Range denudation, such as zircons or garnets, but the low number of samples (2) did not allow statistical analyses.

4.5.2 | Loess

Figure 9 and Table 1 show the percentages of different mineral species (VHM) identified in loess. In C, NC and SC, the sum of zircon, staurolite and rutile tends to be higher than the inosilicates, while in MB and EC the inosilicates are more important. The rest of the minerals have a low representation or do not show sufficient contrast between areas, as happened in the terraces; therefore, they are not considered as tracers of source areas.

Zircon is the mineral with the highest contrast in the spatial distribution (Figure 8). In the zone of the greatest accumulation (C), zircon accounts for 29.84 ± 4.09%, significantly decreasing away from the
The concentration of staurolite is highest in C and decreases significantly in the rest of the areas, including MB. Rutile concentration is highest in C and SC, and lowest in MB and EC. Inosilicates have different behaviour to the previous minerals: they are more abundant in MB and EC than in C.

The concentrations of these four groups of tracer minerals indicate that the signatures of the central loess are similar to those of the ME terraces; likewise, the signatures of loess of the MB are related to the LE terraces.

These relationships are better seen in Figure 10. It displays a ternary plot with staurolite–inosilicates–zircon on the vertex, where the percentages of river sands and loess were plotted. Two outstanding affinities between loess and source areas are observed: loess from the central zone (NC and SC). Towards the east (EC zone) the decrease is more accentuated (7.62 ± 4.22%) and similar to the MB average (9.33 ± 3.37%).
central zone (C) and Middle Ebro terraces (ME), and loess from the Móra Basin (MB) and Lower Ebro terraces (LE). The latter contain a mixture of the mineralogical associations of the Cinca–Segre complex added to the Middle Ebro. EC loess deposits also coincide in the triangle with the Lower Ebro (LE) assemblage. NC and SC loess areas range between ME and LE sample regions.

Table 1 reveals a zircon enrichment in the loess with respect to their fluvial terrace sources in two cases: the central loess of Ebro Basin (C) and the loess of the MB. This enrichment is also noticed in the inosilicates. On the other hand, there was a certain depletion in staurolite. Rutile remained more constant.

The very heavy mineral assemblages of the oldest sequences (Mas de l’Alerany in MB area and Chiprana in C area) are practically identical in mineral variability and abundance to the rest of the (younger) sequences.

4.6 | Gypsum and carbonates

Gypsum does not appear in the terrace sands due to its high solubility. Nevertheless, it is ubiquitous in the Miocene sediment outcropping in the interfluves of the Ebro Basin (Poch et al., 2021). In some lithological formations of the western area (Zaragoza–Sástago), it can reach concentrations >50%. MB sediments do not contain gypsum (except for very rare areas corresponding to Eocene formations in the north and south borders of the depression). In essence, from the point of view of source areas, gypsum is a very important tracer mineral that can only come from the Miocene sediments of the Ebro depression, that is, from the interfluves.

Therefore, the presence or absence of gypsum in loess confirms the differentiation of the two areas of deposition (Figure 11): (a) loess deposits of the Ebro Basin with varying gypsum contents (1–24%) and...
(b) gypsum-free deposits in the Móra Basin. There was no clear pattern of spatial distribution of gypsum, as it is very soluble and can easily be washed and redistributed in vertical soil profiles. However, it is ubiquitous as desert rose crystals at the bottom of the profiles or as vermiciform gypsum, single crystals or nodules. Although there was no definite pattern, some deposits in peripheral areas tended to have higher gypsum contents (up to 24%), probably due to being more exposed to the arrival of gypsum from the Miocene evaporitic formations. It should be noted that, at all times, we are talking about primary loess where the adjacent rocks do not contain gypsum, giving rise to the fact that gypsum can only come from the aforementioned wind materials.

The carbonate mineral contents (calcite and dolomite) were fairly homogeneous throughout the terrace deposits, with averages of ~25%. Loess of the Ebro and Móra basins, with average carbonate contents of 35% and 32.6%, respectively, tended to be higher than the sources of fluvial origin. This increase can be explained by the contribution from the interfluvial sedimentary areas when this component is dominant.

5 | DISCUSSION

The Eastern Ebro Valley loess have a very coarse mean (40–52 μm) and mode (50–79 μm) grain size, comparable with that of sandy loess in Belgium (Bertran et al., 2021) and Tunisia (Coudé-Gaussen, 1990; Faust et al., 2020). These coarse textures suggest very proximal source areas (Pye, 1995), mainly related to fluvial alluvial plains (Fenn et al., 2022; Lehmkühl et al., 2016; Smalley et al., 2009). The most relevant criteria to establish source areas and loess depositional regions are the geomorphological settings, grain size distribution and specifically tracer mineral composition. In the last case, gypsum presence/absence and the heavy mineral signature (zircon, staurolite, rutile, inosilicates) were the most representative markers to define loess regions.

