




Research Article

Grain size analyzers: results of an intercomparison study

Hugo Lepage¹  · Matthieu Masson² · Doriane Delanghe³ · Chloé Le Bescond²

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Abstract

In sciences involving soil and sediment, particle size distribution (PSD) has been one of the parameters given most attention over the past few decades. Formerly measured by sieving and sedimentation techniques, it is nowadays routinely characterized by the laser diffraction method (LDM). Many manufacturers develop particle size analyzers using LDM, but each device is characterized by specific parameters that can lead to different PSDs. At the Rhône Sediment Observatory, suspended particulate matter collected along the Rhône River is analyzed for PSD by four different LDM devices. Analyses were conducted on certified materials and sediment samples for each device. The tests highlighted the difficulty of accurately characterizing PSD, even in the case of certified materials. First, differences observed for a specific device were linked to the heterogeneity observed in the subsamples due to the presence of organic materials such as tree leaves. Second, the difference regarding the certified materials was linked to the laser diffraction method which leads in some cases to underestimating clay content and sand. Third, the main difference observed between the devices was linked to sonication. The results demonstrate that its power is rarely investigated and that it has a considerable impact when used. However, despite significant differences, the trend was similar for each device, with accurate characterizations of the main modal class in most cases. Thus, in the absence of exact knowledge of parameters such as sonication power and pump speed, it is recommended to compare only the trends of the results obtained from the different devices.

Keywords Particle size distribution · Laser diffraction · Ultrasound sonication · Suspended particulate matter · Sediment

1 Introduction

Much research has been conducted over recent decades to understand the dynamics and behavior of sediment and suspended particles in rivers. Understanding the processes that affect the transport of these particles is crucial for ecological, economic and societal purposes, as the world's rivers are becoming more anthropized [1, 2]. The investigation of their nature and origins has now become essential [3]. Among the different parameters used to characterize particles when studying their behavior (e.g., mineralogy, shape, color), particle size remains one of those most

investigated [4, 5] and it is often an issue in geoscience studies.

The characterization of particle size distribution (PSD) can be performed using several techniques such as sieving, by pipette based on Stokes diameters, and laser diffraction [6–8]. The choice of the optimum grain size analysis technique depends on the aim of the study and the type of sediment [8]. For loamy sediments, laser diffraction instruments produced the best results for the various criteria [8]. The main differences observed between the laser diffraction method (LDM) and the other methods are related to particle morphology and clay mineralogy

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✉ Hugo Lepage, hugo.lepage@irsn.fr | ¹Institut de radioprotection et de Sûreté Nucléaire (IRSN), PSE-ENV, SRTE/LRTA, BP 3, 13115 Saint-Paul-lez-Durance, France. ²Irstea, UR RiverLy, centre de Lyon-Villeurbanne, 5 rue de la Doua CS 20244, 69625 Villeurbanne, France. ³CEREGE UM34, CNRS, IRD, CDF, Aix Marseille Université, 13545 Aix-en-Provence, France.



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[5, 9]. Clay content can be underestimated in suspended sediments [10].

The LDM is widely used on sediment and suspended particles as it covers a wide range of grain sizes and requires short analysis time [5, 8, 11]. For example, particle size analyses are processed on cores recovered from marine, estuarine and coastal zones for paleoceanographic and sea-level reconstructions [12–15] and paleo-environmental questions associated with human history [16–18]. PSD is now commonly used to improve soil and sediment fingerprinting [19] and to investigate the concentration of trace metals in sediments. Apart from the geosciences, many domains such as forensic studies [20], pharmaceuticals and industry use this technology and require greater detail on the data acquired from different lasers.

Nowadays, many devices from different manufacturers are used to measure PSD by LDM, with different designs and software applications, leading to slightly different distributions [8, 21]. Bieganowski et al. [5] suggested that about 24 factors can influence the PSD results and lead to uncertainty or error. Differences arise as all these parameters vary between devices and laboratories, since resolving the issue of the homogeneous resuspension of sediment is a complex [22]. For example, it has been demonstrated that the use of ultrasonic power can destroy aluminum and silica particles due to the dissolution mechanism [23] and that the addition of oxidizing solutions to remove organic matter can also alter clay particles such as vermiculite [24]. However, intercomparisons of PSD measurements were performed more between the different PSD techniques described above than between different LDM devices. Nevertheless, differences were observed for the PSD and the proportion of clay measured by the devices of different manufacturers [8, 10]. Unfortunately, in the study conducted by Goossens (2008) on loamy sediments (sieved at 90 μm), the impact of sand particles and the use of ultrasound sonication (US) have not been addressed.

