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COMPARISON OF PIPETTE AND LASER DIFFRACTION METHODS IN DETERMINING THE GRANULOMETRIC CONTENT OF FLUVIAL SEDIMENT SAMPLES

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Abstract

Nowadays there is a growing demand for rapid and accurate determination of grain size distribution. The conventional pipette method is time-consuming and provides less detailed data compared to recently introduced methods. However, in Hungarian practice the pipette method is still considered to be the standard one, as there are a long series of measurements, and grain size thresholds used in sedimentology and soil sciences are based on this approach. The aim of our research was to determine the comparability of the laser diffraction method (LDM) with the conventional pipette method (PM), in order to investigate the controversial question on the interchangeability of the two methods. Based on our measurements on some representative fluvial sediment samples, we found that the largest difference in results can be expected in the silty grain size range. However if the main fractions (clay, silt, sand) are considered the methods provided similar very results, and correlation factors were above 0.92. In all, the LDM has a clear advantage because of its speed, reproducibility and fewer possibilities for operator failure.

Keywords: laser diffraction method, pipette method, fluvial sediments, grain size distribution

INTRODUCTION

Grain size distribution is a fundamental physical parameter in soil and sediment related researches. Physical and chemical conditions of the sediment and soil samples are mostly determined by the main grain size fractions, which contain the majority of the particles. Grain size classes are determined by almost each research field differently (Blott and Pye, 2012). Grain size distribution can be determined in several ways, however in most research applications a fast and unified method is demanded, which offers reproducible and automated grain size measurements.

In the past few decades, several researches have dealt with the methods of grain size distribution (Konert and Vandenberghe, 1997; Buurman et al., 1997; Beuselnick, 1998; Goossens, 2008; Di Stefano et al., 2010; Hernádi et al., 2008 stb.). A controversial question has been which method is the most proper for different applications. The aim of our research was to examine at what extent the conventional pipette method (PM) and the laser diffraction method (LDM) are interchangeable in terms of unsorted fluvial samples.

The PM is internationally accepted for grain size distribution analysis, hence it was used as the basis during the comparison of the two methods. The PM is based on the Stokes law, i.e. sedimentation rate is depending on particle size. There are however some conditions of its applicability: grains are presumed to be spherical and smooth, sedimentation rate must be constant, the density of particles equals to that of quartz ($2.65 g/cm^3$), particle-to-particle interference and boundary effects from the walls of the sedimentation column are negligible and particles have no impact on the viscosity of the suspension (Di Gleira el al. 1957; Konert and Vandenberghe, 1997; Di Stefano et al., 2010). For example, if the first condition is not met the resulted clay content will be highly influenced by the shape of the particles. The settling velocity of the non-spherical grains in the fine fraction can lead to the underestimation or overestimation of the clay content depending on the shape of the particle. Platy shape grains lead to fine fraction overestimation while disc or rod shape grains result the underestimation in the range of 0.1 μm to 100 μm (Di Stefano et al., 2010).

LDM is based on the dispersion and diffraction of a laser beam that is let through the suspension. The dispersion of light creates special diffraction rings on the sensor and the grain size distribution is determined by the position, size and distance between rings. The application of LDM has also got certain conditions (Konert and Vandenberghe, 1998), namely grains are spherical and particle orientation is random throughout the measurement time. However the flow of the measurement medium will likely to determine the orientation of non-spherical particles (De Vos, 2001).

The accuracy of the measurement also depends on the color of the suspension, the mineral composition of particles, the organic material and carbonate content of the sample and the applied measurement theory.

In general two measurement approaches are applied for the LDM, the so called Fraunhofer and Mie theories. The Fraunhofer theory is operating with the portion of light deflection that occurs as a result of diffraction. One major advantage of Fraunhofer theory is lies on the fact that no knowledge of the optical properties of the examined material is required. However, the Fraunhofer diffraction model provides inaccurate result if the size of the particles is less than 10 λ (wavelength of the laser light) (Loizeau et al., 1994; Xu and Di Guida, 2003; Di Stefano et al., 2010). For particles with diameters not significantly larger than the wavelength of the light used, the Mie theory is applied usually for the analysis, in this case, however, the refraction index and the absorption index of the sample must be known. According to Konert and Vandenberghe (1997) the Fraunhofer theory is well suited for non-spherical clay particles and the same conclusion was established by Di Stefano et al. (2010).

