Paleoenvironmental reconstruction based on particle size and shape of aeolian dust deposits

K120620 – funded by NKFIH (National Research, Development and Innovation Office)

Project closing final report by György Varga (PI)

The research project NKFIH K120620 entitled **'Paleoenvironmental reconstruction based on particle size and shape of aeolian dust deposits'** officially ended on 30 September 2021, after a prolongation of 12 months (the project started on 1 October 2016).

The project was aimed at

(1) to **analyse the size and shape properties of individual particles of aeolian dust deposits** which are standing in the focal point of environmental studies;

(2) to provide **new quantitative data of particle shape properties** of mineral dust particles;

(3) to critically examine the **applicability and reliability of widely used grain sizing techniques**;

(4) to develop a new **granulometric paleoenvironmental proxy**;

(5) to **identify the potential inadequacies and oversimplifications of interpretations** of grain size data in paleoenvironmental reconstructions.

The objectives of the project emphasized the **clarification of critical methodological issues**, without which we would not have granulometric data of sufficient quality and without which, of course, the paleoenvironmental reconstructions would be subject to errors. To achieve the above-mentioned objectives, a multi-proxy approach was applied by simultaneous usage of field works and laboratory measurements by applying complex granulometric characterisation by (1) laser diffraction and (2) automated static image analysis, and Raman spectroscopy).

The results of the project were published in peer-reviewed research papers (published in **Sedimentary Geology** in 2018 and 2019 [Q1 – IF: 3.244 and 2.728]; in **Quaternary Research** in 2019 [Q1 – IF: 2.31]; in **Quaternary International** in 2019 and 2020 [Q1 – IF: 2.003 and 2.130]; in **Frontiers in Earth Science** in 2020 [Q1 – IF: 3.498]; in **Environment International** in 2020 [Q1/D1 – IF: 9.621]; in **Scientific Reports** in 2021 [Q1/D1 – IF: 4.379]), and were presented at major international conferences (International Symposium on Eolian Dynamics, Paleosols and Environmental Change in Drylands 2017; EGU2018; EGU2019; EGU2020; EGU2021; Goldschmidt2019; Goldschmidt2021; IASC-2019 Workshop on Effects and Extremes of High Latitude Dust in Reykjavik, Iceland; IASC-2020 Workshop on Effects and Extremes of High Latitude Dust in Reykjavik, Iceland;, in social media (blogosphere, Twitter and Facebook) and in popular science papers (Élet és Tudomány). All details, events, scientific background and results of the project have been published at homepages and blogs of the PI, at http://porvihar.blogspot.com/p/nkfi_24.html and http://aeoliandust.blogspot.com/p/nkfi-120620.html.

Samples

Samples were collected from

(1) **loess-paleosol sequences** of Central Europe: Carpathian Basin (Hunagary: Paks, Dunaszekcső, Beremend, Basaharc, Tamási, Debrecen, Outer-Somogy, Tokaj; Croatia: Zmajevac, Ilok, Sarengrad, Pozega, Velika; Serbia: Stari Slankamen, Ruma, Titel; Slovakia: Malá nad Hronom), Germany: Grimma, Pegau; from local colleagues: Poland, Czechia, Romania, Bulgaria

(2) **red (paleo)soils, aeolian sands and Saharan dust**: Canary Islands (Fuerteventura and Lanzarote);

(3) **terra rossae soils** Mediterranean Basin: Spain and France (Ebro and Rhone Basins), from local colleagues: Turkey; Greece;

(4) **Saharan dust**: Carpathian Basin; European Alpine regions (the Pyrenees, Alps, Carpathians); from local colleagues: Saharan source areas (Chad, Egypt, Libya, Mauritania, Tunisia, Western Sahara);

(5) **cold environment sources** (Hagavatn, Sandkluftavatn, Landeyjarsandur, Maelifellsandur, Myrdallsandur and Dyngjusandur) and Saharan dust Iceland.

(Due to the ongoing global pandemic of coronavirus disease (COVID-19), a large number of samples were gathered from local colleagues.)

Applied particle sizing methods

Laser light scattering particle sizing involves the indirect estimation of grain size distribution, which is back-calculated using the measured angular variations in the intensity of light scattered (diffracted, refracted and reflected) and absorbed, as the laser beams are passing through the sample suspension. GSDs of loess and paleosol samples were analysed using three different instruments: a **Fritsch Analysette 22 Microtec Plus**, a **Horiba Partica La-950 v2** and a **Malvern Mastersizer 3000** (with a Hydro Lv wet dispersion unit) devices. Particle size data are given as volume percentages of particles classed into ~100 logarithmic bins in a size range from several nanometres to millimetres.

Laser light scattering patterns of two light sources were measured by focal plane detectors, side, and backward scattered light detectors. The exact construction and set-up of detectors are different from manufacturer to manufacturer. For larger particles, light is dominantly scattered by diffraction leading to high light intensity and low angle. At the same time, the refraction and absorption efficiency is higher for smaller particles resulting in low-intensity light with wide scattering angles. The acquired signals were transformed into particle size distribution data by the laser device software using **Fraunhofer approach** and/or **Mie-scattering theory**. It must be noted, however, that the algorithms used by the different manufacturers are not public, thus the mathematical calculations cannot be compared.

In our studies, a **Malvern Morphologi G3-ID** device was used as automated static image analyser. Approximately 7 mm^3 of mineral material per sample was dispersed onto a flat glass slide with an instantaneous (10 ms) pulse of compressed air (4 bar) and a settling time of 60 s. Particles were scanned using a 20× magnification lens (0.025 μm^2 per pixel resolution) attached to a Nikon eclipse microscope with z-stacking enabled (two focus layers were added above and below the initial focal plane, equivalent to a ± 13.5 µm z-height focus range).

