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An investigation in laser diffraction soil particle size distribution analysis to obtain compatible results with sieve and pipette method

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ABSTRACT

Recent studies have shown that soil particle size analyses using laser diffraction method (LDM) can give compatible results compared with traditional sedimentation based methods, if the clay-silt particle size cutoff is transformed. Additionally, procedures including separation of the sand fraction by wet sieving and running a well dispersed sample of only fractions smaller than sand during laser diffraction measurement, have given promising results. The main purpose of the present study was to test a combination of these approaches for determining cutoff transformed LDM values on 44 soil samples from agricultural sites spread over Sweden, including its compatibility with the sieve and pipette method (SPM). Furthermore, these results were compared with results of transformed LDM values based on pedotransfer functions between measured LDM and SPM. Also LDM related aspects concerning scattering parameters, repeatability and organic matter calculations were studied. To find the optimum clay-silt cutoff, Lin's concordance correlation coefficient (Lin's CCC) was calculated. The highest value (0.977) was found with the 3.409–3.905 μm bin (a refractive index of 1.52 and an absorption coefficient of 0.1 was used). The pedotransfer-transformed LDM approach showed equally high Lin's CCC as the cutoff-transformed approach for the different soil particle fraction size classes. With the cutoff-transformed LDM approach, 36 out of 44 samples were assigned to the same texture class as SPM, and with the pedotransfer-transformed LDM, the corresponding number was similar (34 out of 44 samples). The results here are promising for application in routine soil analyses, but more specific transformed clay-silt cutoffs and pedotransfer functions for LDM versus SPM should ideally be established for different types of soils. For this, microscopy and image analysis methods to help understand and quantify the influence of particle shapes on obtained particle size distributions are useful.

1. Introduction

The accurate description of the soil particle size distribution (PSD) of the fine earth fraction (<2 mm) plays a pivotal role in many research applications since it influences numerous soil processes and properties including e.g. pore size distribution, water retention and hydraulic conductivity (Bieganowski et al., 2018). These soil properties are fundamental to soil vadose zone functions for ecosystems services such as regulation of water and nutrient cycling, habitat for microorganisms, carbon storage and agricultural production suitability (Drobnik et al., 2018).

Two principal approaches to determine soil PSD are currently used in research and commercial applications, (i) the sieve and sedimentation method (SSM) relying on a pipette or hydrometer, and more recently on a pressure transducer, and (ii) the laser diffraction method (LDM). The SSM is an internationally commonly used method and have an internationally agreed standard for the pipette and hydrometer applications in the International Organization for Standardization (ISO 11277, 2009, 2020). It should be noted, that the standard leaves room for divergences between laboratories. For an alternative method of soil PSD analysis to replace them, it should preferably give comparable results (Yang et al., 2019). The development of LDM for particle size measurements began in the 1970's (Ma et al., 2000). Its applicability for soil PSD has been extensively studied (Vandecasteele and De Vos, 2001; Arriaga et al., 2006; Pieri et al., 2006; Taubner et al., 2009; Di Stefano et al., 2010; Yang et al., 2015; Fisher et al., 2017; Šinkovičová et al., 2017; Faé et al., 2019). Several studies have reported large discrepancies between LDM and SSM, especially for the clay (LDM underestimated) and silt (LDM overestimated) fractions (Eshel et al., 2004; Taubner et al., 2009; Di Stefano et al., 2010). The sand fraction has been found to sometimes

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Table 1
Mean contents of clay, silt, sand (determined with sieve and pipette method) and organic matter (OM) (estimated from loss on ignition) for the 44 soil samples in this study.

Soil property	Mean	Median	Standard deviation	Minimum	Maximum
Clay (%)	25.9	21.2	20.4	0	65.9
Silt (%)	35.9	33.6	23.1	3.1	81.9
Sand (%)	38.2	33.4	33.1	2.1	94.8
OM (%)	3.9	3.1	4.7	0	23.9

agree well between the two methodologies and sometimes to be over- or underestimated (Eshel et al., 2004).

The discrepancies between LDM and SSM have been attributed to the theoretical assumption of particles being of spherical shape. For SSM, this assumption results in a discrepancy, as real particles of non–spherical shape reach lower settling speeds than spheres of equal volumes, resulting in higher effective clay content (e.g. Eshel et al., 2004). LDM describes platy particles as larger than that of a sphere of equal volume, as it projects a mean particle diameter which is an average from many axes of view, resulting in higher effective silt content (Taubner et al., 2009). These considerations have been supported by imaging techniques for SSM using scanning electron microscopy (Yang et al., 2019) and LDM using transmission electron microscopy (Pieri et al., 2006).

With LDM, photodetectors record the scattering pattern that occurs when particles are illuminated by a light source. By a mathematical algorithm, pre-set by the manufacturer of the LDM device, the recorded patterns are matched with the theoretical patterns (assuming spherical particles) so that a volumetric PSD with the best fit to the measured scattering pattern is achieved. The theoretical scattering pattern is given by the Mie-theory or the Fraunhofer approximation (ISO 13320, 2020). The Mie-theory requires the two scattering parameters refractive index (RI) and absorption coefficient (AC) to be known or estimated. Deciding which RI and AC to use is problematic for soil, since it consists of several minerals which all have different RI ranges, and information on AC is generally limited. For example, the soil mineral hematite, common in tropical soils, has a RI range of 2.9–3.2 whereas quartz and many other common minerals have ranges somewhere between 1.4 and 1.6 (Ozer et al., 2010). For these reasons, some studies have favored the use of the Fraunhofer approximation (Taubner et al., 2009; Šinkovičová et al., 2017). However, in a strict sense, the Fraunhofer approximation is only valid for particles greater than five to six times the wavelength of the light source. Consequently, particles smaller than ca 3–5 μm should be analyzed with the Mie theory (Keck and Müller, 2008). Ozer et al. (2010) proposed that X-ray diffraction can be used to determine which minerals the soil sample consists of, in order to select a representative RI. However, they also acknowledged that this is not always possible. In these cases, Ozer et al. (2010) recommended a RI value of 1.55, which corresponds to the common mineral quartz. For AC, which can range between 0 and 1 for optically transparent to fully opaque materials, Ozer et al. (2010) recommended to apply the value 0.1, the same as proposed by Bieganowski et al. (2018), the latter on the other hand proposed an RI value of 1.52. It should also be mentioned, that different LDM devices seem to respond differently to changes in scattering parameters, some being more sensitive than others (Varga et al., 2019).

A main advantage of LDM in comparison with SSM is that it gives a more detailed PSD with a higher number of size classes (Varga et al., 2019). The variation within the clay, silt and sand fractions can thus be more distinguished (Faé et al., 2019). It also provides with PSD classes below the clay-silt boundary, thereby making studies of finer clay fractions possible (e.g. Dur et al., 2004). LDM also has the advantage of being less labor demanding in analyses and requiring a smaller sample mass to be used than SSM (Arriaga et al., 2006). Thus, if the discrepancies in relation to SSM can be overcome, LDM is a promising alternative for soil PSD estimation. Faé et al. (2019) argued that if the PSD results obtained with LDM were consistent enough, making them compatible to those of the SSM is simply an empirical problem with different solutions. They suggested that one solution is to wet-sieve the sand fraction and measure it by weighing and, furthermore, to modify the clay-silt cutoff from 2 μm to ca 6 μm. In this way, results from LDM measurements differed only about as much from the pipette method (RMSE values of 5%, 6% and 3% for clay, silt and sand) as the hydrometer method did (RMSE values of 4%, 7% and 5% for clay, silt and sand).