To investigate PSD, most of these studies used statistical parameters such as mean, median and standard deviation to summarize PSD or Kurtosis and the Skewness index to describe the variability of PSD [25, 26]. However, such parameters might not be sufficient to describe PSD in a non-log-normal or multimodal distribution [27, 28]. Suspended sediment PSDs are mainly multimodal, and the location of modes can be related to the transportation processes and the origins of the particles [28]. Also, modes can be used to investigate the dynamics of sediment deposition [29]. Several mathematical solutions were developed to summarize and describe raw PSD such as two or five parameters log-normal equations [30, 31], demodulation [32] and surface plots [28]. Unfortunately, these approaches can be complex for people unfamiliar with advanced mathematics. This may explain why scientific

publications and technical reports are still published with common parameters [33–35]. Also, depending on the study context, it is not necessary to rely on advanced solutions as common parameters may be sufficient for investigating PSD.

Since 2009, within the context of the Rhône Sediment Observatory (OSR) program, scientists have studied the transport of particles and their associated contaminants along the entire course of the Rhône River (from Lake Geneva to the Mediterranean Sea), the main source of sediments in the northern Mediterranean [36]. All these investigations are well-documented due to the presence of several monitoring stations located along the Rhône River and in its main tributaries [37]. Particle samples for chemical analysis are routinely collected. Analyses of PSD are systematically conducted not only to study the transport of particles but also to improve the interpretation of contaminant concentrations, and they can also be used for transport modeling. For example, the use of different sampling techniques for suspended particle sampling (particle trap and continuous flow centrifugation) leads to a bias in particle size [38]. In the case of large environmental monitoring networks such as the OSR program, PSD analyses are processed by an array of laboratory lasers.

It is therefore crucial to evaluate the potential biases induced by the use of different devices in the PSD results. The intercomparison was conducted by studying the differences observed on common parameters, such as the proportion of clay/silt/sand resulting from the analysis—with and without US—of reference standards and samples collected in the field. The objective was not to define which device is the most accurate but quantify the relative difference that might be observed and how the results can be interpreted and compared. Answering this question is crucial for the organizations responsible for river surveys, as we provide tools for comparisons between all the PSDs analyzed within the OSR.

2 Materials and method

2.1 Particle size analyzers

In the OSR program, grain size is assessed using the following devices: a Beckman Coulter LS 13,320 (Beckman Coulter, Fullerton, CA, USA), a Cilas 1190L (Cilas Company Ltd., Orléans, France), a Malvern Mastersizer 2000 (Malvern Instruments Ltd., Malvern, UK) and a Sequoia LISST-Portable|XR (Sequoia Scientific, Bellevue, WA, USA). These devices will be referred to as Coulter, Cilas, Malvern and Portable, respectively, for the rest of the article. As expected, the main characteristics of these devices differ (Table 1).

Table 1 Main parameters of the grain size analyzers used within the OSR

Brand	Cilas	Beckman Coulter	Malvern	Sequoia
<i>Main characteristics of the devices</i>				
Model	1190L	LS 13,320	Mastersizer 2000	LISST-Portable XR
Range of measured size (μm)	0.04–2500	0.04–2000	0.02–2000	0.34–500
Number of channels	100	132	100	33
Max. volume of the vessel (mL)	450	1000	1000	117
<i>Main parameters set for analysis</i>				
Ultrasonic use	30 s before and during analysis	During analysis	During analysis	30 s before analysis
Type of liquid and dispersant	Deionized water without dispersant	Tap water without dispersant	Tap water without dispersant	Deionized water without dispersant
Duration of a single measurement (s)	60	60	10	60
Optical model	Mie	Mie + Fraunhöfer ^a	Mie	Mie
Refractive index	1.55	1.5	1.57	1.54
Absorption coefficient	0.1	0.0	0.1	0.1
Ultrasound sonication power	50%	6 on scale of 8 levels	100%	30%
Stirrer speed	150 RPM	n.a.	1000 RPM	n.a.
Pump speed	120 RPM	80–90%	2500 RPM	20–25%
Obscuration (Obs) or optical transmission (OT)	Obs: 5–25%	Obs: 8–16%	Obs: 5–25%	OT: 0.75–0.95

n.a. not available

^aFraunhöfer used when particles are coarse (LS-13320 Manual)