Particle size and shape parameters of hundreds of thousands of individual particles per sample were automatically recorded by the device software. Circle-equivalent (CE) diameter, aspect ratio, circularity, convexity, and solidity granulometric parameters were determined. The CE diameter is calculated as the diameter of a circle with the same area as the projected two-dimensional particle image. The aspect ratio is the ratio of the particle width/particle length. Circularity is a proportional relationship between the circumference of a circle equal to the object's area and perimeter. Convexity is the ratio of the convex hull to a particle's perimeter, while solidity is the ratio of the area of the convex hull to the particle area. Aggregated and stacked particles were filtered by their certain parameters; particles with a circularity of < 0.6 were excluded from the granulometric characterisation. A cluster analysis was applied to determine the similarities of size and shape patterns in samples from different sources via a quantitative approach. Hierarchical cluster trees were created using the Euclidean distance pairs of the selected parameters.

A **built-in Kaiser Rxn1 Raman spectroscope** was used for the chemical analysis of particles. The spectra acquired using a 785 nm (< 500 mW) laser over 5 s were correlated to library spectra (BioRad-KnowItAll Informatics System 2017, Raman ID Expert) to get mineralogical information.

Methodological questions

The particular importance of grain size measurements in loess research

Large variety of methodological approaches (stratigraphy, pedology, geochemistry, mineralogy, micromorphology, magnetic susceptibility, biomarkers, paleontology-malacology, colourimetry, etc.) have been applied to give information on the paleoclimatic and paleoenvironmental conditions of loess and paleosol forming periods. **Particle size data of clastic sediments provide insights into the physicochemical environment of entrainment, transport, accumulation and post-depositional alterations of mineral particles**, and therefore changes in size distributions and granulometric proxies are widely used in paleoclimatic reconstructions.

A large number of research papers on grain size data uses simple statistical descriptors to infer the stratigraphic, sedimentological and evolutionary significance of grain size data. The amount and physical properties (e.g. grain size) of mineral dust mobilised, transported, deposited (and/or redeposited) and altered after deposition are modified by several simultaneous environmental factors. Approaches that provide a direct relationship between simple statistical descriptors (mean, mode or median) of particles size distributions and wind speed and strength, distance from the source or dryness/area of the source region are rather simplistic, as the grain size of sediment samples from aeolian dust deposits is influenced by the integrated effect of several concurrent processes. Size-fraction ratios and unmixed sedimentary subpopulations provide a more sophisticated picture on sedimentation mechanisms and post-depositional alterations. However, the open issues of interpretation of grain size distributions, and the already uncertain measurement approaches (from pipette and sieve methods to laser diffraction) and measurement results create additional difficulties of particle size based paleoenvironmental reconstructions.

Sieve and pipette methods, laser diffraction and image analysis of pictures taken by optical or scanning electron microscopes are in general used to determine granulometric parameters of sedimentary rocks. These methods are based on different physical principles and often provide discrepant datasets of granulometric parameters. During sieve analysis, the second-largest dimension is measured and the particles are optimally oriented to pass through the mesh. In this approach, grain size distribution is calculated from the mass of different size classes represented by the progressively decreasing mesh sizes of a series of sieves. Techniques based on sedimentation rates of suspended particles assume that larger and heavier particles settle more rapidly than smaller and lighter ones. Unfortunately, shape effects (e.g. platyness) are generally not taken into consideration. In image analysis, both the size and shape parameters of scanned particles are recorded. For this method, the basic granulometric size parameter is the circle-equivalent (CE) diameter of particles. CE is calculated as the diameter of a circle having the same area as the two-dimensional projected area of the particle. In this case, number size distributions are generated by assigning each particle to logarithmically-spaced size bins. Subsequently, these distributions are transformed into volume size distributions by weighting each size bin with the total sphere-equivalent volume (calculated from the CE diameters) of particles classed into given size ranges.

Laser diffraction challenges

Laser diffraction is the most commonly used technique in particle sizing due to easy operation and high sample throughput. The volumetric amount of particles arranged into ca. 100 size bins are determined in a size range from hundreds of nanometres to several millimetres. Datasets acquired with laser diffraction measurements are regarded as more robust, accurate and reliable than those obtained with sieving and the pipette method. Laser diffraction particle size data provide indirect information on the volumetric, sphere-equivalent diameter of the particle. Diffraction patterns of the laser beam passing through the particulate suspension are used to calculate volume size distributions with different optical models (Fraunhofer and Mie theories). The angle and intensity of monochromatic light modified by diffraction, scattering and absorption are proportional to the particle size. The Fraunhofer approach (FA) assumes that the particles are large enough so that refraction and absorption effects are reduced to negligible levels. At the same time, application of the Mie scattering theory (MST) requires knowledge about the complex refractive indices of both the sample material and optical properties of the dispersant.

However, unlike regular, spherical objects, grain size characterization of irregular-shaped sedimentary particles is much more difficult due to diffuse scattering patterns. Even the size of non-spherical grains is a matter of debate and estimated in general by applying so-called equivalent diameters (ED). For this, the real, irregular particle is replaced by an imaginary sphere or circle having a similar volume, surface or area to the measured particle. Consequently, any size description of a non-spherical particle using simple indices (sphere equivalent [SE] or CE diameter) implies oversimplification. Manufacturers, however, have developed their own algorithms to compensate for these effects associated with the measurements of irregularly shaped particles.

