Granulometric analysis of recent Saharan dust

KH130337 – funded by NKFIH (National Research, Development and Innovation Office)

Project closing final report by György Varga (PI)

The research project NKFIH KH130337 entitled 'Granulometric analysis of recent Saharan dust' officially ended on 31 August 2021, after a prolongation of 9 months (the project started on 1 December 2018).

The project was aimed at (1) provide complex grain size and grain shape analysis of recent Saharan dust samples; (2) determine the synoptic meteorological background of long-range Saharan dust transport (towards European Alpine areas; Central Europe; Iceland; Canary Islands); (3) discuss the possible relationship of climate change and dust transport pattern changes; and (4) unfold the discrepancies and causes of measured and simulated dust deposition data.

To achieve the above-mentioned objectives, a multi-proxy approach was applied by simultaneous usage of (1) field works and laboratory measurements (complex granulometric characterisation by laser diffraction and automated static image analysis; Raman spectroscopy); (2) remote sensing (aerosol optical depth and aerosol index data of different satellites); and (3) numerical simulations of dust transport and dust deposition.

The results of the project were published in peer-reviewed research papers (published in Sedimentary Geology [Q1 – IF: 3.244]; Environment International [Q1/D1 – IF: 9.621]; Scientific Reports [Q1/D1 – IF: 4.379]), and were presented at major international conferences (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; 16th Carpathian Basin Conference for Environmental Sciences [invited plenary lecture]), 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.hu/p/nkfi_20.html.html and http://aeoliandust.blogspot.hu/p/nkfih-kh130337.html.

Climate change: more dust?

The research project addresses several aspects of the ongoing climate change issue. Through these studies, we have provided new results on the relationship between increased atmospheric meridionality – i.e. the increased dominance of south-north flow systems – and the increased warming of the Arctic, as well as the resulting decrease in the temperature difference between higher and lower latitudes and changing atmospheric flow systems. Reliable instrumental measurements have been available since the 1880s. Since then, the global average temperature has risen by almost 1 degree Celsius. The vast majority of this warming has occurred in the last 10-15 years, and its spatial distribution is not uniform: warming in polar regions is several times greater than the temperature change at lower latitudes (a process known as Arctic amplification). The evolution of Rossby waves, which are responsible for cyclonic activity in the temperate zone, varies with the meridional temperature difference: the smaller the difference, the slower the formation of high atmospheric waves with larger amplitudes. Because of these large-amplitude swings, choppy flows from the desert areas of North Africa can carry large amounts of dust northwards (like blowing the dust off a table with a strong wave motion of the tablecloth). This can sometimes be seen across Europe, and a link has been found in Hungary between the meandering jet stream and the increasing amounts of Saharan dust being transported into the Carpathian Basin. What is particularly interesting, however, is that Saharan dust can travel as far as Iceland on so many occasions.

Giant Saharan dust particles and the climate system

The significance of the research project is further enhanced by the fact that the analysis of hundreds of thousands of individual mineral particle sizes and fractions has shown that many more large particles are being released into the atmosphere than previously thought. Because of their size, these dust particles – unlike fine-grained dust – absorb rather than reflect radiation from the Sun, so they have a heating rather than a cooling effect. For this reason, the parameterisation of their role in the Earth's energy balance in global climate models needs to be modified.

Samples

Samples were collected from (1) Saharan source areas; (2) Canary Islands (Fuerteventura and Lanzarote); (3) Mediterranean Basin (Crete); (4) European Alpine regions (the Pyrenees, Alps, Carpathians); (5) Carpathian Basin; and (6) Iceland. Due to the ongoing global pandemic of coronavirus disease (COVID-19), a large number of samples were gathered from local colleagues (primarily from members of COST inDust Project).

Sampling sites for field works in the Chott Melrhir and Chott Jerid area were assigned together with Nadia Gammoudi (Tunisian PhD-student supervised by the PI of the project), and samples were also collected from the area (by her) for analyses. Mineral dust samples were collected from Alpine regions of Austria (around the Sonnblick Observatory, Hohe Tauern), while additional (snow and quartz fiber filter) samples from the region were provided by Anne Kasper-Giebl (TU Wien) and Marion Greilinger (ZAMG, Austria). Granulometric analysis of samples deposited Saharan dust material from the Pyrenees (by Jorge Pey, Pyrenean Institute of Ecology, Spain) and North African source area samples (from Chad, Egypt, Libya, Mauritania, Tunisia, Western Sahara by Zongbo Shi and Clarissa Baldo, University of Birmingham). Long-range transported dust (as well as local source area samples from Iceland [Hagavatn, Sandkluftavatn, Landeyjarsandur, Maelifellsandur, Myrdallsandur and Dyngjusandur]) were characterised from rare Icelandic Saharan dust episodes.

Samples from the Carpathian Basin were collected from the systematic monitoring campaign of Saharan dust episodes. Additionally, this monitoring campaign was completed with the analyses of Saharan dust material extracted from precipitation sampling at Csopak in 2016 by Ágnes Rostási (University of Pannonia).

Methodological aspects – granulometry of mineral dust

General overview

Accurate determination of grain size of clastic sedimentary deposits has attracted great interest in Earth sciences, especially in sedimentology and sedimentary geology, while particle size data in atmospheric sciences was treated as a secondary problem. Granulometric data provide valuable information on the physicochemical environment of sediments from particle mobilisation to transport and deposition. Grain size measurements are particularly important for fine-grained windblown deposits when particle sizes fall into a fairly narrow range. The selective nature of aeolian transport, owing to the relatively low energy and low-density transport medium, results in a unique granulometric fingerprint left on aeolian dust and dust deposits.

Particle size measurements and grain size distributions of mineral dust can be used for a wide range of scientific purposes. However, the specific diverse aspects of aeolian sedimentation, including wind strength, distance to source area or multiple source regions and modes of transport can only be isolated and reconstructed in favourable cases, and using precise and appropriate grain-size data.

A large variety of instrumental techniques for particles size measurements are accessible in the field of sedimentology. 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

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, the 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 characterisation 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 published study (Varga et al., 2019), particle size data of dust deposits are 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.