Measured grain size distribution is greatly affected by the applied pre-treatment method. The necessity of the removal of organic matter and carbonate content are strongly controversial. Several authors justify the pre-treatment on sediment samples with high organic matter content (7-8%) (e.g. Murray, 2002). According to his study, only pre-treatment with hydrochloric acid and hydrogen-peroxide could provide any degree of reproducibility. Beuselnick et al. (1998) also investigated the effect of organic matter content, and found that the pretreatment with acids was unnecessary in case of samples with low organic matter content, and in spite of the different pretreatment procedures the results had a strong correlation in the 3 main fractions (clay, silt, sand). A similar statement was made by Ryzak and Bieganowski (2011), namely physical (ultrasonic) dispersion can be equivalent to chemical dispersion methods.

MATERIALS AND METHODS

In order to determine the applicability of the LDM method, several authors have performed measurements on soil samples, loess- and marine sediments (Konert and Vandenberghe, 1998; Di Stefano et al. 2010; Ryzak and Bieganowski 2011; Madarász et al., 2012). However unsorted fluvial samples are very rarely studied (Buurman et al., 2001). Consequently, the analysis was performed on sediments obtained from point bars and swales of a Maros River paleochannel, near Sannicolau Mare, Romania. Samples were derived from 5 boreholes from depths of 30, 50, 70, 90 and 110 cm. In all 25 samples were analysed.

Samples were dried on 105 ˚C and sieved at a 2mm mesh size. The organic matter and carbonate content of sediments were also measured. Carbonate content was under the measurement threshold (Scheibler calcimeter) in case of 15 samples, while 10 samples had carbonate

content between 0.42% and 3.35%. Organic matter content was between 0.32% and 2.01 %. Due to the low organic matter and carbonate content, and since the Hungarian standard of the PM (MSZ-08 0206/1-78) does not contain orders of pre-treatment, we considered the removal of these components unnecessary in case of the investigated samples.

The dispersion of the particles was enhanced using sodium pyrophosphate and shaking: 25 g of sample was weighted for the PM analysis, 0.5 g sodium pyrophosphate and 400 ml distilled water was added, then samples were placed in a shaking machine for 6 hours in order to disperse the aggregates into primary particles.

After this pretreatment the suspensions were poured to 1000 ml sedimentation cylinders, which were then filled up with additional distilled water. Based on the schedule of the Khön's table, 10 ml suspension was pipetted and put into a known-mass evaporating vessel, and the following grain size classes were determined <2 μm, 2-5 μm, 5-10 μm, 10-20 μm, 20-50 μm, >50 μm.

For the LDM measurements a Fritsch Analysette 22 MicroTec instrument was applied. Its measurement range is 0.08-2000 μm and it is equipped with 2 linearly polarized lasers: green (λ=532 nm, P=7 mW) and infra-red (λ=940 nm, P=9 mw). During the measurement, samples were homogenized with ultrasonic treatment (f=36 kHz, P=60 W) for 3 min. The grain size distribution was determined at 108 channels. Samples were measured sequentially for 3 times, during the measurement the ultrasonic dispersion was continuous. The difference of the distributions between the 3 measurements was at maximum 3-4% (*Fig. 1*), hence the third measurement was considered as the primary grain size distribution. In order to compare the results of the two methods, intervals of the pipette method were generated out of the continuous distribution curve yielded by the LDM measurements. Grain size distribution analysis was performed by Statgraphics software.
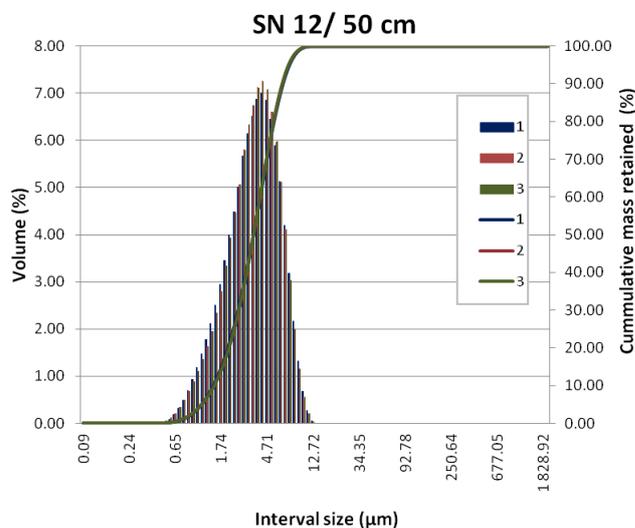