To date, only a few studies investigated in detail the potential effects of different optical approaches on laser diffraction results for non-spherical particles. Furthermore, there is an obvious lack of scientific studies rigorously testing the performance of commercially available laser diffraction devices. Robustness, reproducibility, and comparability of grain size data obtained with various devices is a basic issue and associated uncertainties are rarely considered. Previous studies on the optical setting dependence of laser diffraction measurements suggested appropriate values of complex refractive indices suitable for only one laser diffraction device currently applied by the authors, however, different devices with different set-ups have not been compared yet.

In our study pubished in Sedimentary Geology (Varga, Gy., Gresina, F., Újvári, G., Kovács, J., Szalai, Z. (2019). **On the reliability and comparability of laser diffraction grain size measurements of paleosols in loess records**. Sedimentary Geology 389, pp. 42-53.), particle size data of paleosols in loess sequences was presented as measured by three different laser light scattering instruments: the Fritsch Analysette 22 Microtec Plus (Fritsch), Horiba Partica La-950 v2 (Horiba) and Malvern Mastersizer 3000 (Malvern). In addition, particle size and shape distributions were obtained from Malvern Morphologi G3-ID automated static image analyses performed on the same samples. To complement these datasets, scanning electron microscope images were taken as references for particle shapes, and X-ray powder diffraction measurements provided insight into the mineralogical compositions of sample materials.

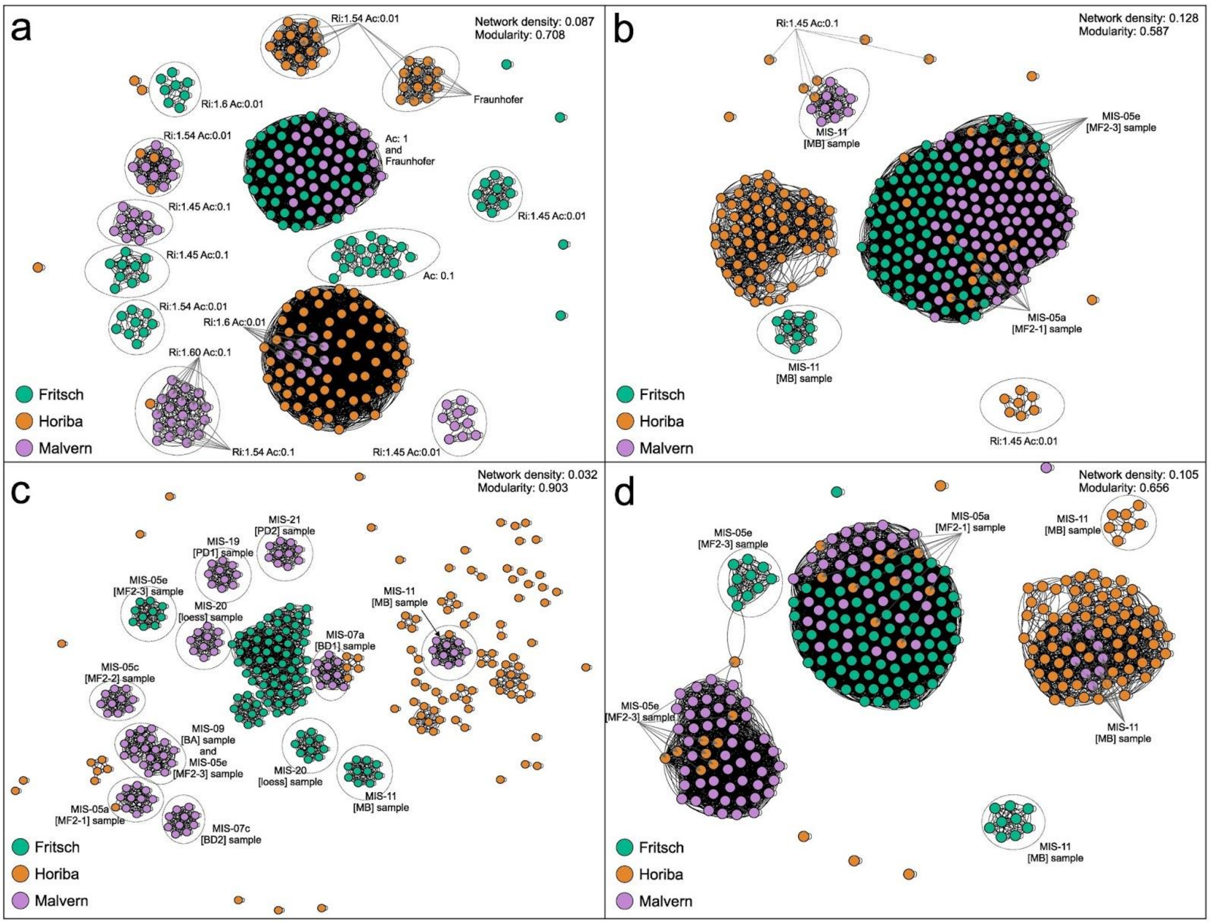

Figure 1. Network analysis results based on volumetric proportions of particles in size bins of different fractions (a) clay; (b) silt; (c) sand; (d) bulk. (Connected nodes represent similar size distribution patterns in the given fraction.)

Our investigations focussed on grain size distributions (GSDs) of **10 selected samples** measured using **3 state-of-the-art laser diffraction instruments**. The applied **68 different optical settings** of these instruments resulted in **2040 grain size data-series**. One of the most important findings was that calculations with the Mie theory provide more accurate data on the GSDs of paleosols in loess sequences. Significant differences between the Mie and Fraunhofer approaches were found for the finest grain size fractions, while only slight discrepancies were observed for the medium silt fractions.