The main purpose of the present study was to evaluate soil PSD using LDM by wet sieving the sand fraction and by performing different modifications to the LDM measurements to obtain results compatible with the reference sieve and pipette method (SPM). For this, 44 soil samples from agricultural sites distributed across Sweden were investigated. The specific modifications tested were: (i) pedotransfer functions, (ii) changing the clay-silt cutoff, (iii) using two sets of recently recommended scattering parameters, (iv) applying ultrasonic dispersion and (v) omitting organic matter content in the LDM calculations.

2. Material and methods

2.1. Soil samples

The National Soil and Crop Inventory (Miljödata-MVM, 2020) has sampling points located in arable fields across Sweden. A subset of 44 soil samples (17 topsoil, 27 subsoil) was selected, which had been analysed for PSD in 2019 with the sieve and pipette method (SPM). The PSD fraction limits were principally based on the Atterberg classical fractionation and its sub-fractions, i.e. 2, 6, 20, 63, 200, 600 and 2000 μm. The subset was selected to represent a range of texture classes with a broad span of clay, silt and sand as well as different contents of organic matter (OM) (Table 1 and Fig. 5). The samples originated from 44 sites spread between longitudes 12°12' E in the west (of Sweden), 22°00' E in the east, and latitudes 55°39' N in the south and 65°55' N in the north, representing the continental, nemoral and boreal south agro-climate zones in Europe (EEA, 2019). The soil parent materials are of glacial and postglacial origin.

2.2. Soil pre-sample preparation and analysis for the sieve and pipette method (SPM)

The procedure included air-drying, grinding and sieving the soil samples to < 2 mm (fine earth), before 20 g of this fine earth was taken for analysis. Sample organic matter was removed with 45 ml of deionized water and 10 ml of hydrogen peroxide (35%). The organic matter (OM) content was estimated from loss on ignition (550 °C for 4 h) on separate samples, and corrected by a factor related to clay content to account for crystal water content.

Dispersion was done overnight after OM had been removed, using an end-over-end rotator with 25 ml dispersant (Na_2CO_3 7 g l^{-1} + $(NaPO_3)_n$ 33 g l^{-1}) and diluted with deionized water to reach 1 l. The particles of 200–2000 μm fraction were separated by wet-sieving and stored. After stirring the solution for sedimentation analysis (with particles <200 μm), samples of 10 ml were taken out with the pipette after 56 s at 20 cm depth (fraction <63 μm), 4 min 38 s (fraction <20 μm) and 51 min 29 s (fraction <6 μm) at 10 cm depth. The method followed the protocol for sedimentation according to Stoke's law, adapting to ISO 11277 (2020). After 5 h 48 min the final sample was taken out at a depth of 7.5 cm (fraction <2 μm). Wet sieving was then done with a 200 μm and a 63 μm sieve for the samples, including also the stored 200–2000 μm fraction. After drying and weighing all fractions, dry sieving was performed on the 200–2000 μm sand with a 600 μm sieve to determine the 200–600 and 600–2000 μm fractions. In the present study the sand sub-fraction contents were not utilized, only the total sand content (63–2000 μm).

Table 2
Linear regression relationship between sieve and pipette method (SPM) and laser diffraction method (LDM).

Class (μm)	Equation	R^2	P-value	Equation with intercept locked to zero	R^2	P-value
Clay[a] < 2	LDM= 0.714SPM-2.163	0.917	< 0.001	LDM= 0.662SPM	0.897	< 0.001
Fine silt[a] 2–6	LDM= 1.222SPM+ 2.721	0.874	< 0.001	LDM= 1.400SPM	0.882	< 0.001
Medium silt[a] 6–20	LDM= 1.133SPM+ 3.180	0.934	< 0.001	LDM= 1.280SPM	0.93	< 0.001
Coarse silt[a] 20–63	LDM= 0.833SPM+ 1.786	0.888	< 0.001	LDM= 0.919SPM	0.894	< 0.001
Silt[a] 2–63	LDM= 1.083SPM+ 6.758	0.939	< 0.001	LDM= 1.217SPM	0.936	< 0.001
Sand[b] 63–2000	LDM= 0.976SPM+ 0.746	0.999	< 0.001	LDM= 0.987SPM	0.998	< 0.001

[a] pipette (SPM) and laser diffraction (LDM) analyses (LDM fraction limits based on nearest bin).
[b] sieving for both SPM and LDM analyses.

2.3. Soil sample pre-treatment and analysis for the laser diffraction method (LDM)

The soil samples were taken from the same fine earth fractions as prepared for the SPM analyses (which had thus already been air-dried, ground and sieved to <2 mm (fine earth)). Three subsamples (replicates) of 5 g were taken for each sample. Between each withdrawal, the bag containing the original fine earth was shaken and flipped to minimize the segregation of particles. The three subsamples were placed in separate 600 ml glass beakers.

To remove the organic matter, the same procedure as for the SPM samples was followed (to reduce influence of pre-treatment), however with adjusted volumes. Thus, 20 ml of deionized water was added to the subsample in each of the beakers, which were then placed on a water bath and 5 ml hydrogen peroxide (35%) was added to each of them. Each beaker was covered with a glass dish to prevent evaporation and was left overnight. The following day, the water bath was brought to boiling for 6 h. The samples were occasionally stirred and the evaporated deionized water was replenished. After 6 h, the heat was switched off and the samples were allowed to cool down to allow the particles to settle. The clear supernatant was removed with a pipette before the subsamples were transferred to 100 ml test tubes. The tubes were then filled with 12.5 ml of dispersant (Na_2CO_3 7 g l^{-1} + $(NaPO_3)n$ 33 g l^{-1}) and deionized water to a final volume of 80 ml. The samples were shaken overnight on a reciprocating shaker, set to 210 strokes per minute. The dispersed subsamples were then wet-sieved through a 63 μm sieve and collected in 600 ml beakers. The retained sand fraction (63–2000 μm) was oven-dried for 24 h and then weighed on a balance to obtain the sand mass.

The laser diffraction of each of the subsamples (<63 μm) was performed on a Horiba Partica La-950 v2 (Horiba, Ltd.) which has a 650 nm red laser diode and a 405 nm blue LED, with a measurement range of 0.01 – 3000 μm. Each measurement run is saved in 93 logarithmic size increments (bins). Deionized water was used as dispersion medium, which was degassed by the internal ultrasonic probe. A blank measurement with deionized water was taken to create a baseline value. Then, whilst the sample was being stirred, an aliquot was withdrawn with a wide-mouth (2 mm) pipette and enough sample was added to reach a transmission of 80 ± 0.5% (red laser). The measuring sequence used a pump speed of setting 7 (1633 rpm), and agitator setting 5 (2000 rpm). Before the first replicate subsample of each sample was discarded, they were exposed three times to 30 s of ultrasonic dispersion (output power 24 W) (3 ×30 s), with a PSD measurement taken in between each application. This was done as a means to ensure that sufficient dispersion had been achieved. The results were calculated with a set of scattering parameters, i.e. RI= 1.52 and AC= 0.1, as proposed by Bieganowski et al. (2018) to be an international standard to improve comparability between laboratories. In the results section, also alternative settings of RI and AC are elaborated.

The upper limits of the bins that the Horiba saves the data into do not match perfectly with the classes used in the SPM analyses, so the closest limits were selected. Thus, in the LDM analysis the SPM fraction classes < 2, 2–6, 6–20 and 20–63 μm were represented by < 1.981, 1.981–5.867, 5.867–19.904 and 19.904–58.953 μm bins.