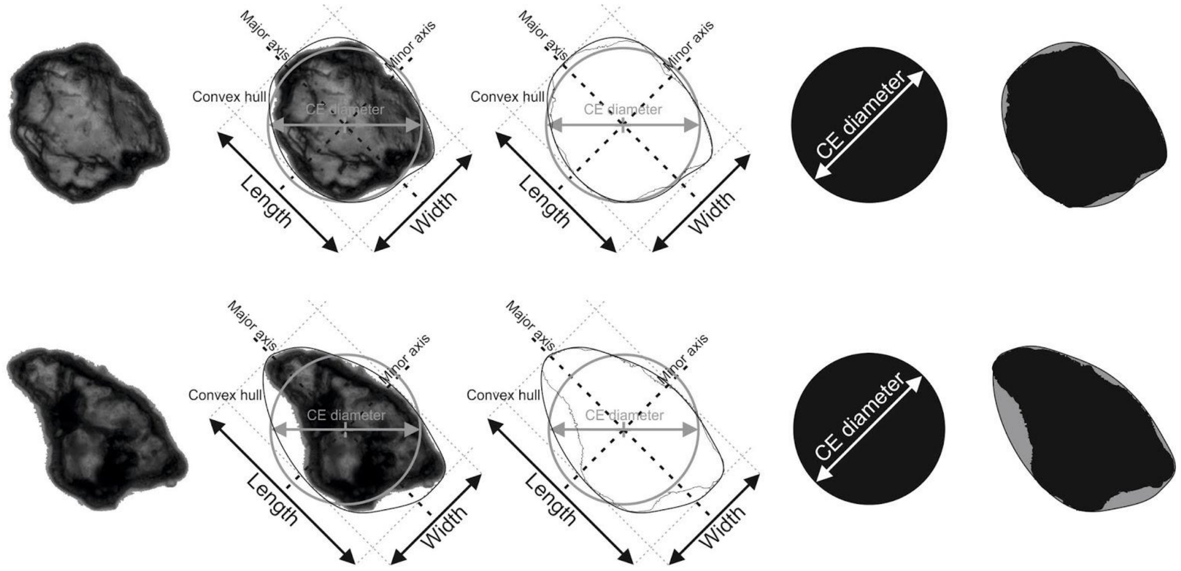
One of the most important findings was that calculations with the Mie theory provide more accurate data on the GSDs of dust deposits. Significant differences between the Mie and Fraunhofer approaches were found for the finest grain size fractions, while only slight discrepancies were observed for the medium silt fractions. Since the two approaches gave similar results for the medium to coarse silt fractions, 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 environmental studies.

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

Direct optical image-based granulometric measurements of mineral particles provide a unique opportunity to determine the size and shape parameters of dusty material. The automated microscopic approach allows the characterisation of hundreds of individual mineral particles, thus providing the opportunity for a robust statistical analysis.

In our studies, a Malvern Morphologi G3-ID advanced automated static image analyser was used. 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).



Aspect Ratio = Width/Length

CE Diameter: diameter of a circle with the same area as the projected 2D particle image

Circularity = $(2 \times \pi^{0.5} \times \text{Area}^{0.5}) / \text{Perimeter}$

Convexity = $\text{Perimeter}_{\text{Convex hull}} / \text{Perimeter}$

Elongation = 1 - (Width/Length) = 1- Aspect Ratio

SE Volume: volume of a sphere with the same CE Diameter as the projected 2D particle image

Solidity = $\text{Area}_{\text{Convex hull}} / \text{Area}$

Fig.1. Main particle size and shape parameters of individual mineral dust particles.

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 identify quartz particles among the targeted sedimentary grains.

Developed complex Saharan dust event identification procedure

Several billion tonnes of mineral dust is emitted every year from arid-semiarid areas and are transported up to several thousand of kilometres by winds. North African dust hot spots located in the Sahara and Sahel contribute to 50–70% of the global mineral dust budget. Dust-loaded air-masses originated from these sources also affect remote areas; vast amounts of mineral particles are transported to North and South Americas across the Atlantic Ocean, toward the Middle East, and in the direction of Europe.

Satellite measurements

For appropriate monitoring of recent Saharan dust events, aerosol products of several satellite campaigns were used. The long-term daily aerosol measurements of NASA's Total Ozone Mapping Spectrometer (TOMS Nimbus-7 TOMSN7L3 v008; TOMS Earth-Probe TOMSEPL3) and Ozone Monitoring Instrument (OMI – Daily Level 3 Gridded Products; OMTO3d – source: NASA Goddard Earth Sciences Data Information Services Center (GES DISC) via Giovanni online (Web) environment for the display and analysis of geophysical parameters https://giovanni.gsfc.nasa.gov/) were applied. The TOMS Aerosol Index (AI) and OMI's TOMS-like AI measure the relative amount of aerosols based on the differences in the measured backscattered ultraviolet radiation of the atmosphere (containing aerosols) and a calculated pure molecular atmosphere. Its positive values indicate absorbing aerosols (dust, smoke from biomass burning, and volcanic aerosols). As a result of the geographical location of the investigation area (Carpathian Basin is situated far from active volcanoes, and in common agricultural practices in the European countries, biomass burning is not applied), positive values mostly indicate mineral dust.

As the satellite-based dust detection over land surfaces, especially in mid- and high-latitude regions, is rather complicated due to local aerosol emissions and cloud cover, additional confirmation is needed to identify SDEs. Possible SDE-days were initially selected based on the AI_{st} values, but SDE-days were only accepted after being confirmed by backward-trajectory calculations, where the Saharan surface origin had to be established both from the path of the trajectories and from the vertical profiles of air-mass transport. Multiple endpoints from different heights (1500, 3000, and 4500 m a.s.l.) were used during the 72–120 h backward-trajectory analyses, performed by NOAA HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) model to determine the main dust transportation pathways. The meteorological input for the trajectory model was also obtained from the NCEP/NCAR (National

Centers for Environmental Protection/National Center for Atmospheric Research) Reanalysis Project dataset. Atmospheric presence of mineral dust was also verified by true colour satellite images of NOAA Advanced Very High Resolution Radiometer (AVHRR), ESA Meteosat Spinning Enhanced Visible and Infrared Imager (SEVIRI), or Terra/Aqua Moderate Resolution Imaging Spectroradiometer and Aerosol Optical Depth data (NASA MODIS Aqua/Terra), from 2000 dust models (BSC-DREAM8, NMMB/BSC-Dust-model, and SKIRON), and surface observations at the proposed source areas (visibility-reducing surface weather reports of Naval Research Laboratory: https://www.nrlmry.navy.mil/aerosol/#aerosolobservations) were also applied in the verification procedure. Verification of episodes from the ’80 s and ’90 s was based on much fewer available data sources, but AI_{st} and back-trajectories were used as the basis of identification.