Fig. 1 Differences between three consecutive LDM measurements on the same sample

Comparison of pipette and laser diffraction methods in determining the granulometric content
of fluvial sediment samples

51

RESULTS

The texture of the samples was different, but similar attributes characterized the samples along the boreholes (regardless of depth). The highest clay content was measured in borehole SN5, where nearly identical sand content was measured with the two methods. In the meantime borehole SN11 showed the lowest clay content, while these samples had the highest sand contents. Beyond texture analysis, median diameter (D50) and cumulative distribution with 10, 25, 75, 90% values (D10, D25, D75, D90) were also measured. Differences and ratio of the resulted values derived by the two methods were used for the comparative analysis (*Fig. 2*).

Samples of borehole SN11 – with the lowest clay and highest sand contents – showed the greatest differences of cumulative distribution values (D25, D50, D75, D90) obtained by the LDM and the PM. The sources of the dissimilarity have been searched in the differences of sand contents. During the PM, removal of sand content was performed after sedimentation; therefore presence of sand grains could have disturbed the sedimentation speed of grains of other fractions. Moreover, borehole SN12 contained a large amount of sand, D75 and D90 values were also high.

Differences between D values were the lowest in the samples of boreholes SN3, SN4 and SN5. These samples have the lowest sand content, and amount of sand grains was roughly the same after the measurements of the two methods.

Based on the D values, samples can be divided into two groups: SN3, SN4, SN5 and SN11, SN12. Classification is also confirmed by the spatial distribution of the samples, as the samples of the first group is originated from the point bars and swales of the Aranka river, while SN11 and SN12 were obtained from a 6.1±1.1 ka old, meandering riverbed (Kiss et al., 2012). Measured organic matter content also re-flects this difference of sample groups. The first group (SN3, SN4, SN5) has an average organic matter content of 2.1%, while the second has an average of only 0.8%. Dissimilarities of samples in grain size distribution and organic matter reflects to the river dynamics of the formerly accumulating river. The samples of the first group belong to the clayey and organic matter rich landforms of a meander with a low discharge, while samples of the second group reflects to a considerably large meandering river (Kiss et al., 2012). Since cumulative and differential size distributions of the samples were similar in each group, distributions will be represented by one sample per group.

Comparison of the two methods can be performed by examining the distribution curves. In the case of sample SN3, the shape of distribution curves derived by the two methods are very similar (*Fig. 3*). Results of the LDM were evaluated using the defined fraction intervals of the Hungarian Standard. Curves are bimodal and their main modus is equivalent, however the secondary modus of the LDM curve is displacing towards the coarser fractions. The peak of the LDM curve is at a larger particle size compared to the PM curve.

The differences between the measurements have occurred at different fractions that depend on the proportion of the fractions. The samples SN5 with high clay content shows significant difference in clay proportion, regardless that difference between D values are the lowest in this borehole (*Fig. 4*). The largest difference can be identified in clay contents, confirmed by former statements as the increase of clay content is also increasing the error and probability of underestimations during LDM (Beuselick et al., 1998; Konert and Vandenberghe, 1998).