2.4. Data processing and statistical analysis

The volumetric clay and silt fractions were recalculated to mass units, as proposed by Faé et al. (2019):

$$f_{cl} = \frac{(M_T - M_{sa}) \times f_{cl-LD}}{M_T} \quad (1)$$

$$f_{si} = \frac{(M_T - M_{sa}) \times f_{si-LD}}{M_T} \quad (2)$$

where f_{cl} and f_{si} are the clay and silt mass fractions, M_T total mass, M_{sa} the sand mass, $f_{cl\text{-}LD}$ the percent cumulative volume of particles less than clay-silt cutoff and $f_{si\text{-}LD}$ is the cumulative volume larger than the clay-silt cutoff (i.e. up to silt-sand cutoff). The assumptions for this approach are equal particle density and spherical particles, i.e. the same as for the sedimentation methods (Faé et al., 2019). Since organic matter had been removed, the sand mass was weighed without organic matter. For this reason we used the OM values from SPM measurements (recalculated from loss on ignition values) to get pre-estimated mass of OM that was subtracted from the total mass, before calculating the sand fraction. The three replicate subsamples of each soil sample were averaged before being used in the subsequent calculations and results.

To test the compatibility between the LDM and SPM, including finding possible pedotransfer relationships between them, the data was transformed by aid of linear regression (here called pedotransfer-transformed LDM). Linear regression with intercept set to zero was tested to avoid negative values in the subsequent calculations. All PSD fractions were proportionally adjusted to make their sum 100 for each sample (normalization).

Secondly, to identify the cutoff between clay and silt and between the silt subdivisions in the LDM data that result in the best agreement with SPM, Lin's concordance correlation coefficient (CCC) was used (Lin, 1989, 2000). This measure is a modification of the Pearson correlation coefficient that includes a correction factor in order to assess not only how close the data lie to the best fit, but also how far the fitted line is from the 1:1 line. The measurement can therefore be used to compare two methods when there is a standard, such as SPM in this case (Fisher et al., 2017; Makó et al., 2017). The optimized cut-off limits were used in the subsequent calculations (here called cutoff-transformed LDM). Furthermore, Lin's CCC was also used to evaluate the performance of the pedotransfer-transformed LDM values.

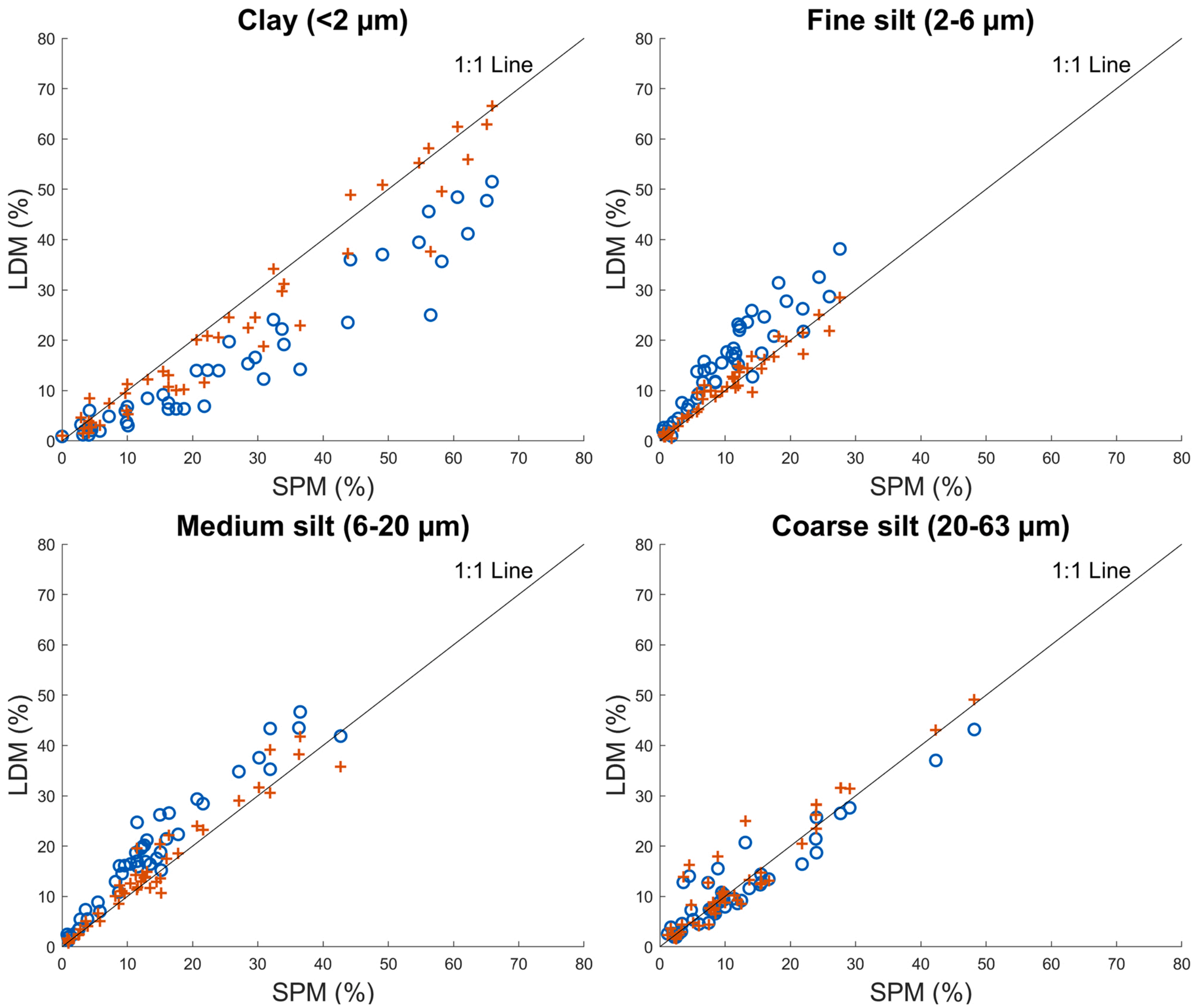

Fig. 1. Particle fractions for laser diffraction method (LDM) and sieve and pipette method (SPM) before (blue circles) and after (red crosses) LDM transformation by linear equation (using equations with intercept locked to zero in Table 2) and normalization.

To investigate the variance between the three replicate subsamples measured by LDM (i.e. the clay and silt fractions), the coefficient of variation (CV) was determined for the cumulative value at 10% (D_{10}), 50% (D_{50}) and 90% (D_{90}) of the distribution (ISO 13320, 2009).

Texture classification was done for all 44 samples for both transformed LDM and SPM, to investigate the agreement in textural classification. The USDA system was adopted, which delimits sand and silt at 50 μm. So for this texture classification (thus not for the other results in the present study), the silt and sand contents for both methods were adjusted by aid of loglinear interpolation (Nemes et al., 1999) in R using the Soil Texture Wizard (Moeys, 2018), the latter also used for automatic classification.

3. Results

3.1. Compatibility between LDM and SPM

The relationships between LDM and SPM were equally significant for the equations with intercept not locked (to the left in Table 2) or locked to zero (to the right in Table 2). The clay content was significantly underestimated by LDM in comparison with SPM, with lower values in 41 out of the 44 samples, and with a mean of 34% lower values as based on the slope of the least-square line with zero intercept (Table 2). Consequently, the silt content was overestimated by LDM for 42 out of 44 samples (mean 22% higher) with substantial inter-class variations. Thus, fine and medium silt was overestimated (mean 40% and 28% higher, respectively), and coarse silt was underestimated (mean 8% lower). The sand contents, determined by wet-sieving for both LDM and SPM, were almost identical (mean 1% lower for LDM). The relationships with zero intercept were used to transform the LDM values (Fig. 1). It is seen that the pedotransfer-transformed values (red crosses) have become fairly well distributed around the 1:1 relationship for clay and fine and medium silt. For coarse silt, already the untransformed LDM values (blue circles) were fairly well oriented around the 1:1 line.