Numerical simulations

In contrast to the dust load, there is much less information regarding dust deposition. Although dust has long been observed in the Mediterranean and Europe, it remains uncertain about how much dust is transported to the region and how much and where the dust is deposited. There are very few direct measurements of Saharan dust deposition in Europe. Therefore, estimates of dust flux were performed via model calculations using the data of BSC-DREAM8b (Barcelona Supercomputing Center’s Dust REgional Atmospheric Model) v1.0 and v2.0 and Non-hydrostatic Multiscale Model NMMB/BSC-dust models and a mineral dust model database. Simulation results of the BSC-DREAM8b v1.0 are available from 1 January 2000 to 31 December 2012, whereas the results of the updated v2.0 calculations are ready for the period between 1 January 2006 and 31 December 2014. The BSC-DREAM8b models predict the atmospheric residence of the eroded fine-grained aeolian material by solving Euler-type partial differential non-linear equations. The meteorological fields are initialised every 24 h, while the boundary conditions are updated every 6h. The available numerical time series is short; however, it has to be considered because modelled daily values of BSC-dust models have already been proven to represent the atmospheric dust load well and the simulated values are the only useable quantitative data sources of daily dust deposition in the area.

NASA’s Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2) provides modelled monthly data from 1980. Summarised dry and wet deposition data of the five available dust size-bins of the model were obtained from NASA Goddard Earth Sciences Data Information Services Center (GES DISC) via Giovanni application for visualisation and access Earth science remote sensing data (https://giovanni.gsfc.nasa.gov/giovanni/).

Synoptic background

To define the synoptic meteorological patterns associated with dust intrusion episodes in the investigation area, mean geopotential height (700 hPa), wind vector, meridional, and zonal flow maps as well as a 250-hPa jet stream wind flow map were compiled for the dusty days using the Daily Mean Composite application of NOAA Earth System Research Laboratory (http://www.esrl.noaa.gov/psd/). According to previous studies, the 700 hPa level represents the typical dust transport altitude.

Days with meridional wind components exceeding 10 and 15 m/s at 700 hPa were investigated for every decade and season to uncover possible strengthening of the atmospheric flow meridionality in Central Europe and in the Central Mediterranean. Thresholds were defined based on the compiled meridional flow maps of individual SDEs, where 10 and 15 m/s isotachs indicated the strongest flow and main pathways of dust transport. Surface air temperature anomalies (based on 1981–2010 climatology) were also calculated using the gridded NCEP/NCAR (National Centers for Environmental Protection/National Center for Atmospheric Research) Reanalysis Project dataset.

Saharan dust in the Carpathian Basin

As a result of the systematic analysis of satellite measurements completed with the above-detailed verification process, 218 SDEs were identified between 1979 and 2018 (Varga, 2020).

SDEs were classified into three main synoptic meteorological types based on the daily 700 hPa geopotential height, wind maps, and dust transport pathways of SDEs. The different types were defined by specific deterministic atmospheric patterns: Type-1 SDEs were connected to deep atmospheric troughs over Western Europe and north-western Africa; dust transport of Type-2 episodes was caused by Central Mediterranean cyclones; Type-3 events were defined based on the rare dust transport when dust-loaded air-masses approached the Carpathian Basin from the north-western directions.

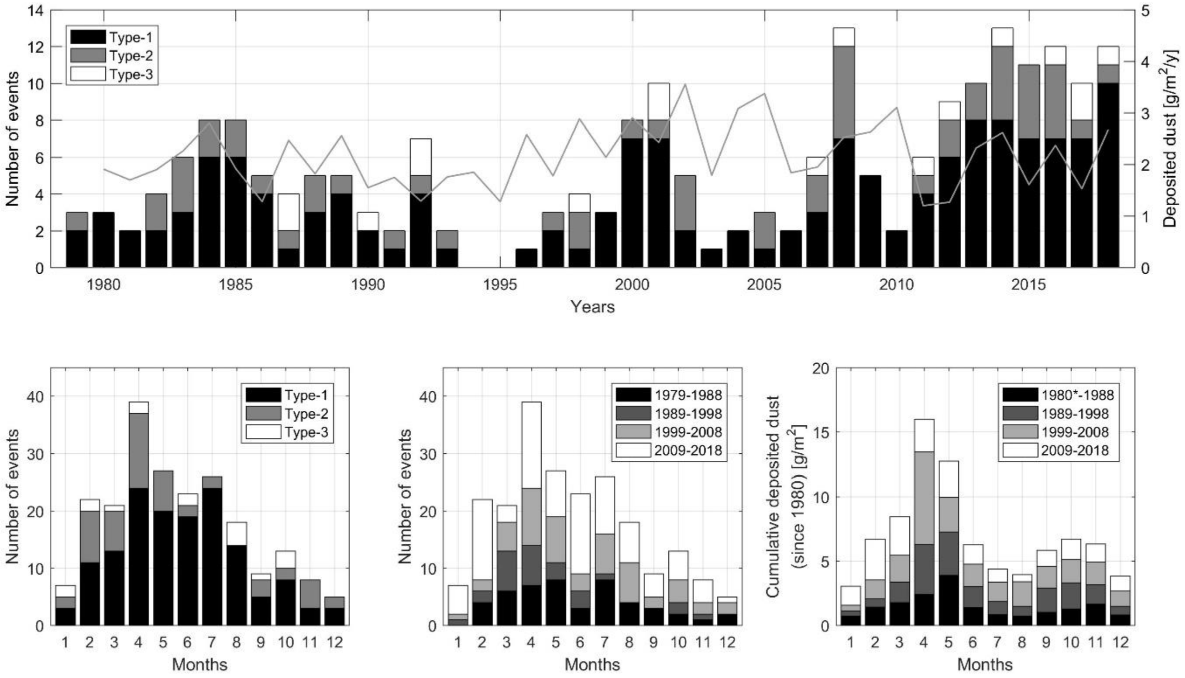


Fig.2. Annual and monthly frequencies and deposition rates of Saharan dust events.