In case of high clay content the LDM underestimates the clay fraction in favor of the silt fraction. However the PM overestimates the fine fraction because of the non-spherical shape of the clay particles. Underestimation of clay content with LDM was also
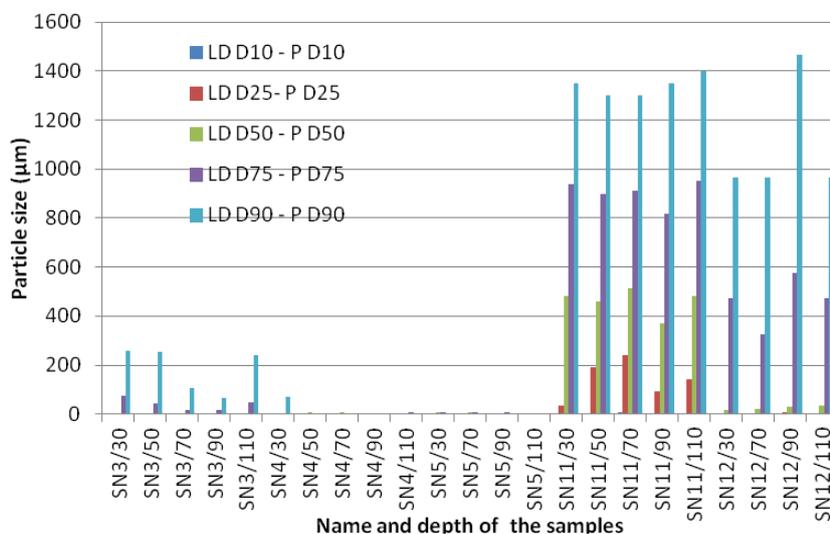

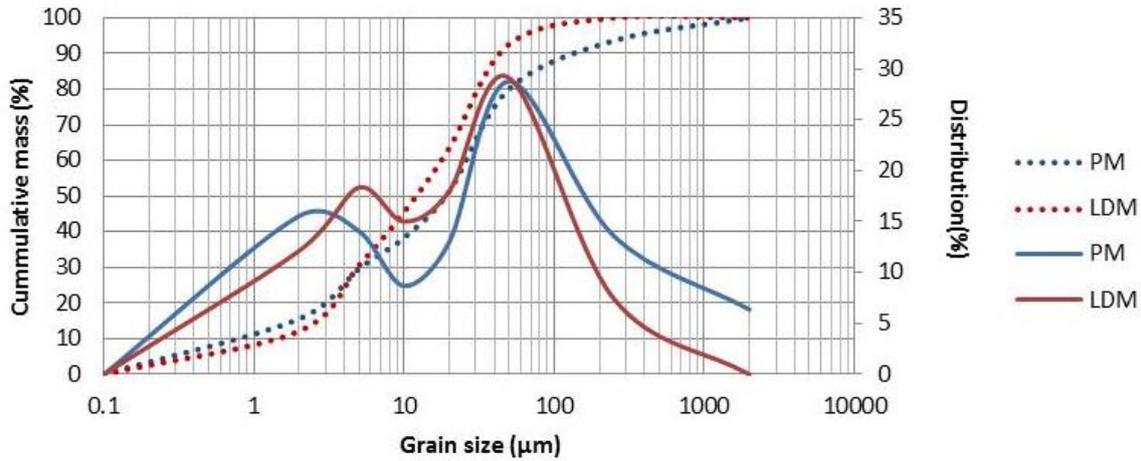*Fig. 2* Difference between the D values derived by PM and LDM

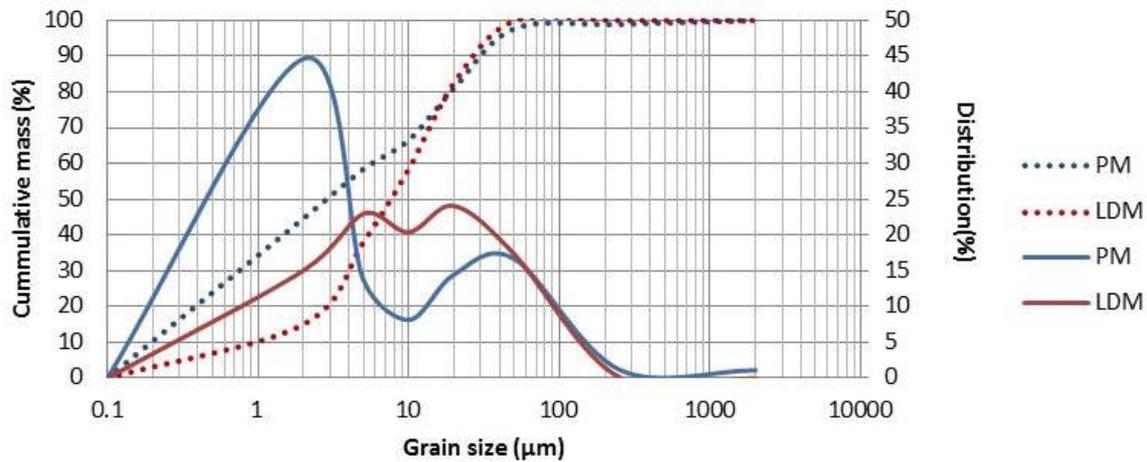*Fig. 3* PM and LDM grain size distribution and cumulative curves of sample SN3/70