According to these criteria, the data clearly separate two loess populations in the Eastern Ebro Valley, each one related preferentially to a different source area (Figures 4, 10 and 12).

To the west, the Ebro Basin represents the largest loess area, about 1500 km², located almost on the right bank of the Ebro River in the Eastern Ebro Valley. It is made up of proximal deposits with a large accumulation of sediments, with variable gypsum contents, and has been considered the sedimentary aeolian basin depocenter. It has several peripheral areas with a progressive decrease in the deposit thickness (NC, SC and EC). Heavy mineral composition and abundance of the depocenter area (C) show a very good correspondence with the Middle Ebro terraces (ME). In the case of EC and NC areas, we notice a mixed contribution of the ME and LE terraces, due to the proximity of the deposits to the LE system. SC loess shows the signature of ME components with a potential influence of Miocene substrate sandstones, assuming winds from the WNW.

To the east, a much less extensive loess area of about 150 km² outcrops in the Móra Basin. Its depocenter would be located on the eastern platform of the Tivissa landscape. The gypsum from the Zaragoza lithologies does not appear in the Móra Basin, indicating the efficiency of the Catalan Pre-Coastal Range (Pàndols–Cavalls Range) as an aeolian barrier. Heavy mineral composition and percentages of the Móra Basin loess were very similar to those of the Lower Ebro terraces (LE), with a possible influence from the Siurana River terraces. Therefore, the provenance of this loess is mainly the Lower Ebro (LE).
There is a high dispersion of textures in the C, NC and MB loess areas (Figure 5), which could be explained by differences in wind transport competence of dust storms and distancing from the source area located to the west. All these aeolian sediments would have come mainly from sand and silt deflation of old alluvial plains of the ME and LE, which have a width up to 10 km during wind deflation time. They correspond to the terraces about 8–20 m above the current Ebro River talweg. A much smaller proportion of particles would have come from deflation of silt and sand from the slopes of the Miocene substrate, in the interfluves at the north and south of the Ebro River. They are rich in carbonates and gypsum, but poor in VHM (zircons, staurolite, inosilicates). Fine particle availability to wind deflation is clearly higher in terraces than in Miocene substrate interfluves (sandstone, limestones, marls), where very low rock weathering rates and vegetation cover result in low sediment supply and availability (Bullard et al., 2008, 2011).

In the Ebro depression, a 50% reduction in %HM took place between the source area (ME) and the depocenter (C). The same is valid for loess from the MB; they have half the %HM of that of the Lower Ebro River. This dynamic is due to a heavy mineral depletion by weight sorting and the contributions of aeolian material from other areas, such as Miocene substrate interfluves with a lower heavy mineral concentration. The Ebro Valley loess %HM is higher than for other European loess (Chmielowska & Salata, 2020; Wolf et al., 2019, 2021), except in the case of Pannonian Basin, with which they were very similar (Thamó-Bozsó et al., 2014).

There was an enrichment in zircon and inosilicates in the loess compared to the terraces, and a certain impoverishment in staurolite. Zircon enrichment in loess could be explained by the preference for lower diameters during wind transport and a possible contribution from Miocene sandstones. Zircon crystals (50–70 μm) are found in the smallest particle sizes of fluvial terraces (average size 180–200 μm), favouring wind entrainment. In contrast, the transport of the larger-sized staurolite grains would have been more difficult. Similar patterns of grain size sorting can be found in Bosq et al. (2020).

The estimated wind direction from the WNW to ESE, which coincides with the actual ‘cierzo’, is parallel with the ME terrace plain, where wind deflation would be favoured in a cold and dry climate due to the lack of vegetation and lack of cohesion of the particles. Downstream from Sástago, the Ebro River enters an area of very tangled and incised meanders where wind access to detrital materials would be diminished. Downstream the Sástago + MB confluence, winds deflect LE materials. On the contrary, the Miocene substrates were more coherent, found as soils under a certain vegetation cover.

Quaternary alluvial sediments from the Middle Ebro (Zaragoza to Sástago) form a NW–SE corridor, longer than 50 km and with a width of up to 10 km. Winds blowing along this corridor would generate the central loess, which show the maximum similarity of the variables

![Diagram](https://example.com/diagram.png)
studied. The corridor is flanked in a parallel manner in the north and south by Miocene substrates, where gypsum is especially present east of Zaragoza. The central area (C) presents similar patterns as the Middle Ebro. The northern area (NC) would differ from the central area due to a higher contribution of Miocene and Lower Ebro sources, while the eastern area (EC) would mainly have a mixture of contributions from the Middle and Lower Ebro. Finally, Moça Basin loess does not show any contribution from the Middle Ebro, being completely formed from Lower Ebro sediments. According to these sources and the main wind direction, the minimum transport distances would range from about 100 to 25 km in the central zone, to only 15 km for the MB.