In our previous study (Varga et al., 2016), data of BSC-DREAM8b (Barcelona Supercomputing Center's Dust REgional Atmospheric Model) v1.0 and v2.0 dust models and the mineral dust model database was used to assess the deposited Saharan dust. These modelled values corrected by a few direct surface observations of published European measurement campaigns indicated an average annual dust deposition of 3.2–5.4 g m^{-2} in the Carpathian Basin (Varga et al., 2016). MERRA-2 modelled data of the present study showed a slightly lower annual mineral dust deposition in Central Europe (1.2–3.55 g m^{-2}), dominated by wet deposition (77–93% mass of the total). The temporal distribution of Saharan dust deposition coincides with the seasonal pattern of Type-2 events (Mediterranean cyclones bring precipitation to Central Europe) by a dominant spring maximum with relatively high values in autumn and in February (especially from 2003).

Interannual changes of the number of SDEs and deposited dust material in the Carpathian Basin are obvious; however, no direct relationship among these frequencies and other climatic parameters (annual mean temperature, precipitation, and North Atlantic Oscillation phases) could be revealed. There is no general trend in the number of identified events; however, an increase in the annual occurrences in the last decade is rather apparent. In addition, the seasonal distribution of deposition changed in the last quarter of the studied period, and almost 25% of the Saharan dust deposition occurred during winter.

Intense dust deposition events

Several considerably intense Saharan dust depositional events (SDDEs) were recorded after 2014. These were identified based on reported surface observations of mineral dust washout situations from the study area. An exceptionally large amount of mineral dust material was washed out during these episodes, which also attracted the attention of local society and (social) media. All of these SDDEs occurred between the end of October and February; before 2009, these episodes could be regarded as unseasonal episodes.

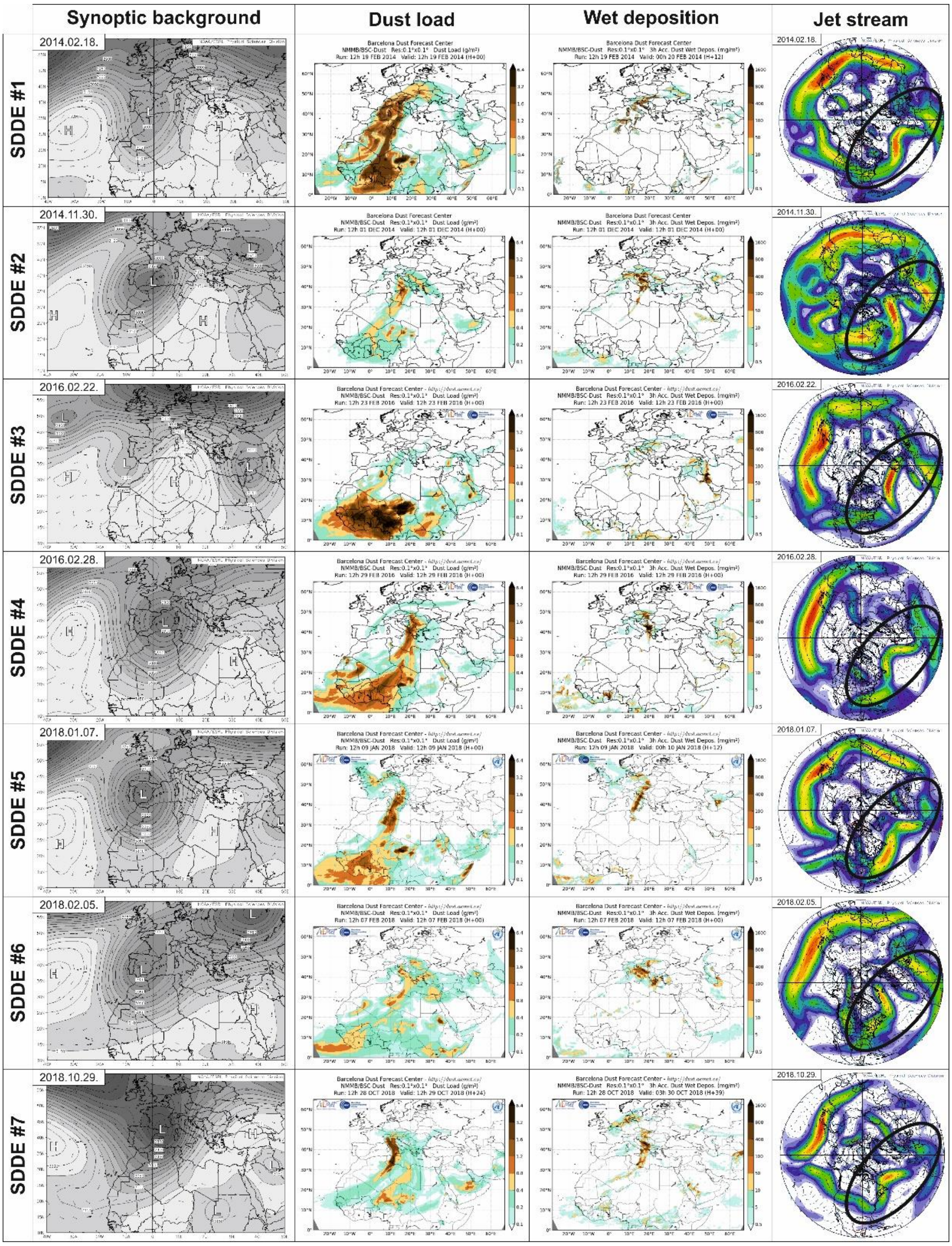


Fig. 3. General properties of the discussed intense Saharan dust depositional events (geopotential height and wind vectors at 700 hPa; modelled dust loading (NNMB/BSC); modelled wet deposition (NNMB/BSC); jet patterns (wind vectors at 200 hPa)).