2.2 Protocol of intercomparison

The intercomparison of the devices was conducted using certified reference materials (CRMs) and natural samples from the Rhône River and its tributaries:

- Measurements were performed on CRMs to investigate the accuracy of the different devices and highlight the differences between them. Three CRMs were selected in order to accurately represent the ranges of suspended particle size, mostly silt-size [38], observed in the Rhône River: BCR-066 (0.35–3.5 μm) and BCR-067 (2.4–32 μm) [39, 40]; SRM1003c (25–50 μm) [41].
- Artificial multimodal distributions were created to assess the capacity of the device to efficiently characterize the different modes. To do this, three mixtures of the 3 CRMs were prepared by weighing masses in order to reach the following proportions (BCR-066/BCR-067/SRM1003c in %): Mix_A: 25/50/25, Mix_B: 50/25/25 and Mix_C: 25/25/50.
- As CRMs are mostly composed with glass–quartz particles, they are not qualitatively representative of the suspended particulate matter (SPM) encountered in the Rhône River [42], especially regarding their aggregation properties. Therefore, an intercomparison was also conducted on the SPM samples collected in the

observatory. Four samples were collected using different methods at different stations to characterize the diversity observed in this observatory:

- Arc River (45°33'44.6"N 6°12'22.8"E): lag deposits that settled during a flood event were collected manually with a plastic spatula. This sediment was sieved in order to keep only a sandy fraction between 100 μm and 2 mm.
 - Azergues River (45°56'11.5"N 4°43'24.6"E): bottom sediment collected by a sediment dredge (Eckman),
 - Jons monitoring station in the middle Rhône River (45°48'42.3"N 5°05'09.2"E): SPM collected by particle trap [38] during baseflow in March 2017,
 - Barcarin station in the downstream Rhône River (43°25'12.5"N 4°44'50.7"E): SPM collected using a Niskin bottle 1 m above the surface during a flood event in November 2017. This was the only liquid sample comprising SPM in Rhône River water.
- The last step was conducted on a mixture of the samples collected in the Azergues and Arc Rivers to control the proportion of modes and the impact of coarse particles. The mixtures were prepared by weighing dry particles in the following proportions (Azergues/Arc in %):

Mix_1: 95/5, Mix_2: 90/10, Mix_3: 85/15, Mix_4: 80/20 and Mix_5: 70/30.

Each sample was homogenized and then split into 4 subsamples before being sent to the different laboratories for PSD analysis. For each sample, measurements were repeated on 2 or 3 distinct aliquots depending on the quantity available. Prior to measurement, the subsamples were dispersed in water according to Arvaniti et al. [11].

The Mie model was used when possible as the optimal model, as it is more useful in samples characterized by fine silt and clay content [43]. The Coulter device was the only device in this intercomparison that uses both models. Depending on the device, the refraction indexes were chosen to be similar and close to silica as the variation of this index can result in different PSDs [11]. The other parameters of the analysis method (Table 1), such as stirrer time and speed, pump speed adapted to the volume of the bowl, ultrasonic duration and measurement duration, were determined by the users to obtain the best possible results. (These parameters were optimized for each device with particulate samples collected in the OSR program.)

After a series of tests on the certified standards (data not presented), measurements were performed by conforming to the best obscuration range given by the manufacturer (Table 1). The results of the tests show differences occurring outside those ranges.

Each subsample was analyzed at least three times without US and then three times with US to investigate the repeatability of the measurements and the effect of sonication on the natural samples.

2.3 Comparison parameters

One of the main difficulties in conducting an intercomparison on PSD is the need to use synthetic parameters, as the raw distributions cannot be directly compared due to channel inhomogeneity. In the following study, intercomparison will be conducted on several of the parameters used most frequently in environmental studies and especially in the OSR program:

- The median (d_{50}) that synthesizes the distribution by one factor. However, this parameter is not the most relevant in the case of multimodal distribution, as is mostly encountered in rivers [28, 31].
- The proportions of clay ($< 1.95 \mu\text{m}$), silt ($1.95\text{--}2.5 \mu\text{m}$) and sand ($62.5\text{--}2000 \mu\text{m}$) are commonly used for normalization, as contaminants are mostly fixed on clay and silt. Again, this does not give information on distribution shape, especially in the case of multimodality [28] and the range of the classes can vary depending on the scale used [26, 44]. This separation into 3 classes

will be referred to as “simple classes” for the rest of the text.