Since the two approaches gave similar results for the medium to coarse silt fractions that are the most abundant in loess, the use of these two approaches has no appreciable effect on the mode and other single statistical descriptors of the distributions. In conclusion, the different applied optical parameter settings and device selection have had significant effects on measured volumetric amounts of the fine-grains present in the investigated sedimentary samples, which are widely applied in paleoenvironmental reconstructions as proxies of chemical weathering, the degree of pedogenesis and/or external dust addition from secondary sources. An additional uncontrolled factor is the shape-dependence of measurements, which is critical as both of the applied scattering approaches assume spherical particles. In reality, however, sedimentary mineral grains are generally anisotropic in shape. According to our investigations by automated static image analysis, coarse silt- and sand-sized particles have a more diverse granulometric character than the finer fractions while the smaller grains exhibit more homogeneous shape characteristics. Nevertheless, 3rd-dimensional anisotropy cannot be revealed using 2-dimensional images. This later finding could be another source of major uncertainties of laser diffraction measurements as the volumetric proportion of platy clay minerals cannot properly be determined. Thus, it seems that all these grain size analyses methods are compromised to some extent.

Automated static image analysis: shape matters

Not only size, but shape parameters of particles hold vital information on sedimentary transport and deposition mechanisms and postdepositional, environment-related alterations. Traditional image analysis techniques have been applied widely, however previously published studies have been carried out on populations with much smaller number of particles compared to automated analyses. Measurement of particle shape is time-consuming. Automated static image analysis is still uncommon and underexploited for particle size and shape distribution analysis of sediments. The use of automated digital image analysis solves the issues generated by low number of measured particles as it is more precise, less time-consuming and easier to use compared with traditional methods. The average particle number of automated imaging amounts to ca. 10^4-10^6 particles, which allows us to gain statistically robust and objective insights into the morphological characteristics of particles. Various size and shape parameters, as well as optical intensity values of each particle, are routinely measured and number-size distributions can easily be converted to volumetric distributions, thus the direct comparison with results obtained by laser diffraction can be done. To date, only a few studies have been published on automated image analyses of particle size and particle shape parameters of sedimentary deposits, and therefore much uncertainty exists about the relationship between the different methods.

In our paper published in Sedimentary Geology in 2018 (Varga, Gy., Kovács, J., Szalai, Z., Cserháti, Cs., Újvári, G., (2018). **Granulometric characterization of paleosols in loess series by automated static image analysis**. Sedimentary Geology 370, pp. 1-14.) the grain size results obtained by widely used laser diffraction technique and by a new, high-precision granulometric characterization approach, namely automated static image analysis was compared; the major differences and underlying causes were discussed; and problematic issues of grain size and shape determinations of the automated static image analysis technique were identified.

Granulometric investigations of Pleistocene interglacial paleosols intercalated into loess sequences in the Carpathian Basin revealed the major discrepancies in results obtained by the laser diffraction and automated static image analysis measurement techniques applied. The data acquired by widely used, indirect laser diffraction and direct observations by automated image analysis provided complementary, but different information on grain size. While the particle size distributions provided by laser diffraction measurements are dependent on the complex refractive index of a given particle (which can only be approximated in case of polymineral samples) assuming a spherical shape, the image analysis techniques are based simply on the direct, optically-acquired images of grains.

Comparisons of measured grain sizes indicated that the fine populations are consistently and significantly underestimated by the image analysis technique compared to laser scattering results. Modelling data demonstrate that the anisotropic character of irregular particles, especially the thickness of platy minerals, are responsible for the observed disagreements. The acquired two-dimensional images of dispersed particles sitting with their largest area on the glass slide were classified into grain size bins being too large based on their circle-equivalent diameter. In addition, their volumetric-weighting scores (sphere-equivalent volume derived from the CE diameter) were also found to be too high in volume-based conversions. Consequently, this led to overestimation of particle sizes and volumetric amounts of wrongly classified platy grains due to the cubic relationship. Application of the rotation averaged and SE/CE ratios as correction factors successfully reduces the discrepancies between results obtained by the two approaches. Nevertheless, the most definite factor, the unknown thickness of particles still remains an unresolved problem. The other presented innovative way of estimating the uncertain 3rd dimension of particles using their intensity-size relationships allows us to further minimize deviations between the two particle sizing methods.