Seasonal intensity changes of meridional wind components were studied for the four investigated decades. The number and relative proportion of days when the speed of meridional wind components exceeded 10 and 15 m/s at 700 hPa showed a dominance of fall in both cases, but more clearly for higher wind speeds. Summers are represented by a relatively low number of such days, but springs and winters provide suitable conditions for gusty southerly atmospheric flow patterns. Decadal variability of days with enhanced (>15 m/s) meridionality was rather significant, partly because of the low number of these days; however, the increased relative proportion in the winters of 2009–2018 (48.5% of the total decadal episodes) is remarkable.

The grain size of deposited dust material

The deposited mineral dust samples of the above presented SDDEs were characterised by particle size and shape analysis techniques using automated static image analysis and proved the presence of a larger volumetric proportion of medium (6.25–20.00 µm) and coarse silt-sized (20.00–62.50 µm) particles.

Occasionally, fine particles stacked on each other to form larger aggregates. This phenomenon was observed during the measurements of the samples of SDDE #1 (19–20 February 2014), when the laser diffraction measurements resulted a modal grain size of particle size distribution at 6.3 µm; however, images analysis results showed the clear presence of a larger quantity of coarse silt-sized aggregates, indicating that these Saharan minerals were not transported as individual fine-medium silt-sized particles. Large aggregates were accidentally dispersed during laser diffraction measurement, leading to the underestimated grain sizes. The dispersed grain sizes cannot be representative of the strength of the transport agent (e.g., wind speed); therefore, appropriate, non-destructive sizing technique should be chosen.

In other cases, a higher individual grain per aggregate ratio could be identified with a large proportion of single mineral grains (quartz, feldspar, calcium-carbonate, and dolomite) with a particle diameter over 30 µm (e.g., during SDDEs #2 and #3; 22 and 26 February 2016), and even larger ratios were observed during SDDE #6 on 7–8 February 2018. At this moment, because of the scarce homogeneous grain size data and general lack of grain shape data of Saharan dust source areas, source appointment cannot be performed solely based on granulometric data.

The published characteristic particle sizes of Saharan dust materials deposited in Europe are in the range of 2 to 30 µm. Particle size data (various single statistical descriptors were used) measured by different analytical techniques: Crete: 8–30 µm (modal), 4–16 µm (median); Spain: 4–30 µm (mean); Germany: 2.2–16 µm (median); Italy: 16.8 µm (modal), 14.6 µm (median); South France: 4–12.7 µm (median), 8–11 µm (median); France (Paris Basin): 8 µm; Swiss Alps: 4.5 ± 1.5 µm (median); and Central Mediterranean: 2–8 µm (modal). The identified Saharan quartz particles from the geological samples collected from Fuerteventura, Canary Islands (located significantly close to the African continent in the Atlantic Ocean, 100 km west of Morocco), showed ~ 70 µm modal and 68.5 µm median values (Varga and Roettig, 2018, Roettig et al., 2018).

Wintertime dust events

All of the identified unusual dust events characterised by severe washout of mineral dust material in the Carpathian Basin were related to very similar synoptic meteorological situations. The first phase of the dust storm development was an enhanced southward propagation of a high-latitude upper-level atmospheric trough. The orographic blocking of Atlas Mountains played a vital role in the formation of severe surface wind storms and dust entrainment. The low-level winds and dynamics are associated with the penetration of the atmospheric trough. The northward dust transport across the Mediterranean towards Central and south-eastern Europe was also related to the main flow patterns and the eastward-moving low-pressure system.

The occurrence and southerly penetration of high-latitude high-level atmospheric trough to low-latitudes and the increased meridionality of dominant flow patterns have been associated with the remarkable southerly meander of the jet stream, often associated with Arctic amplification (AA). AA is the enhanced warming of high-latitude regions compared to mid- and low-latitudes. AA, along with its alternative metric, the difference in the 1000–500 hPa thickness change in the Arctic relative to that in mid-latitudes, is leading to more (less) meridional (zonal) flow at high altitudes and increasing planetary wave amplitudes.

Francis et al. (2018) identified the importance of more meandering polar jets in the Saharan cyclone formation and poleward mineral dust transport from North Africa. According to their findings, in winter, the dust storm formation at the lee side of Atlas Mountains is generated by high-latitude upper-level troughs and associated low-level dynamics, as well as meridional temperature gradient-driven development of Saharan cyclones. Therefore, the Saharan cyclone formation and poleward dust transport from Africa are affected by the increasing amplitude of planetary waves.

General airflow patterns of severe Saharan dust depositional events of the last decade in the Carpathian Basin, as well as increased frequency of gusty meridional flows and enhanced warming of high latitudes of Northern hemisphere, coincide. Thus, here, we confirm the findings of Francis et al. (2018), that the increasing occurrence of extreme Saharan dust events in Central Europe has been associated with enhanced warming of the Arctic, thereby leading to more meandering jet streams.

Grain size uncertainties-driven deposition data issues

The general increasing trend of numbers of SDEs and the increasing wintertime amount of deposited dust material should not necessarily correspond to the increasing intensity of dust events, and even the opposite can be concluded from the raw data. Intensification, however, was indicated by direct surface observations of intense dust washout episodes when the deposited reddish-yellow dust material had blanketed parking cars, roof-windows, and other exposed obstacles. Numerical simulations of dust deposition also revealed a wintertime increase of the number and magnitude of dust episodes. Nevertheless, the extent of it was not equivalent to the growth of the SDE numbers. A few previous studies confirmed the significant underestimation of deposited mineral dust material by numerical simulations. Quantitative values of valuable but scarce surface measurements and satellite-based assessments have been several orders of magnitudes larger than the modelling results, but the spatial and temporal (e.g., interannual, seasonal) patterns of dust deposition have been properly simulated by the dust models.