The similar mineralogical composition of the oldest and more recent sequences in Mas de l’Alerany and Chiprana, attributed to a MIS-6 phase and a MIS-2 phase, respectively, suggests that there were no substantial changes in the main source of sediments during this time interval and that the direction of prevailing efficient winds did not differ either (Gutiérrez et al., 2013; Mensua & Ibáñez, 1975; Plata et al., 2021; Schatz et al., 2015; Wolf et al., 2019).

Terraces of the Middle Ebro have a particle deflation level located between 150 and 200 m a.s.l. and Ebro Basin loess outcrop between 150 and 450 m a.s.l. By contrast, the Pàndols–Cavalls Range is an orographic key structure, perpendicular to the wind direction, reaching heights of 550–705 m a.s.l. This very effective wind transport barrier has disconnected the two main loess depositional areas (Ebro and Moça basins), as shown by the mineral signatures. Although the mountain barrier only represents an elevation increase of about 200–350 m over the loess platforms, dust storms apparently failed to surpass it.

Loess formation implies the simultaneous activity of several geomorphologic processes and environmental systems (glacial, periglacial, semiarid/arid) acting in cold and dry paleoclimatic conditions (Bertran et al., 2021; Li et al., 2020; Smalley et al., 2009). The proposed morphoclimatic processes and systems involved are as follows. (1) Preparation of fine materials by abrasion and glacial transport in the Pyrenean headwaters, and to a much lesser degree in the Iberian System. (2) Fluvioglacial and alluvial transport from mountain braided rivers to the wide Middle and Lower Ebro meandering alluvial plains, amplified by strong flood events during thawing seasons, cleaning the riverbed from vegetation. (3) Silt and sand wind deflation in the Middle and Lower Ebro floodplain enriched with interfluvial Miocene substrate supply under poor vegetation cover. This aeolian transport would take place by saltation and suspension mechanisms at low altitude (dust storms). (4) Accumulation of loess deposits in suitable aerodynamic positions on landscapes located against the direction of the dominant wind (E–SE leeward hillsides and valley bottoms). Phases 3 and 4 took place in the colder periods of MIS-2 and MIS-6. The main winds followed a WNW–ESE direction, along the Ebro Valley axis.

Regarding the methodology used in mineralogical analysis, the use of diiodomethane is revealed as a useful technique to separate and identify VHM signatures of loess and source areas, because it shows a good correlation with the classic bromoform separation techniques and reduces noise from rock fragments and micas. Besides, it would be interesting to increase the number of samples to obtain more significant results, especially in the EC and SC distal areas.

6 | CONCLUSIONS

The significant loess deposit cover in the Eastern Ebro Valley is formed by fine sands and silts with a grain size mode between 50 and 79 μm. They are coarser than the classic loess and this implies a closer source area.

Textures and particle mineralogical signatures of the Ebro, Cinca and Segre rivers, as well as of interfluve areas, are sufficiently contrasted to separate source areas and relate them to different groups of loess deposits. In particular, the proportion of HM and some mineral concentrations in loess (gypsum, zircon, staurolite, inosilicates) can be used as effective tracers for particle provenance.

Our results show two clearly differentiated deposition areas related to two major source areas. The first is the loess deposits of the Ebro Basin, up to the border with the Catalan Pre-Coastal Range, which are rich in gypsum, zircon and staurolite, and are poor in inosilicates. Their origin is mainly the deflation of sedimentary materials located in the Middle Ebro alluvial plain, between Zaragoza and Sástago, and of the interfluvies on both sides of the Ebro River, with gypsum outcrops. On the right bank of the Ebro River, a depocenter can be identified around Batea, richer in zircon. In the lateral zones surrounding the central area, the Middle Ebro has less influence and a certain increment of contribution of interfluvies and Lower Ebro terraces is noticed. Transport distance by wind would have been between about 25 km in proximal areas and about 100 km or even more in distal areas.

The second depositional area is the Moça Basin, with gypsum-free loess. It is poor in zircon and staurolite, and rich in inosilicates. In this case, it would have come from the remobilization of alluvial materials from the Lower Ebro River, after the incorporation of contributions from the Cinca and Segre rivers. The provenance would be very local and the wind transport distance much shorter, less than ~15 km, confirming its isolation with respect to the Ebro depression.

The transporting wind would have had a WNW–ESE direction, coinciding with the current direction of the prevailing winds in the Ebro Valley (known as Cierzo). Aeolian sediment provenance and wind direction would have remained constant during the two deposition cycles related to MIS-2 and MIS-6 glacial phases, as the respective mineralogical signatures are very similar. Finally, the Pàndols–Cavalls Range relief, perpendicular to wind direction with a height difference of 200–350 m, acted as a wind barrier separating the loess of Ebro Basin from the loess of Moça Basin.

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DATA AVAILABILITY STATEMENT

Data are available in the online Supplementary Material.
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SUPPORTING INFORMATION

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