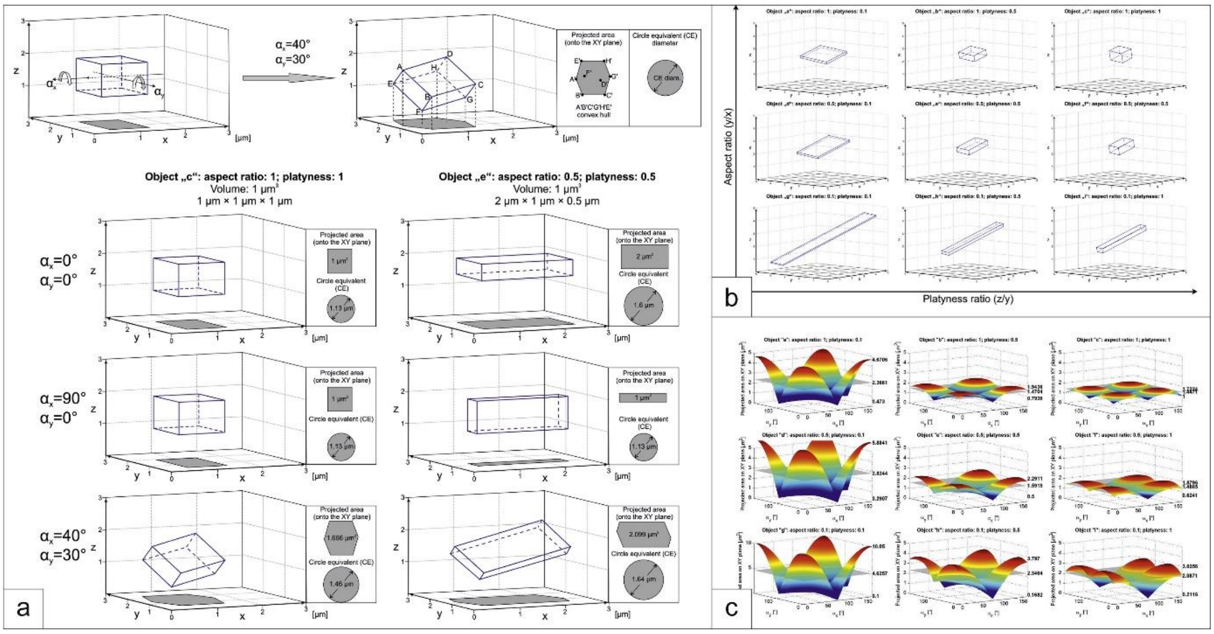

Figure 2. Assessment of projected area of a randomly oriented geometric objects: (a) general problem of shape and rotation determined projected area; (b) presented nine simple geometric objects; (c) rotation averaged projected areas as a function of rotation angles.

However, since particle sizes of paleosols covering several orders of magnitude, even a small number of coarse grains can modify significantly the grain size distribution curves in the larger fractions distorting the whole measurement spectrum, and so the full agreement between laser diffraction and image analysis results cannot be reached. There are discrepancies of these above discussed methods, but these can be handled by deeper understanding of physical background of them.

Paleoenvironmental interpretation of graunulometric data

The studied paleosols are the product of a complex depositional environment: granulometric characteristics of paleosols are dependent on (1) the grain size properties of the underlying windblown loess material from which the soil was formed; (2) post-depositional alteration governed by the weathering intensity characteristic for the given interstadial/interglacial period; and (3) possible syn-sedimentary dust material additions (and/or removal). However, it must be emphasized that this study was not aimed at obtaining genetically meaningful sedimentary interpretations of the samples. In our Quaternary International paper (Varga, Gy., Újvári, G., Kovács, J. (2019). **Interpretation of sedimentary (sub)populations extracted from grain size distributions of Central European loess-paleosol series**. Quaternary International 502, Part A, pp. 60-70) differences of frequently applied grain size data interpretation approaches and methods were discussed, main reasons of the revealed differences were ascertained; and a comprehensive overview on the geological importance and genetically meaning of the subtracted sedimentary (sub)populations and clusters were provided.

Major characteristics of the granulometric properties of the studied high-resolution loess-paleosol profile of Dunaszekcső, South Hungary (Central Europe) were represented reasonably well by all applied techniques. General upward coarsening of the grain size values of the section was clearly indicated by all of the investigated granulometric descriptors, indices and proxies. However, results of single statistical indices provide very similar section-down curves without clear environmental signals; indicating only some changes in main granulometry. Not to mention the problems of simple statistical indices which are not likely to be representative for polymodal size distribution curves.

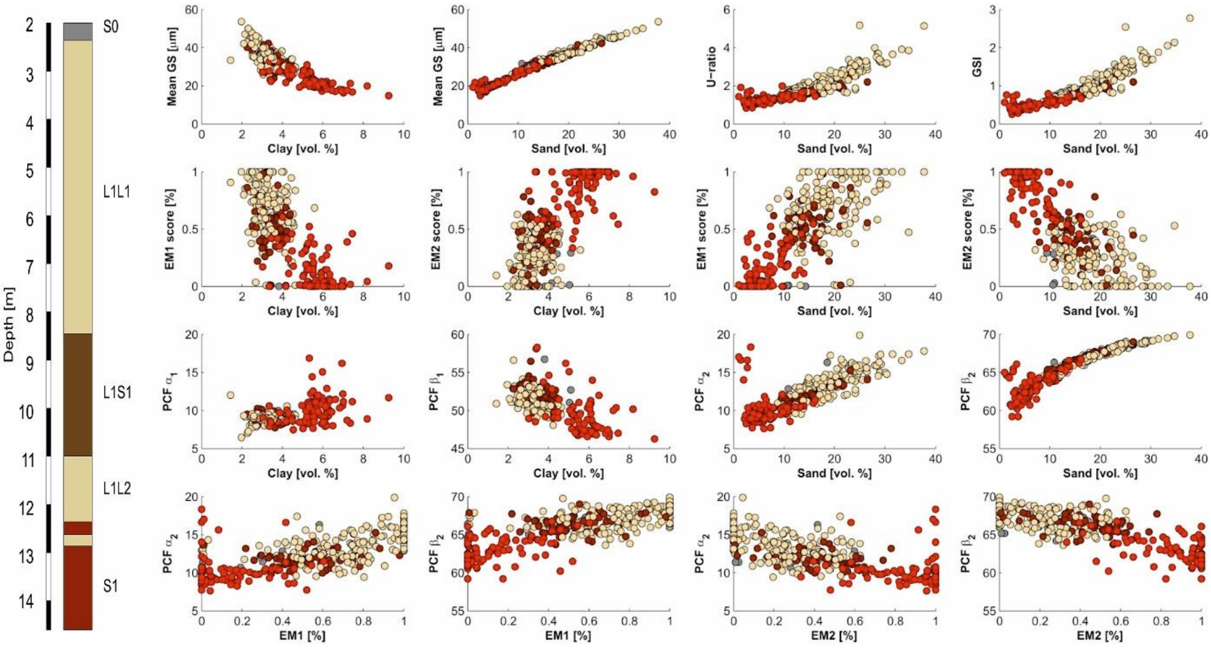

Figure 3. Relationships between different grain size indices and proxies (EM: end member; PCF: parametric curve fitting; colours indicate the stratigraphic units).