According to our suggestions, grain size should be the key to resolve this contradiction. Several recent papers reported the measurements of giant mineral particles found in far-travelled mineral dust. Our automated static image analyses of Saharan dust material in the Carpathian Basin also indicated that the grain size of transported Saharan dust material could be significantly larger than the grain sizes predicted by the model (Varga et al., 2016). Mineral grains of>20 µm are usually not accounted for in the numerical simulations (in the majority of global and regional dust models, only some size-bins of a relatively narrow size range are applied). Because of the cubic relationship between particle diameter and volume (mass), even a small change in applied size-bins can lead to a significant increase of modelled dust fluxes. A deeper understanding of grain size-related uncertainties would also be useful to explore the relationship between uncertain granulometric parametrisation in retrieval algorithms of satellite observations and mineral dust flux estimations.

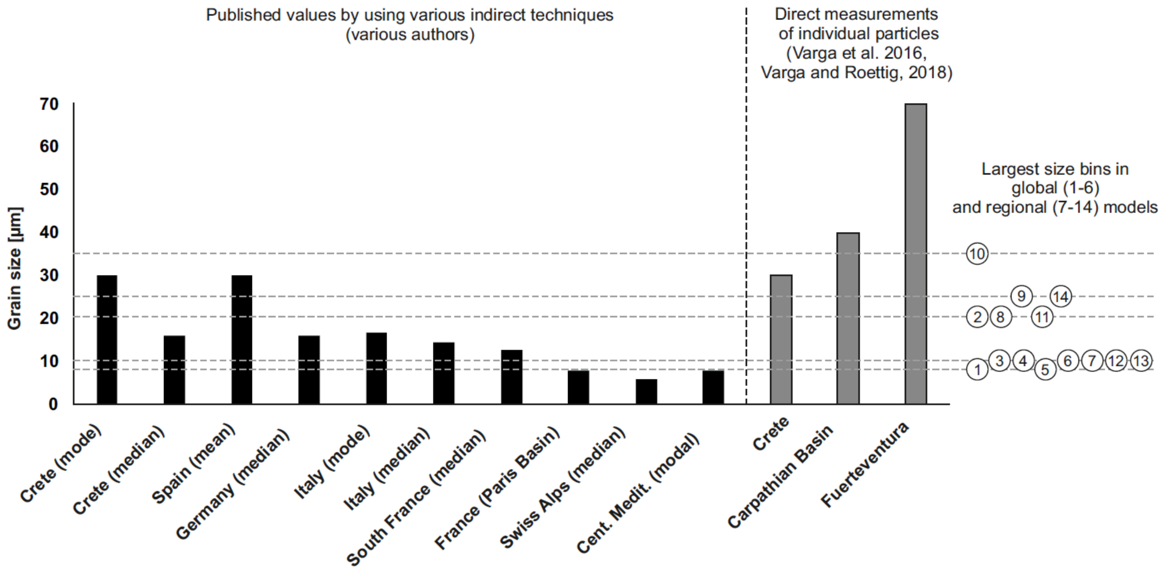


Fig.4. Reported grain size values of Saharan dust in the Mediterranean (indicated models: 1: GEOS-5; 2: MACC_II; 3: MASINGAR; 4: MetUM; 6: NGAC; 7: NMMB/BSC-Dust; 8: CHIMERE; 9: CMAQ-KOSA; 10: COAMPS; 11: CUACE/Dust; 12: BSC-DREAM8b; 13: DREAM8-NMME-MACC; 14: TAQM_KOSA;).

In addition, the physical background of the long-range transport of such a giant mineral material is a matter of debate in the scientific literature. This lack of understanding of driving mechanisms makes the evaluation of radiative forcing of atmospheric mineral dust difficult because the net climate effect of dust (scattering and absorption) depends on grain size distribution; larger particles act like greenhouse gases by absorbing and emitting longwave radiation and have a heating effect, whereas fine dust cools the atmosphere.

Giant Saharan dust in Iceland

Fifteen (#1–15) Saharan dust events were identified in Iceland between 1 January 2008 and 29 February 2020. A systematic combined analysis of Moderate Resolution Imaging Spectroradiometer (MODIS) aerosol optical depth (AOD) data and Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) data resulted in the identification of nine of these fifteen dust events. These nine episodes matched distinct AOD peaks. The remaining six episodes were determined based on the numerical simulations of the NASA's Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2) and Barcelona Supercomputing Center (BSC), and verified using satellite images and backward trajectories of the HYSPLIT model.

All 15 dust episodes were independently examined by analyses of Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) vertical aerosol subtype profiles. The presence of dust over Iceland was confirmed by the lidar data for all 15 episodes. Among the events, two rare wet depositional episodes were detected; in these cases, the sampled Saharan dust materials were analysed using automated static image analysis.

Based on geopotential height maps and wind vectors at 700 hPa, two different synoptic meteorological situations were distinguished. The Omega block-like pattern determined the Saharan dust transport of nine episodes. The high-pressure centre of the blocking system was located over Western–South-western Europe. The steepest pressure gradients and connected strongest flows were situated at the southwest side of the anticyclonic system. Synoptic patterns of the remaining six events were associated with a low-pressure system situated over (or close) to the Iberian Peninsula. The highest wind speeds were observed at the foreside of the stationary cyclone. Central and Eastern European blocking systems were also responsible for the development of the extended meridional atmospheric ‘conveyor belt’.

The most important dust sources can be regarded as NW African dust hot spots, like (1) the ancient lakebed deposits of the Taoudenni Basin, (2) large alluvial fans and extensive wadi-system on the western and north-western slopes of the Hoggar Mountains (Ahaggar Plateau and foothills), (3) intramountain sediments of the Atlas Mountains, and (4) coastal sebkhas of Western Africa.

Saharan dust depositional events in Iceland

The general meteorological background and related transport routes, as well as the possible source areas of the two depositional events, in Iceland were very similar. The first deposition event (SDE #1) occurred during the last days of March 2014, when a low-pressure system governed the atmospheric patterns of the eastern basin of the North Atlantic. The cyclonal flow of the system reached Northwest Africa, where the steep pressure gradient between the mid-latitude low and Saharan highs triggered sand and dust storms. A high amount of lofted mineral dust was transported northwards along the eastern flank of the stationary atmospheric trough system blocked by the Central European ridge. Dust material reached the atmosphere of France on 28 March 2014 and consequently Great Britain and Ireland on 29 March 2014. Intense wet deposition of dusty material and ‘blood rain’ events were reported for the affected areas. Atmospheric dust was deposited in Iceland on 3 April 2014, as shown by wet deposition in the Icelandic domain in the dust numerical simulations of the NMMB/BSC (Non-hydrostatic Multiscale Model/Barcelona Supercomputing Center).