Fig. 2 shows Lin's CCC changes when different cutoffs are used for LDM for the different SPM particle size cutoffs. The LDM peak for clay is notably higher than the SPM 2 μm cutoff, i.e. at 3.9 μm, and for fine silt higher than SPM 6 μm, i.e. at 7.7 μm. For medium and coarse silt the LDM peaks agree well with the SPM cutoffs at 20 and 63 μm, respectively. Fig. 3 shows the compatibility between individual fractions, when the optimized cutoffs are used. As seen, the values are well oriented around the 1:1 line, to be compared with the original LDM values (blue circles in Fig. 1) that to a large degree deviate from 1:1 line.

Lin's CCC calculations were also utilized to compare untransformed original results (blue dots in Fig. 1) with pedotransfer-transformed (red crosses in Fig. 1) and cutoff-transformed (Fig. 3) results. Both

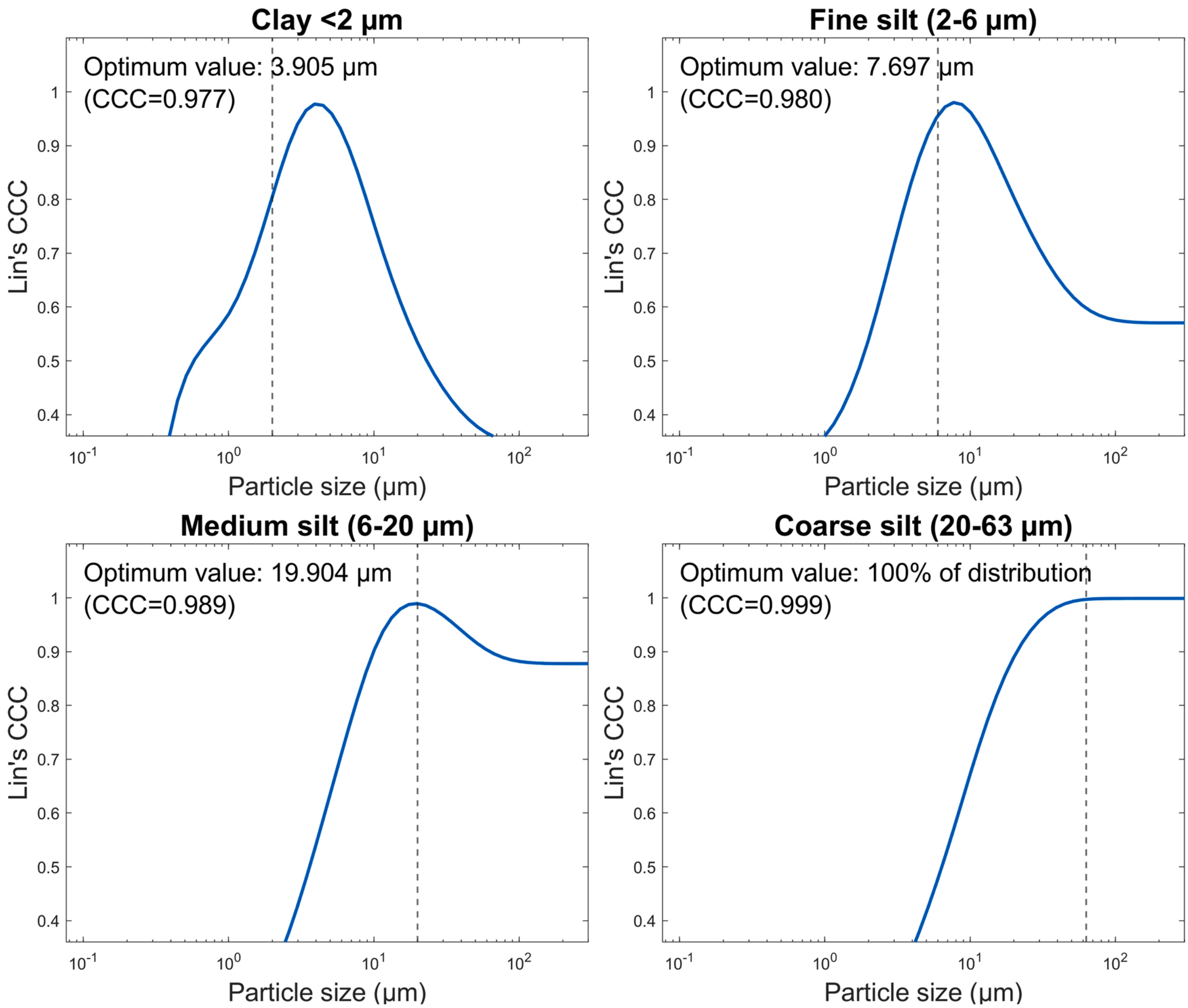

Fig. 2. Lin's CCC values obtained with a range of possible laser diffraction method (LDM) cutoffs for the upper limit of clay, fine silt, medium silt and coarse silt. The peaks indicate the cutoff that best matches the standard cutoffs used with sieve and pipette method (SPM) (the latter indicated by the dashed line).

pedotransfer and cutoff transformations improved Lin's CCC satisfactorily and to similar values (from 0.802 to 0.962 and 0.977 for clay, and from 0.891 to 0.977 and 0.983 for silt), with slightly higher Lin's CCC for transformed cutoffs (Table 3). For the subsequent analyses of the results below, unless otherwise mentioned, the LDM series with transformed cutoffs are considered.

The cutoff-transformed LDM median deviation from SPM for clay, silt and sand were, respectively, 1.83, 2.30 and 0.92 percentage points, and the maximum 14.81, 12.08 and 4.27 percentage points (Fig. 4). The soil texture classification resulted in that 36 out of 44 samples (82%) were assigned the same texture class for cutoff-transformed LDM and SPM (Fig. 5). None of the mismatches stretched further than to a neighboring texture class. In the case of pedotransfer-transformed LDM, 34 out of 44 samples (77%) were assigned to the same texture class as SPM. Thus, like for Lińs CCC analysis (Table 3), pedotransfer and cutoff-transformed values were almost equally good to reproduce SPM texture classes.

3.2. Influence of scattering parameters

The LDM device (Horiba) software allows recalculating the results for any given set of scattering parameters and we tested, in addition to those of Bieganowski et al. (2018) in the results above, the ones proposed by Faé et al. (2019): RI of 1.543 and AC of 0.01. Changing to these scattering parameters affected the correlation between the untransformed values (clay-silt cutoff 1.981 μm) of LDM and SPM in such a way that the R^2 decreased from 0.92 to 0.85 for clay and from 0.94 to 0.91 for silt (Appendix A, Table A1). Moreover, the optimum clay-silt cutoff increased from 3.905 to 5.867 μm, which leads to a Lin's CCC of 0.974 for clay and 0.980 for silt (to be compared with 0.977 for clay and 0.983 for silt for the original scattering parameters with clay-silt cutoff 3.905).