- The proportions of detailed classes follow the Doeglas scale [45]: clay ($< 1.95 \mu\text{m}$), very fine silt (V_F_Silt: $1.95\text{--}3.91 \mu\text{m}$), fine silt (F_Silt: $3.91\text{--}7.81 \mu\text{m}$), medium silt (M_Silt: $7.81\text{--}15.63 \mu\text{m}$), coarse silt (C_Silt: $15.63\text{--}31.25 \mu\text{m}$), very coarse silt (V_C_Silt: $31.25\text{--}62.5 \mu\text{m}$), very fine sand (V_F_Sand: $62.5\text{--}125 \mu\text{m}$), fine sand (F_Sand: $125\text{--}250 \mu\text{m}$), medium sand (M_Sand: $250\text{--}500 \mu\text{m}$), coarse sand (C_Sand: $500\text{--}1000 \mu\text{m}$), very coarse sand (V_C_Sand: $1000\text{--}2000 \mu\text{m}$). It allows a good and comparable description of the distribution, but the higher classes are less-well adapted for statistical analyses than previous parameters. This separation will be referred to as “detailed classes” for the rest of the text.

2.4 Specific case of the LISST-Portable|XR device

Due to its lower range of PSD than the other devices (Table 1), and as it cannot characterize the particles with a diameter lower than $0.34 \mu\text{m}$, this LDM device is sensitive to rising tails [46–48]. Rising tails can be linked to the refractive index for small particles [49] or shape effects [12], and most studies recommend removing the lower classes and keeping the larger size classes that are consistent [46]. Thus, as rising tails were observable for almost all the samples, the first 5 classes ($< 0.72 \mu\text{m}$) of all the samples measured by the Portable were removed.

However, for this study, both raw and corrected PSDs were used to investigate the impact of rising tails on the PSD. Corrected PSD will be referred to as “Portable_C” for the rest of the text. Only the results of Portable_C will be presented in the graph for the sake of clarity, and the results without correction are available in the Electronic Supplementary Material. Also, the presence of bubbles during the analysis led to the presence of modes in the higher classes. This was especially relevant for the CRMs that should not be characterized by such coarse particles. Corrections were performed on references (BCR-066 & BCR-067) and a mixture of references (Mix_C), by deleting the coarser classes.

3 Results

3.1 Accuracy of the devices

The first part entailed measuring the CRMs. For this step, the use of US had a negligible impact as CRMs are characterized by non-cohesive materials (quartz). Only the results with US will be presented in this chapter.

First, the results obtained by each device from the different aliquots of the CRMs (Fig. 1) were characterized by low standard deviations (SD) except for mixture C (Mix_C) measured by the Coulter (high SD on clay and coarse silt). For this device with this mixture, the mean d50 ranged from 2.4 ± 0.1 to $6.2 \pm 1.4 \mu\text{m}$ for the three aliquots measured and the mean proportion of clay ranged from 27 ± 2 to $42 \pm 1\%$ (Electronic Supplementary Material).

Second, the values measured by the devices were compared to the certified values (Table 2, Fig. 1). For the finest CRM (BCR-066), all the devices overestimated the d50 (Table 2) and underestimated the proportion of clay (Fig. 1). The absolute difference ranged from 13 to 34% for the clay proportion, which represented 15 to 40% of the relative difference. For BCR-067 and SRM1003c, the

measured d50s were similar to the certified parameters with a relative difference lower than 20% (Table 2). For the simple classes, the relative difference was also lower than 20% but each device measured particles in classes in which the CRMs should not be observed. Indeed, all the PSDs measured by the devices were more spread out with a low amount (< 10%) of particles observed in the finer and coarser classes than the expected classes of the CRMs. This difference was clearly observed on the detailed classes (Fig. 1), and the relative difference exceeded 20% for all the devices on the classes certified by the BCR-067 (from 25 to 175%). For the SRM1003c, the difference exceeded 10% only once with the proportion of very coarse silt measured by the Portable_C device