The second deposition event (SDE #2) occurred on 25 April 2019. The deep cut-off low on 20 April 2019 (closed circulation system from an upper-level trough in the antecedent days) determined the synoptic situation of North Africa, and the cyclonal flow lofted an enormous mass of desert dust into the atmosphere. In the following few days, the atmosphere from Africa through Crete over Europe to Iceland was loaded with an enormous amount of Saharan dust, reaching a PM10 concentration of > 150 µg m−3. The blocking mechanism of an Eastern European anticyclone was responsible for the stationary behaviour of the atmospheric system. Saharan dust arrived over Iceland on 24–25 April via a transport route (from the southern forelands of the Atlas Mountains through the Iberian Peninsula and across the British Isles towards Iceland). The pathway was very similar to that of SDE #1.

Granulometry of SDE samples compared to local Icelandic dust source samples

Detailed laboratory analysis of the samples was necessary to distinguish between local and remotely transported particulate matter and to exclude possible local dust addition and contamination of the deposited dust material. An automated static optical image analysis of several thousand individual particles was performed to obtain a robust granulometric characterisation based on the distribution curves of grain size and shape. The grain size distribution curves of deposited material during the SDEs in Iceland showed clear unimodal distributions, which fell predominantly into the coarse silt (20–62.5 µm) and (very) fine sand (62.5–(125)–250 µm) particle size fractions. The modal grain sizes, median diameters, and mean diameters D[3,4] were 62.9 µm and 81.2 µm, 65.7 µm and 87.0 µm, and 70.6 µm and 89.3 µm for SDE #1 and SDE #2, respectively.

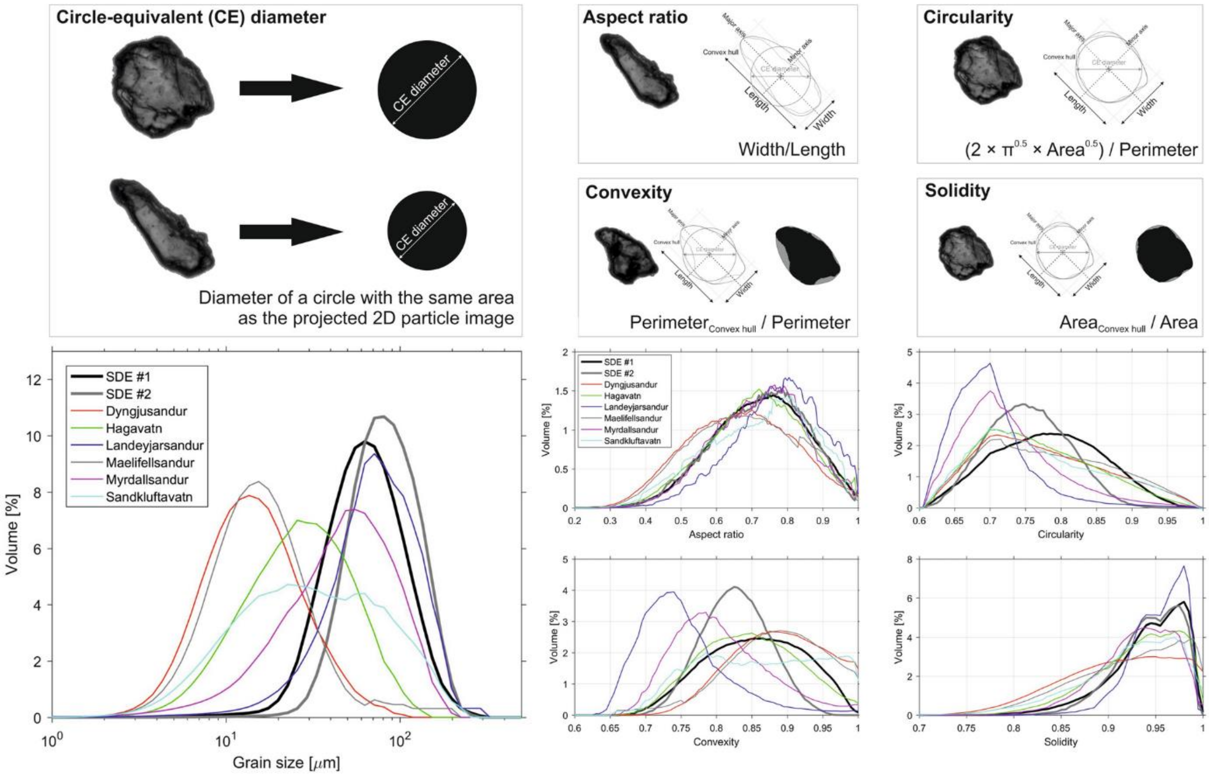


Fig. 5. Distribution curves of the grain size and shape of bulk samples from SDEs and Icelandic source areas.

The circle-equivalent (CE) diameters of bulk samples from Icelandic sources were diverse. The grain sizes of samples from Landeyjarsandur and Mýrdalssandur fell into the coarse silt/fine sand fractions, whereas the particles of other samples were much smaller, especially the samples of very fine-grained deposits from Dyngjusandur and Mælifellssandur. The grain shape parameters of automated static optical image analysis are size-dependent variables (direct comparison of shape parameters of particles with significantly different sizes do not provide realistic results). Thus, particles in size ranges of the SDE samples (i.e. 20–250 µm) were selected to create adequate conditions for a representative particle shape comparison. The SDE samples exhibited a more convex and circular particle shape compared to the local Icelandic samples. The general patterns of the aspect ratio distribution curves were similar, but the solidity curves clearly indicated major differences among the different sample groups. The SDE samples and Landeyjarsandur (a sandy beach that is not directly affected by glacial processes) sample presented similar curves.