3.3. Repeatability of particle size measurements

The mean coefficient of variation (CV) for the cumulative values (three replicates) measured by laser diffraction at D_{10}, D_{50} and D_{90} was 6.4%, 2.1% and 3.3%, respectively (Appendix A, Table A2). The value for D_{10} was largely influenced by three outliers and if removing these (no reasons for these outliers related to soil or site properties could be detected), the value decreased to 2.9%. There are no specific recommendations for soil, but ISO 13320 (2009) recommended for powders and other substances, that the CV of D_{10}, D_{50} and D_{90} should be below 5%, 3% and 5% respectively. However, it is also mentioned that for particles below 10 μm, the maximum values should be doubled so e.g. 10% is the limit instead of 5% for D_{10}. Thus, it seems that the

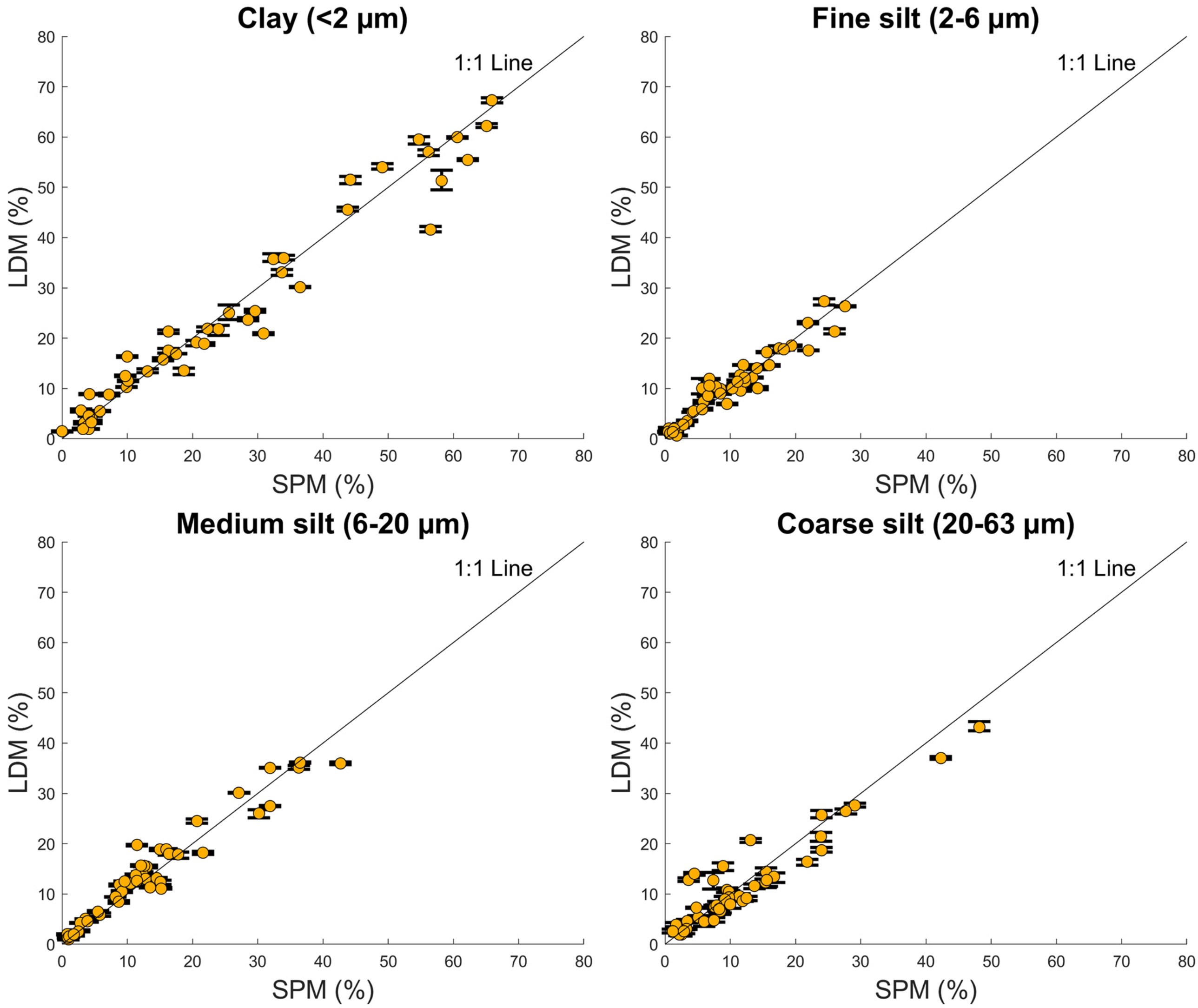

Fig. 3. The agreement for individual particle fraction between laser diffraction method (LDM) and sieve and pipette method (SPM). The LDM cutoffs were optimized to the following: < 3.905 µm is equivalent to < 2 µm, < 7.697 µm to < 6 µm, < 19.904 µm to < 20 µm and 100% of the distribution to < 63 µm. Bars show min/max of the three replicates.

Table 3
Lin's concordance correlation coefficient (Lin's CCC) for the original data, the data transformed using the linear equations in Table 2 with the intercept locked to zero (pedotransfer-transformed) and the data transformed by modifying the clay-silt cutoffs (cutoff-transformed).

Class (µm)	Original untransformed	Pedotransfer-transformed	Cutoff-transformed
Clay < 2	0.802	0.962	0.977
Fine silt 2–6	0.767	0.964	0.959
Medium silt 6–20	0.871	0.964	0.964
Coarse silt 20–63	0.935	0.933	0.935
Silt 2–63	0.891	0.977	0.983

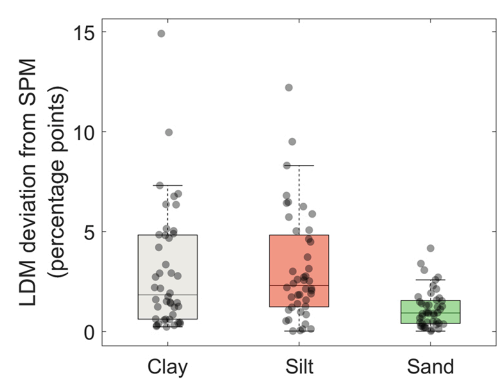

Fig. 4. Boxplot showing laser diffraction method (LDM) (with transformed cutoff) deviation (in percentage points in absolute terms) from sieve and pipette method (SPM) for clay, silt and sand. Each dot represents the average of the three replicates. The median values were 1.83, 2.30 and 0.92 percentage points for clay, silt and sand, respectively.

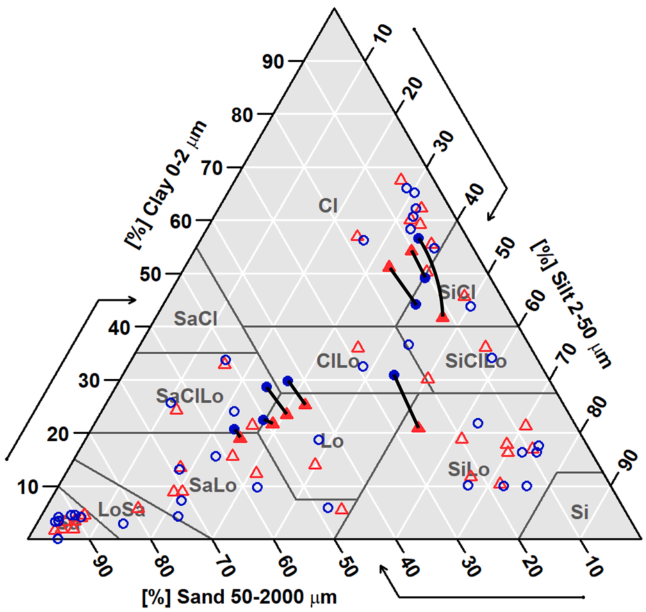

Fig. 5. The texture classes obtained with sieve and pipette method (SPM) (blue circles) and cutoff-transformed laser diffraction method (LDM) (red triangles). Filled circles or triangles and lines indicate the cases where SPM and LDM fall in different textural classes.

repeatability across the replicates was satisfactory. Moreover, the variability between the calculated values between replicates (according to Eqs. 1 and 2) of the clay and silt subclasses was mostly negligible (e.g. 0.8% on average and maximum 4% for clay) (Fig. 3), showing that a high CV of D_{10}, in the laser diffraction measurement influences the calculated fractions only marginally.