Fig. 4 PM and LDM grain size distribution and cumulative curves of sample SN5/110
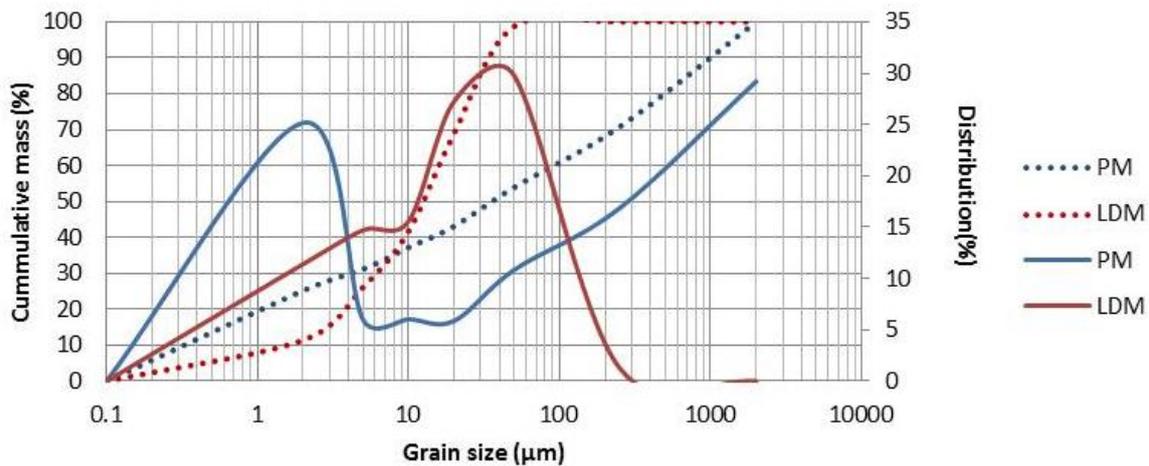

Fig. 5 PM and LDM grain size distribution and cumulative curves of sample SN12/90

typical in other samples of the group. Differences between the LDM and PM are also visible on SN11 and SN12 samples (*Fig. 5*), where the LDM measured lower sand content that can be related to the methodology of sedimentation processes formerly described. Errors during sand content calculations can be explained with the rough surface of grains that leads to the underestimation of sand and therefore overestimation of silt.

Analyzing the results of samples the distribution curves can be very different in certain cases, hence we analysed the result with another approach as well. For every sample we determined the amount of particles in the main fractions (clay, silt, sand) with the two methods, and then we examined the correlation between them. Taking the sediment categories into account, a high correlation was found between the LDM and PM sand, silt and clay content.

Comparison of pipette and laser diffraction methods in determining the granulometric content
of fluvial sediment samples

53

Table 1 Correlation of the main fractions determined by different methods

Size Class (µm)	Texture	Correlation factor	Linear regression
<2	Clay	0.928	Y=0.179·X+6.764
2-5	Silt	0.934	Y=1.036·X+16.841
5-10			
10-20			
>20	Sand	0.951	Y=0.482·X+28.663