Deeper sedimentary and genetically meanings can be deciphered by the application of more complex mathematical algorithms. Parametric curve-fitting is based on the one-by-one break-up of polymodal grain size distribution curves into aggregates of unimodal statistical probability distribution functions (in this case Weibull-functions were applied). End-member modelling provide fix sedimentary populations and the measured grain size distribution curves of the samples are composed from these ‘end-members’ by weighting them with an appropriate score. As the input for this later method is the whole data-series and end-members are held fix and constant, the goodness-of-fit values are not as high as parametric curve-fitting scores.

End-members of loess-paleosol samples are regarded as representation of the average dust grain size distribution of various temporal sediment clusters of seasonal or other short-term intervals, while (sub)populations by parametric curve-fitting are proposed to illustrate process-related elements of background and dust storm depositional components for each sample. The end-members were calculated from the whole grain size database of the investigated section, while the input for the parametric curve fitting is only one sample. The end-members are polymodal, the simple probability density functions of parametric curve fitting are logically unimodal. The polymodal end-members cannot be regarded as the representation of a single dust transportation and/or sedimentation process; these are illustrated by the Weibull functions. The end-members can only be considered as the result of more simultaneous sedimentation mechanisms, which are dominant in a specific period of a year (e.g. seasonal dust signal: spring dust storms connected to the arrival of cold fronts). Results of cluster analysis represent similar grouping conditions as end-member modelling with a reduced sedimentary and genetically meaning.

To develop a full granulometric picture joint application of parametric curve-fitting and end-member modelling is suggested. Parametric deconvolution of the fix and stable end-members the process related seasonal aeolian sedimentary dynamics could be recognized. Another source of uncertainty, the optical setting dependence of laser scattering grain size measurements deserves also a more accurate analytical attitude and correct description in methodological sections of the research papers.

Beyond granulometry-based paleoenvironmental reconstructions

Manuscript by Szeberényi et al. (2020) was published in the Quaternary International (title: **Experiencing new perspectives in the application of reflectance spectroscopy in loess research**) about the application of diffuse reflectance spectroscopy (DRS) in loess research. The hierarchical cluster analysis of DRS curves and a new method (DRS stratigraphy) in terms of its application to DRS results, provide complex, new approaches to the study of loess-paleosol successions. Our method examines the changes within the range of UV, visible light and the shorter wavelength sections of the near-infrared range at the same time. Different cluster groups are determined based on the spectral features of samples, which can be interpreted as stratigraphic units of loess-paleosol successions.

In our paper, published in Frontiers in Earth Sciences in 2020 (Kovács et al., 2020) **silt and clay-rich deposits and paleo-karst fissure sediments** from sites of the northern and southern parts of the Carpathian Basin were investigated. These materials were supposed to be mixed during transport before being captured in karstified fissures. Evidence that the aeolian fissure sediments of Plio-Pleistocene age in the older Triassic–Cretaceous limestones are derived from eolian silt and clay includes compositional and textural matches, especially decreasing grain-size trends observed downwards from the paleo-surface of the former landscape. Various environmental factors could be recognized by the statistical evaluation of grain-size distribution curves of fissure fillings sediments, such as the effects of eolian transport, parent rock type, weathering, and other sediment transport processes. Grain-size distribution curves with a single maximum in the silt size range are typical for the overlying siltstone debris, for the redeposited loess and red paleosol underlying the loess. Red clay fissure fillings yield bimodal grain-size distribution curves with maxima both in the clay and silt fractions. The research reported in this paper identifies for the first time the presence of eolian deposits in karst fissures of the Carpathian Basin and investigates the characteristics and origin.

The suitability of our granulometric analytical approach was also demonstrated by investigating porewater-rock interaction of **Miocene sandstones** which may cause the dissolution and precipitation of different minerals. Results were published in Hungarian Geographical Bulletin (by Király et al. 2019, Granulometric properties of particles in Upper Miocene sandstones from thin sections, Szolnok Formation, Hungary).

Granulometric alteration index

Reconstructive studies of past climate changes are essential for a deeper understanding of the Earth's climate system. This is of particular importance today, since predicting the potential impacts of present climate change and determining the extent of human activity in all these changes cannot be done without understanding the natural dynamics of similar events in the past. The Pleistocene glacial-interglacial alternation represents two main contrasting states of long-term climate systems, with short transition periods. Based on stable isotope analyses of global reference curves from deep-sea, ice core and cave records, it has long been evident that the duration, intensity and climatic conditions of different interglacial periods were significantly different. Identifying the driving forces leading to warm-wet periods and reconstructing the paleoenvironmental conditions of these interglacials may provide an analogy to Holocene interglacials and the dynamics of natural climate change.

While the geochemical composition of the loess deposits is rather homogeneous, the duration and intensity, the climatic and environmental conditions of soil forming intervals were more variable than during the glacials, leading to a geochemically and sedimentologically mixed interglacial pedostratigraphy. It is important to emphasize that the oversimplified stratigraphic approach, whereby loess is purely glacial dust deposit, and paleosols represent interglacial soils, is not tenable. Firstly, the majority of mineral particles of interglacial paleosols were (1) transported, (2) accumulated and (3) cemented to loess during the preceding glacial period; after that (4) interglacial postdeposit pedogene processes played the dominant role and altered the particles and structure, formed the loess into soil. In this sense, none of the loess-paleosol sequences can be regarded as a full stratigraphic record of the glacial-interglacial changes. The detrital mineral material of the last phase of glacial is in the soil material of the next interglacial.