3.4. *Pre-treatment and sample preparation implications*

As identified in the literature, preparation of samples can affect the PSD using LDM. To investigate the sensitivity of the final PSD, we identified three issues related to the preparation of samples, (i) the dispersion of soil samples to be analysed, (ii) the wet sieving of sand and (iii) the influence of pre-estimated OM content in the calculations.

For the most part, using ultrasonic dispersion did not result in much change in the distribution pattern, indicating that the samples were well dispersed without this treatment (Fig. 6). However, for five samples, the use of ultrasonic dispersion noticeably increased the clay content and decreased silt content. Lin's CCC for clay content improved from 0.976 to 0.982 after the application of 3 × 30 s of ultrasonic dispersion (Table 4), indicating that in some cases the applied chemical dispersion was insufficient. For silt, Lin's CCC only improved slightly from 0.981 to 0.982. The use of 3 × 30 s of ultrasonic dispersion had no influence on the outcome of the texture classification.

The analyses of sand contents were very similar between LDM and SPM, i.e. performed by sieving, even though a much smaller soil mass had been used in the LDM study and the sieving was done by another operator and different sieve (smaller size for LDM). Due to the near 1:1 linear relationship between LDM and SPM (Table 2; intercept locked to zero), sand contents were not transformed for LDM as they were for the clay and silt fractions above (Figs. 1 and 3).

Removing the pre-estimated mass of OM from the total mass before calculating the sand fraction improved Lin's CCC for the sand fraction from 0.995 to 0.999 (Fig. 7). From Eqs. 1 and 2 it follows that this also influences the calculated clay and silt values. However, the effect of this correction was minor as seen in Fig. 7 demonstrated by a small shift in Lin's CCC from 0.978 to 0.977 for clay and 0.976–0.983 for silt. Three additional samples ended up in a different texture class compared to the pipette method when this step was omitted. These samples had sand contents over 50% and high OM contents (3.8–9.1%).

4. Discussion

4.1. *Compatibility with SPM*

Our results of final soil texture classification with agreement of 82%

Table 4
Summary statistics of the effect of using 3 × 30 s of ultrasonic dispersion on cutoff-transformed laser diffraction method (LDM) and its relationship with sieve and pipette method (SPM).

Ultrasonic dispersion	Fraction	R^2	RMSE	Equation	Lin's CCC
No	Clay	0.95	0.04	LDM= 0.943SPM+ 0.932	0.976
Yes	Clay	0.96	0.04	LDM= 0.972SPM+ 1.326	0.982
No	Silt	0.97	0.04	LDM= 0.938SPM+ 2.920	0.981
Yes	Silt	0.97	0.04	LDM= 0.920SPM+ 2.420	0.982

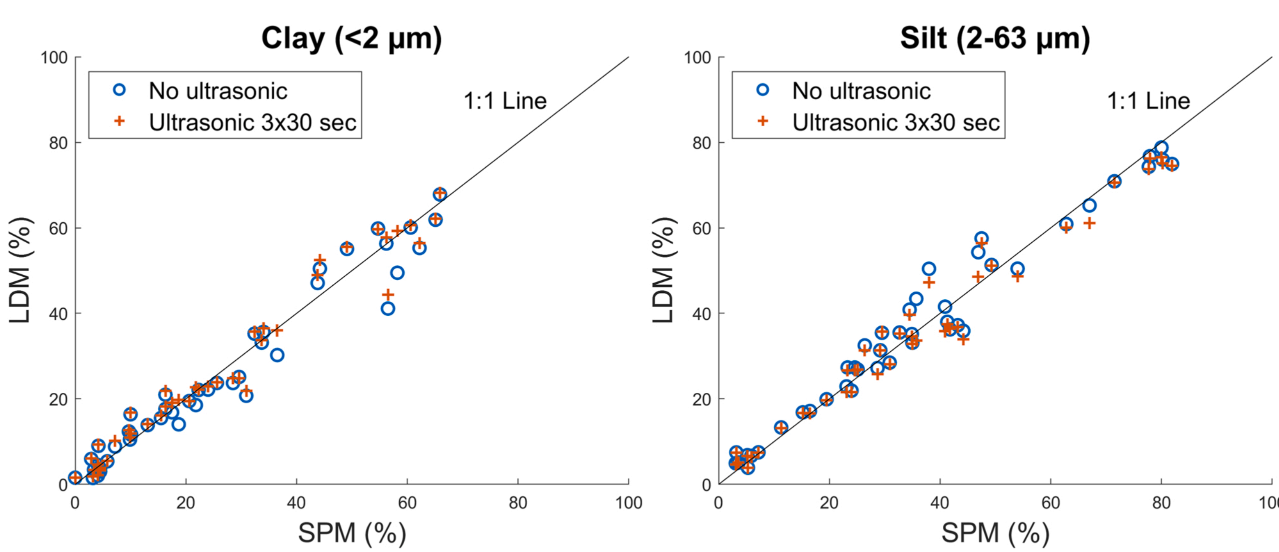

Fig. 6. The agreement between laser diffraction method (LDM) and sieve and pipette method (SPM) clay and silt content with and without 3 × 30 s of ultrasonic dispersion in LDM (for cutoff-transformed values). Summary statistics are shown in Table 4.