Table 1 shows the linear function that is calculated by the amount of particles in the main fractions. Figure 6, 7, 8 shows how the values fit on the linear function per each main interval. There is no case where the values are outside of the confidence interval (95%). The correlation factor is significantly high concerning all main fractions, despite the difference between the dispersion curves based on the two methods. For exploring this difference, we examined the correlation as well in each interval (*Table 2*). The lowest correlation factors were experienced in case of coarse silt (10-20 µm) and the fine sand (20-50 µm) fractions. Based on linear regression, separation of the particles with 10-50 µm diameter is problematic. Several author found the results of this fraction doubtful, hence they expressed the silt fraction mathematically as a function of the known clay and silt fraction (Beuselnick, 1998; Buurman et al., 2001; Ferro and Mirabile, 2009; Di Stefano et al., 2010). The modification of the silt fraction's limit is suggested by Konert and Vandenberghe (1997) in order to correct the underestimated clay fraction.
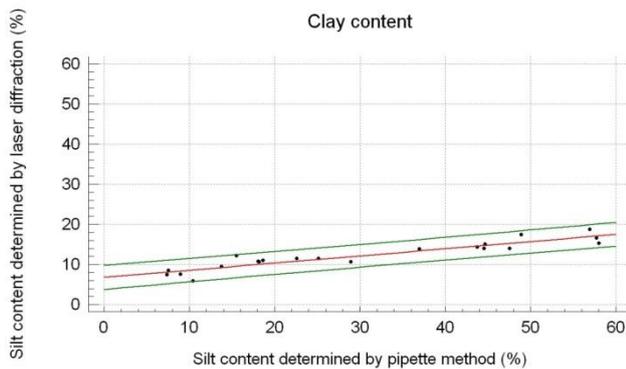

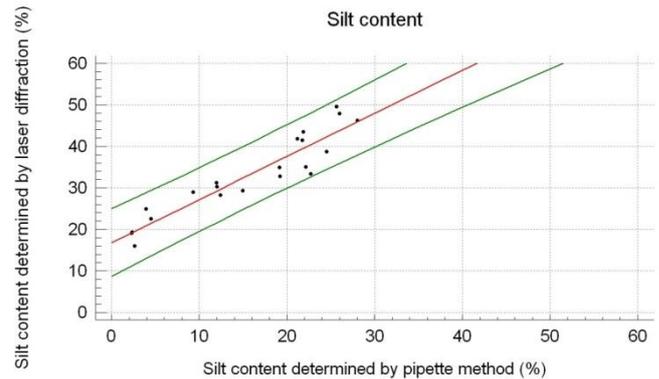*Fig. 6* Correlation between clay content
determined by the two methods

Fig. 7 Correlation between silt content
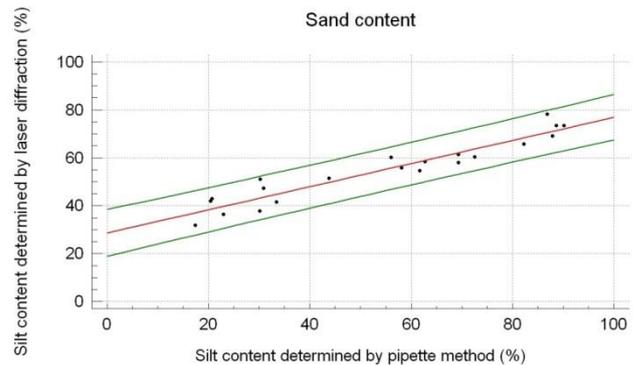determined by the two methods

Fig. 8 Correlation between sand content
determined by the two methods

CONCLUSION

In our research we examined the interchangeability of two different grain size measurement methods in case of unsorted fluvial samples. During the investigation we compared the cumulative and distribution

Table 2 Correlation between results received for different grain size classes

Size Class (µm)	Correlation factor	Linear regression
<2	0.929	Y=0.180·X+6.,667
2-5	0.921	Y=1.014·X+8.992
5-10	0.899	Y=0.974·X+8.336
10-20	0.676	Y=0.566·X+15.124
20-50	0.567	Y=0.447·X+16.871
>50	0.951	Y=0.483·X-3.551

curves produced by the two methods, studied the effect of clay, silt and sand abundance on distribution difference, and determined the correlation between results received for the main fractions (clay, silt, sand).

In case of high clay content, the LDM seems to underestimate the clay content in favor of the silt fraction. In the meantime the PM seems to overestimate clay and therefore the proportion of the silt fraction is significantly lower. The PM systematically overestimates the sand fraction compared to the LDM results. The LDM underestimates the sand fraction for the benefit of silt fraction because of the rough surface of the sand particles.

The interchangeability of the two methods in terms of the silt fraction is doubtful if distribution curves are considered. However, if the bulk values are calculated for the main fractions, differences are not significant. The lowest correlation factors were found in case of coarse silt (10-20 μm) and fine sand (20-50 μm). Therefore, the comparative analysis of particles with a 10-50 μm diameter is problematic.

Acknowledgement

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