Cluster analyses of different size and shape parameters were also performed. Based on the grain size distribution and solidity shape parameter, the SDE samples were clustered close to Landeyjarsandur, whereas other Icelandic dust hotspots consisted of significantly finer grains. The analyses of other major shape parameters (aspect ratio, convexity, and circularity) resulted in various trends for different materials, with the solidity parameters showing a similar clustering for the same particle size-based groups of material. In general, the SDE samples were closely clustered, whereas the Icelandic samples were not; however, there were similarities between the samples from Dyngjusandur and

Mælifellssandur. Irregular circularity and convexity of particles were typical characteristics of material from Landeyjarsandur compared to other Icelandic hotspots.

Raman spectroscopy also confirmed the differences between the materials from SDEs and local sources. Due to the high percentage of particles with nonapplicable Raman signals, robust quantitative analyses could not be made. Our general findings, however, clearly showed that Icelandic surface samples obviously reflected the local basaltic environment and that no quartz particles were found in these samples. Quartz particles, including particles larger than 100 µm, were identified in large numbers in both SDE samples.

Enhanced atmospheric meridionality and climate change

The surface temperature anomaly during the two SDEs compared to the climatological mean for the period from 1981 to 2010 (by applying the reanalysis data of the National Center for Environmental Protection (NCEP) and National Center for Atmospheric Research (NCAR)) was 3.1°C. Enhanced meridionality of the upper-level atmospheric flow was the main synoptic driver of the warm incursions and the identified Saharan dust events. The importance of a more meandering polar jet and associated meridional flow patterns during the formation of the North African cyclone and poleward dust transport was discussed previously in relation to SDEs in the Carpathian Basin. The intense warming of Arctic regions reduces the temperature difference between the high- and mid-/low-latitude areas, thus leading to increasing planetary wave amplitudes and more meridional flow patterns at high altitudes.

The southward propagation of the upper-level atmospheric trough and the orographic blocking of the Atlas Mountains play vital roles in the formation of severe surface wind storms and dust entrainment in the northwest regions of the Sahara. The area of these Northwest African dust hotspots was identified as the main source of the discussed Icelandic dust events; hence, the Icelandic samples were collected approximately 4000–4500 km from their potential Saharan sources.

Long-range meridional transport of giant Saharan dust

It has been shown that coarse particles with a diameter of > 30 µm and giant particles with a diameter of > 100 µm are capable of long-range transport within the Saharan air layer. Previous studies focused on the zonal (east-to-west) transport of giant Saharan dust, whereas meridional (south-to-north) transport towards higher latitudes has only been mentioned in some recent papers. In particular, aerosol studies on the long-range transport of coarse and giant particles to the Arctic are generally lacking. Here, we provide the first evidence of giant Saharan particles deposited in Iceland. Our results confirm the existence of established atmospheric pathways of Saharan dust towards the Arctic, with Saharan dust contributing ~ 32% of the total atmospheric dust load in the Arctic.

The long-range transport of sand-sized and/or giant dust particles, however, is still an open question and little is known about the key drivers of transport mechanisms. Several possible transport mechanisms have been suggested, including strong winds, enhanced atmospheric vertical mixing due to strong turbulence via vertical up-flow of thunderstorms and tropical cyclones, and triboelectrification of dust particles.

Underestimation of the transport and deposition of these coarse mineral particles leads to further biases in the derived climatic effects (and other environmental effects of) aeolian dust (e.g. radiative forcing, which possibly causes net atmospheric warming, and a role in biogeochemical cycles and soil formation). So, the presented size data also has a climatic relevance as the interpretation questions of coarse-sized windblown dust are standing in the focal point of recent studies. Climatic impacts of mineral dust are also sensitive to particle size and shape. Radiative effects of coarse mineral grains in the shortwave spectrum is determined by the lower single scattering albedo compared to finer particles, causing more absorption of solar radiation and so, heating of the atmosphere. Ryder et al. reported an increased atmospheric heating by up to a factor of three when as a result of calculated single scattering albedo dropping from 0.92 to 0.8 when larger particles were taken into account. Kalashnikova and Sokolik showed that the irregular shape character of windblown mineral particles also has an effect on radiative properties via larger surface-to-volume ratio relative to that of an equal volume sphere. According to the calculations of Kok et al., average global dust extinction efficiency results are underestimated by 20–60% for larger than 1 micron particles.

Differences of granulometric properties of Icelandic and SDE samples

Various size and shape parameters of the (1) local Icelandic samples and (2) collected mineral material of the SDEs showed different characteristics. These differences in the two sediment groups were also confirmed by cluster analysis. Clustering of the shape parameters of the SDE and local surface samples showed an outlying nature of the particles from the washout samples. The washout samples mainly contained coarse silt and fine sand-sized mineral dust material, which could be clearly distinguished by their different size and shape parameters in comparison to the samples from local source areas. The more circular and convex shape along with higher solidity values indicated a more mature nature of the SDE particles, which was significantly different to the nature of the fresh volcanoclastic deposits of Iceland. We infer that these granulometric differences could relate to (1) the relatively young geological evolution history of local samples and (2) the initial shape properties and long-range transport processes of Saharan mineral dust particles.

Similar shape features of far-travelled Saharan dust particles have been reported in other studies that applied the same approach and used the same Malvern Morphologi G3-ID device as used in the present study. We are not aware of any other automated static image analysis data of windblown North African mineral dust material (shape data made by other methodological approaches cannot be quantitatively compared).

The well-rounded (but not spherical) shape of coarse silt and fine sand particles (caused by rapid abrasion and edge-rounding) is a known property of windblown deposits; however, sharp edges of finer silt-sized fractions (typical for fine-grained aeolian silts) were also visible on the captured images of individual particles of the SDE samples. Contrary to these properties, the Icelandic samples were much more irregular in shape and dominated by sharp features, even in the case of larger particles.

Mineralogical comparisons of Icelandic and Saharan dust have shown that the latter is mainly comprised of quartz and feldspar, while the former primarily consists of volcanic glass. Mineralogical fractionation is not observed in Icelandic dust due to the lack of larger mineral grains such as quartz. It is clear from the detailed laboratory analyses that the local addition of coarse particulate matter to the sampled deposited particles can be excluded.

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