On the other hand, both glacial and interglacial sedimentation have been subject to syngenetic dust addition from distant sources (e.g., from hot desert areas). Due to high local dust fluxes, the relative contribution of external dust did not have a significant effect on (glacial) loess formation, however, interglacial accretionary soil formation modified the physicochemical composition of soils more effectively, hence influence of fine-grained dust addition should be taken into account in the paleoenvironmental interpretation of proxies derived from paleosols.

In our paper (in prep – will be submitted in the next weeks), the primary aim was to compare the complex shape parameters of interglacial paleosols and underlying remnants of glacial loess deposits from which the soils were formed. By this systematic comparison of the unweathered loess and the covering weathered soil material, the degree of pedogenic processes was assessed. These derived granulometric alteration index was compared to traditional alteration proxies (chemical index of alterations, geochemical proxies) and grain size parameters (size-fractions, fraction-ratios, sedimentary subpopulations). To meet the objectives, automated static image analysis technique was used to directly measure the grain size and shape parameters of several hundreds of thousands of individual silt-sized mineral particles per sample. To provide a wider spatiotemporal range of the work, two different sample sets were investigated: (1) different weathering intensities and changing nature of interglacials from the analyses of a longer time-period from one geographical location: loess-paleosol sequence of the **Carpathian Basin (MIS21 to MIS2)**; (2) different weathering intensity of the last interglacial in different geographical regions: **MIS6-MIS5e loess-paleosol samples from Central Europe (from Germany to Bulgaria)**.

Based on our findings, it could be stated that several **quantitative shape parameters of paleosols are sensitively dependent on weathering intensity**, however, robust relationship among chemical indices and granulometric parameters could not be clearly specified, suggesting our approaches need to be refined. It is worth noting, however, that chemical weathering indices should be treated with caution, according to a number of previous studies. For this reason, it is not clear that the procedures we have developed do not provide accurate information on weathering intensity.

Identification of syn-sedimentary dust addition to sedimentary and pedogene units

Hot aeolian environment: Fuerteventura (Canary Island)

Samples were taken from 24 silty units considered as paleo-surfaces of stable geomorphic periods with reduced sand movements and relatively enhanced **Saharan dust influence, additional dune sand and sand sheet samples** were also investigated as references for intense sand transportation intervals (Varga – Roettig, 2018; Roettig et al., 2019).

Measurements of ~50,000 individual mineral particles per samples provided huge amount of granulometric data on the investigated sedimentary units. Based on the fact that the Saharan dust deposited at Fuerteventura is mainly (1) silt-sized and (2) contains a lot of quartz particles (regarded as exotic in the basaltic and carbonate-rich environment of the island), these two deterministic factors were evaluated separately to identify North African dust particles. Three different assessment methods were applied to determine the amount of Saharan dust material of the samples. In contrast to simple grain size and shape parameters of bulk samples, (1) parametric curve-fitting allowed the separation of different sedimentary populations suggesting the presence of more than one key depositional mechanisms. Additional (2) Raman-spectroscopy of manually targeted individual particles revealed a general relationship among grain size, grayscale intensity and mineralogy. This observation was used to introduce the (3) intensity based assessment technique for identification of large number of quartz particles. The cluster and network analyses showed that only joint analysis of size, shape and grayscale intensity properties provided suitable results, there is no specific granulometric parameter to distinguish Saharan dust due to their irregular shape characteristics. The presented methods allowed the separation of Saharan dust-related quartz grains from local sedimentary deposits.