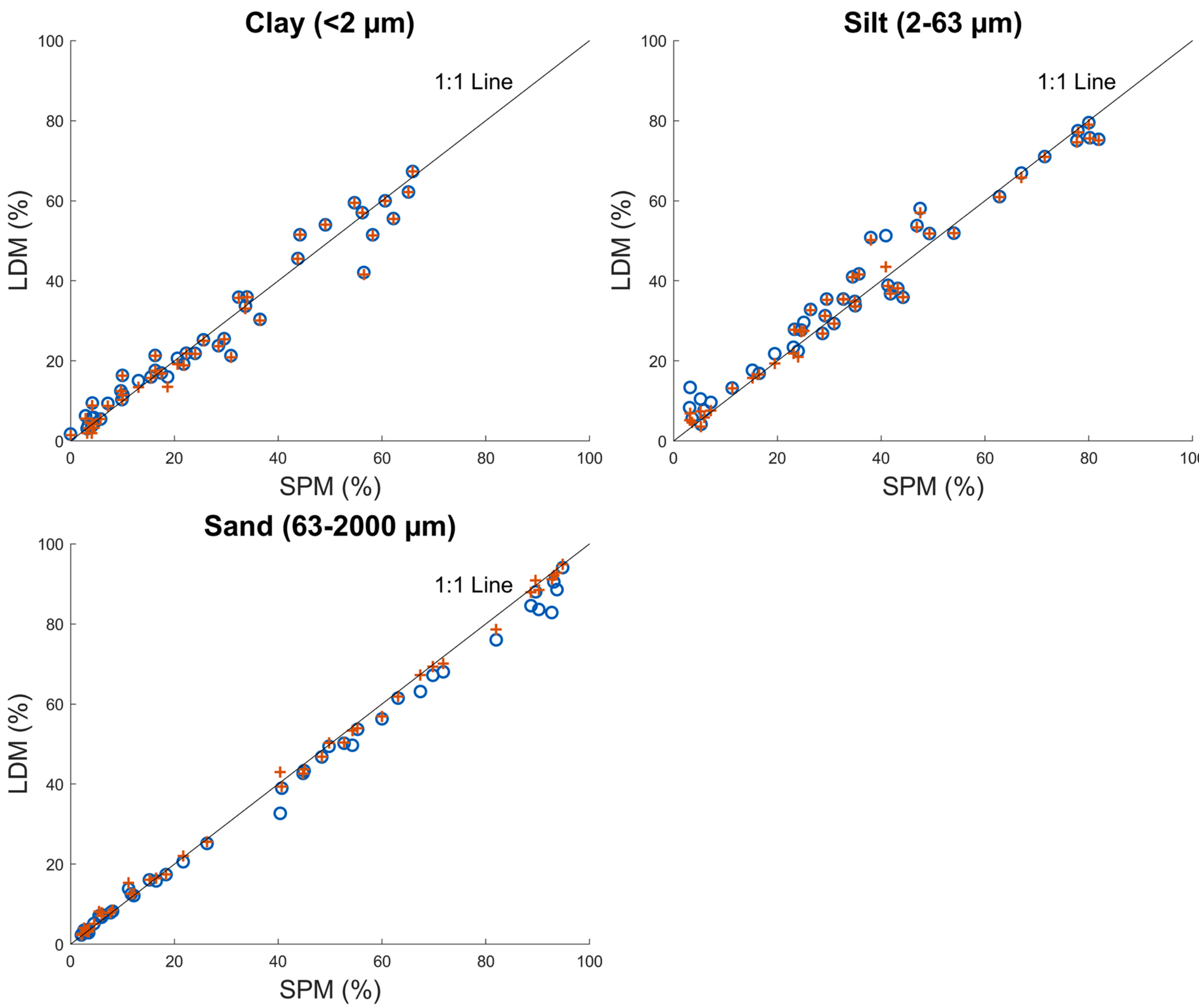

Fig. 7. The agreement between laser diffraction method (LDM) and sieve and pipette method (SPM) with and without removal of the pre-estimated mass of organic matter from the total mass before calculating the sand fraction. Blue circles represent without the correction and red crosses with.

between SPM and LDM (cutoff-transformed) analyses (Fig. 5), was higher than the best case reported by Makó et al. (2017) (62 ± 2%). Some of the mismatches in the texture classification could be explained by soils being close to one or more borders, and a relatively small difference in PSD was sufficient to shift the sample from one class to another. However, some samples differed considerably between the methods. Similar discrepancies were also found by Fisher et al. (2017) and Faé et al. (2019). A plausible explanation for the samples with high deviation could be that they contained more platy particles, thus deviating more from the assumptions of spherical particles. Bittelli et al. (2019) found greater discrepancies between the methods for samples that contained higher contents of platy clay particles, such as kaolinite. The clays in the studied areas are generally dominated by illite, but more detailed data on mineralogy was not available for the samples in this study.

Like for Lińs CCC analysis (Table 3), pedotransfer and cutoff-transformed LDM values were more or less equally good to reproduce SPM texture classes. One reason for the observed (very small) differences may be that deviations from the spherical particle shape may affect the pedotransfer-transformed LDM values slightly different than the cutoff-transformed. The fact that a normalization was required in order for the sums of the individual fractions to be 100% for the pedotransfer-transformed LDM, may explain some of the remaining divergences from perfect distribution around the 1:1 line after transformation (Fig. 1). The generality of the two different ways of harmonizing, i.e. pedotransfer and cutoff transformations, is something that needs to be further looked into, ideally with a separate training and validation set.

In the present study, 3 × 30 s of ultrasonic dispersion had little influence on most samples in this study, which may suggest that it is not needed if the protocol for sample preparation in line with ISO (ISO 11277, 2009, 2020) is followed. Nevertheless, it is a useful tool for discovering poorly dispersed samples and in some cases to disperse additional clay (Fig. 6). Bieganowski et al. (2018), in their literature review, found large variations regarding the usage of ultrasonic dispersion (duration and power) as well as the usage of chemical dispersant and removal of OM. Some studies that used ultrasonic dispersion in combination with a chemical dispersant found it to result in flocculation (e.g. Ryżak and Bieganowski, 2011). In contrast, Buurman et al. (1997) found that flocculation could be eliminated by ultrasonic dispersion alone or in combination with a dispersant. We could not see any indications of flocculation occurring as an effect of ultrasonic dispersion in the present study.

Removing the pre-estimated mass of OM from the total mass before calculating the sand fraction had a minor effect on the clay, silt and sand contents (Fig. 7). For routine PSD analyses with LDM, it may be argued that pre-estimation of OM content could possibly be skipped without influencing the results much if the samples are known to have a low OM or sand content. For new samples, a "pre-test" sample would otherwise have to be run in parallel to see how much mass that is lost due to the initial removal of organic matter.

4.2. Selection of the optimum clay-silt cutoff

The LDM clay-silt cutoff producing the best agreement with the pipette method was more dependent on the scattering parameters (RI and AC) than suggested by Faé et al. (2019), i.e. it moved more than to the adjacent bin in our study when RI and AC were changed from 1.52 and 0.1 to 1.543 and 0.01. This may be due to that a different device (Malvern Mastersizer 3000) was used in the Faé et al. (2019) study. Horiba devices, used in our study, have been shown to be more sensitive to the selection of scattering parameters than other devices (Varga et al., 2019). The optimum clay-silt cutoff of 5.867 µm obtained with the second set of scattering parameters (Table A1) was closer to the findings of Fisher et al. (2017), Makó et al. (2017) and Faé et al. (2019) who found the optimum to be 9, 6.6/5.8 (with/without OM) and 5.92 µm, respectively. The optimum clay-silt cutoff is also dependent on the soil type, making it difficult to find a universal cutoff even for a specific device (Gorączko and Topoliński, 2020). From this point of view, the results in the present study are fairly positive finding strong relationships between transformed LDM and SPM (e.g. Fig. 3 and Table 3) for a large range of PSD variations (Table 1).

The choice of the transmission likely also influences the selection of the best clay-silt cutoff. In this study, 80% was chosen in order to be within the recommendations given by the manufacturer. A small test on four samples in the present study (not included in the results section), suggested that changing the transmission to 90% could have led to that the best cutoff, instead of being found at 3.905 µm would be 1–2 bin size classes higher, i.e. a cutoff of 4.472 or 5.122 µm. Other factors that may have impacted the results is that we sieved to < 63 µm whereas, for example, Faé et al. (2019) sieved to < 53 µm. From Eq. (1) it can be seen that if the sand content is lower, the calculated fraction of clay in mass units will be larger, which should, in turn, influence the optimum clay-silt cutoff.

The proposal by Bieganowski et al. (2018) for a standardized set of scattering parameters is challenging. On one hand it would improve the comparability between studies, but on the other, the LDM scattering parameters leading to the best agreement with the SPM, seem to be dependent on the LDM device and possibly also soil type (Ozer et al., 2010). To some extent, this can be explained by the fact that the construction and mathematical algorithms differ between manufacturers, but unfortunately, the algorithms are generally not open access (Varga et al., 2019). So whilst a standardized protocol for LDM is appealing, each laboratory will likely have to adjust their settings and protocol according to the device at hand.

4.3. Issues related to particle size classification

While the optimum clay-silt cutoff depends on factors like those treated in the present study, it should be mentioned, as pointed out by Fisher et al. (2017), that the definition of clay is arbitrarily selected and that the separation at 2 µm is usually not used by sedimentologists and colloid chemists, who instead use 4–5 and 1 µm, respectively (Guggenheim and Martin, 1995). Moreover, it is worth considering whether determining clay content based on particle size alone is sufficient, since particles without clay characteristics may then be included in the clay fraction (Moreno-Maroto and Alonso-Azcárate, 2018).

The calibration of LDM to SPM is justified because the latter is a recommended standard in soil science (ISO 11277, 2020). However, analyzing microscopy images might be a more appropriate choice of method to validate LDM with, since it has been suggested to be an absolute method which directly measures the observed particles (Pieri et al., 2006). Even so, using microscopy coupled with image analysis is not free of errors, particularly because of the third dimension, which is not depicted by a two-dimensional image. Dur et al. (2004) and Pieri et al. (2006) used microscopy and image analysis for the clay fraction and chose to represent each particle as a platy disk with a thickness of one tenth of its diameter. Dur et al. (2004) found a good agreement with LDM because they represented the clay particles as platy disks for LDM as well. For the clay and silt PSD, a representation of both platy and spherical shapes is possible in LDM measurements. Such an approach should preferably be compared with microscopy and image analysis. Varga et al. (2018) presented an intensity-based thickness estimation of the particles with image analysis, which allowed to further reduce the deviation with LDM. In time, a recognized method for microscopy and image analysis will likely take shape. If so, adjusting LDM is once again an empirical problem, possibly with further studies to identify solutions that are less soil specific.

This study provides further evidence that with a few simple steps, LDM can be compatible with SPM as also shown in some previous studies (Fisher et al., 2017; Makó et al., 2017; Faé et al., 2019). Both the pedotransfer-transformed and cutoff-transformed approaches are promising. Twenty-four samples could easily be done per week with the protocol used in this study. If the initial removal of organic matter had been omitted, which is common in LDM studies (Bieganowski et al., 2018), at least double the amount of samples could be done in the same time. A test carried out in the development phase of this study suggested that if organic matter was not removed, a different shape of PSD was obtained, regardless if ultrasonic dispersion was used or not (see Appendix A, Fig. A1). Therefore it was decided to remove organic matter from all samples, and consequently keep the pre-treatments closer to the ones performed for SPM.

5. Conclusion

This study strengthens the evidence that the laser diffraction method (LDM) can be used to obtain results compatible with the reference sieve and pipette method (SPM) if the clay-silt cutoff is optimized and the sand fraction is wet-sieved. However, sporadic large discrepancies were identified for some samples. In line with other studies, it can be argued that the effect of the particle shape may be the main cause of the observed deviations. Pedotransfer-transformed LDM values for the same data gave good compatibility with SPM, almost to the same degree as the cutoff-transformed LDM values.

With the cutoff-transformed LDM in this study, 36 out of 44 soil samples were classified in the same textural class as SPM and the remaining samples in adjacent classes. With the pedotransfer-transformed LDM, this number was 34 out of 44. The use of ultrasonic dispersion did not affect the final texture classification, but it is recommended to be used as a tool to ensure adequate dispersion.

We conclude that LDM can be used as a compatible alternative to SPM, acknowledging discrepancies due to the fundamental differences in measuring techniques and that LDM results need to be harmonized by altering the clay-silt cutoff or using pedotransfer functions. Future studies would benefit from advancing robust microscopy and image analysis methods to help understand particle shape and the use of particle equivalents (platy or spherical) for LDM and SPM, which can then serve as a new basis for validating laser diffraction as well as sedimentation methodologies.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A

(See here: Appendix Fig. A1 and Tables A1, A2).

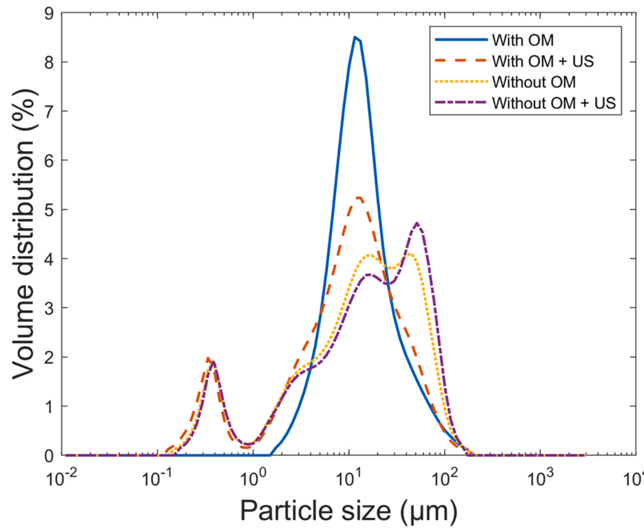

Fig. A1. Particle size distributions of four soil sub-samples in a pilot study, subjected to different pre-treatments (with and without removal of organic matter (OM) and ultrasonic dispersion (US)).

Table A1.
The agreement between laser diffraction method (LDM) and sieve and pipette method (SPM) under two different scattering parameters and with their transformed optimum clay-silt cutoff at 3.905 or 5.867 µm and untransformed cutoff at 1.981 µm.

RI/AC	Cutoff	fraction	R^2	RMSE	Lin's CCC	Equation
1.52/0.1[a]	3.905	Clay	0.96	0.04	0.977	LDM= 0.946SPM+0.909
1.52/0.1[a]	1.981	Clay	0.92	0.04	0.802	LDM= 0.714SPM-2.163
1.543/0.01[b]	5.867	Clay	0.95	0.04	0.974	LDM= 0.953SPM+0.367
1.543/0.01[b]	1.981	Clay	0.85	0.05	0.633	LDM= 0.603SPM-3.703
1.52/0.1[a]	3.905	Silt	0.97	0.04	0.983	LDM= 0.946SPM+2.630
1.52/0.1[a]	1.981	Silt	0.94	0.06	0.891	LDM= 1.083SPM+6.758
1.543/0.01[b]	5.867	Silt	0.96	0.04	0.980	LDM= 0.944SPM+3.049
1.543/0.01[b]	1.981	Silt	0.91	0.08	0.811	LDM= 1.141SPM+9.069

[a] As suggested by Bieganowski et al. (2018)
[b] As suggested by Faé et al. (2019)

Table A2.
Coefficient of variation (%) for the three replicates, for the cumulative value at 10% (D_{10}), 50% (D_{50}) and 90% (D_{90}) of the LDM cutoff-transformed distribution.

Sample ID	D_{10}	D_{50}	D_{90}
	%	%	%
1	4.9	8.4	8.6
2	1.3	1.8	2.5
3	0.3	0.7	1.6
4	0.6	0.3	0.8
5	4.5	2.2	5.0
6	1.3	1.9	2.5
7	0.6	0.3	1.4
8	1.2	1.5	2.5
9	1.0	1.0	3.9
10	1.4	3.6	1.4
11	0.5	0.5	2.0
12	0.6	0.8	5.2
13	0.2	2.1	9.3
14	3.5	4.1	2.9
15	0.7	1.4	2.6
16	87.9	2.8	8.7
17	7.4	4.9	8.7
18	1.7	1.7	5.6
19	10.8	1.9	2.9
20	1.0	1.0	3.2
21	3.1	3.6	2.9
22	1.0	2.0	2.2
23	0.2	0.4	1.1
24	8.8	2.0	4.1
25	2.0	2.2	6.5
26	7.5	2.7	3.5
27	1.5	1.2	0.3
28	3.2	5.3	5.2
29	3.0	1.8	0.8
30	0.6	0.8	1.9
31	7.2	2.0	2.3
32	24.7	4.4	2.3
33	4.8	0.5	5.0
34	0.5	1.6	2.0
35	0.5	1.7	3.2
36	2.9	3.9	7.0
37	0.6	2.2	0.9
38	1.1	0.7	0.6
39	8.0	5.1	1.6
40	2.5	2.1	3.4
41	4.7	0.5	1.6
42	57.7	3.8	3.6
43	1.3	0.5	2.5
44	8.2	1.7	1.9
Mean	6.4	2.1	3.3
Median	1.6	1.8	2.6

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