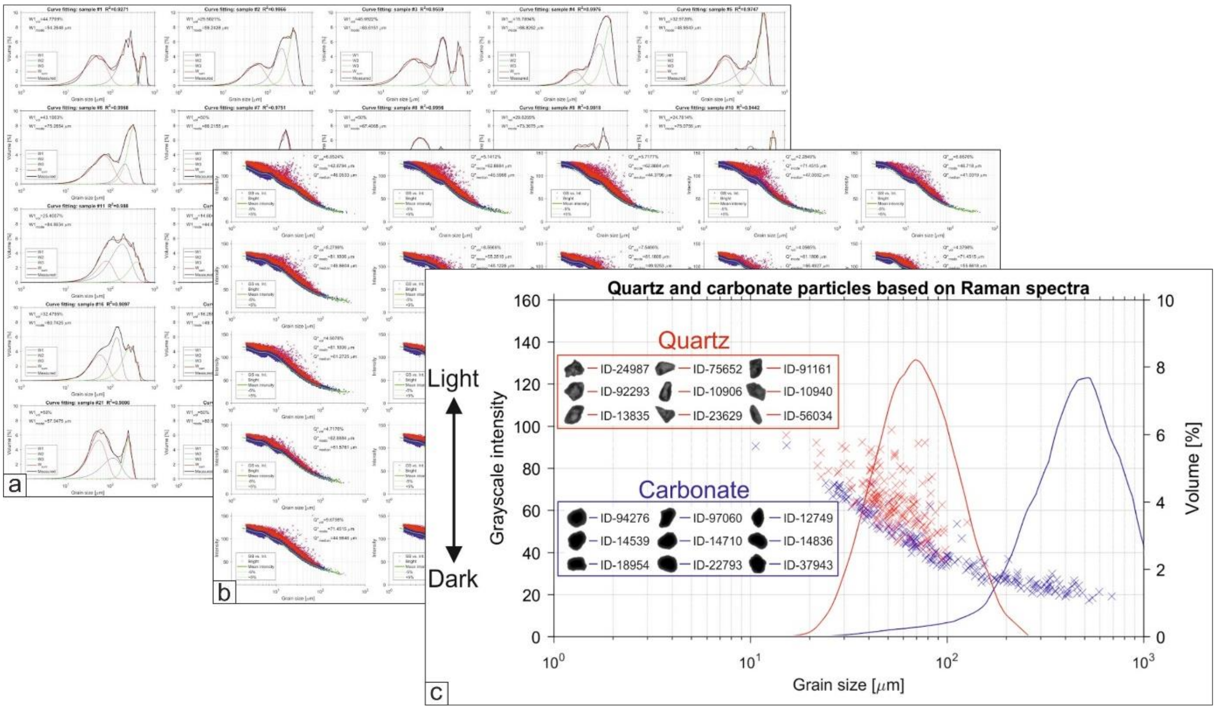

Figure 4. Three different particle characterization methods (a: parametric curve-fitting; b: grayscale intensity-based assessment; c: grayscale and Raman-spectroscopy) to identify Saharan dust particles in Fuerteventura samples.

Cold aeolian environment: Iceland

In our Scientific Reports paper (Varga, Gy., Dagsson-Waldhauserová; P., Gresina, F., Helgadottir, A. (2021). **Saharan dust and giant quartz particle transport towards Iceland**. Scientific Reports 11. 11891), we described Saharan dust storm events identified in the Icelandic region over the past decade and a half, their meteorological background, dust transport routes, possible source areas and general characteristics of the dust particles. The 15 dust storm events identified by remote sensing methods and computer models clearly show that fine-grained mineral dust can travel thousands of kilometres from the Saharan source areas. In fact, particle size and shape analyses of dust collected during two intense events in the Reykjavík area showed that it is not just fine-grained dust of, up to a few tens of microns in diameter that can reach such distances as previously thought, but also large numbers of very large mineral particles of up to 100 microns in size.

We found that the size and shape properties of the dust material from local source areas can be effectively distinguished from each other using this method. Particles of similar size had different shapes and particles of similar shape had different sizes.

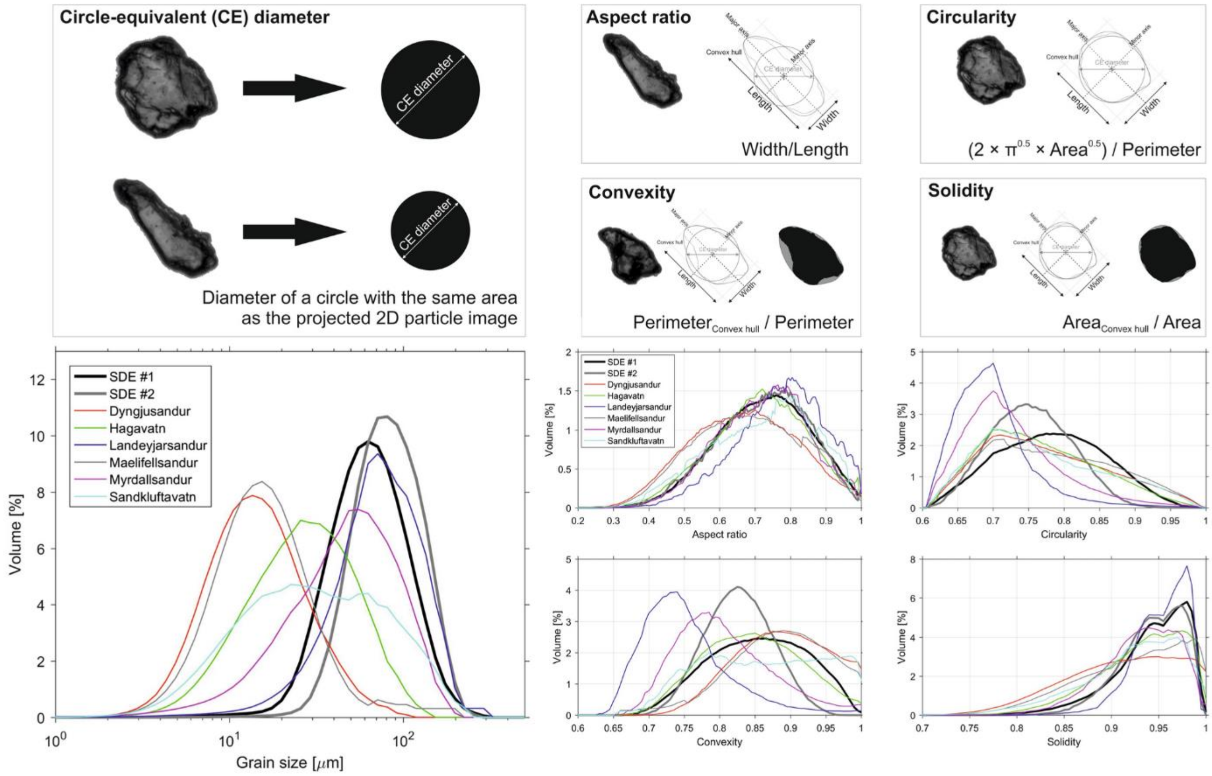

Figure 5. Distribution curves of the grain size and shape of bulk samples from Saharan dust depositional events and Icelandic source areas.

The grain size of deposited modern dust material

Our observations confirmed that the size of **mineral dust particles carried by wind over long distances was previously significantly underestimated**. Our measurements detected large quantities of giant particles in all the regions studied. The role of this in paleoenvironmental reconstructions is that we must assume traces of external dust accretion not only in the material of fine-grained sediment populations, but also in the coarse fractions.

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