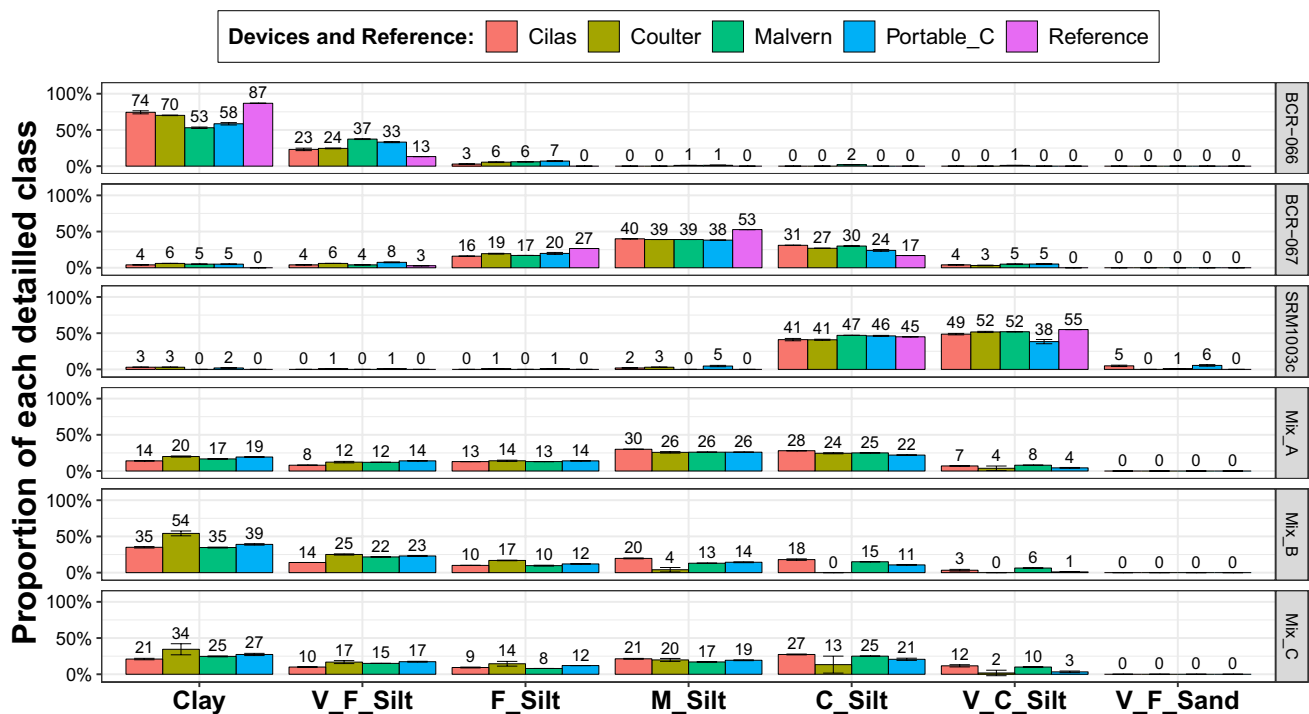


Fig. 1 Proportion of the detailed classes measured with US by the different devices (Cilas, Coulter, Malvern and Portable corrected) on the CRMs (BCR-066, BCR-067 and SRM1003c) and the different

mixtures (a, b and c). Certified proportions are also presented (Reference). Values over the bar represents the percentage of the proportion

Table 2 Mean D50 (μm) and standard deviation determined by the devices without US

	BCR66	BCR67	SRM1003c	Mix_A	Mix_B	Mix_C
Certified range (μm)	0.35–3.5	2.4–32	25–50	n.a.	n.a.	n.a.
Certified D50 (μm)	1.13	10.41	32.1 ± 1.0	n.a.	n.a.	n.a.
Cilas	1.2 ± 0.1	12.4 ± 0.1	32.6 ± 0.8	11.5 ± 0.1	4.2 ± 0.4	11.4 ± 0.5
Coulter	1.5 ± 0.1	11.3 ± 0.1	31.6 ± 0.1	8.9 ± 0.5	1.8 ± 0.1	4.6 ± 2.3
Malvern	1.9 ± 0.1	12.3 ± 0.1	31.9 ± 0.1	10.1 ± 0.1	3.0 ± 0.1	8.9 ± 0.1
Portable_C	1.9 ± 0.1	11.8 ± 0.4	32.6 ± 0.6	9.3 ± 0.1	2.8 ± 0.1	5.9 ± 0.7

n.a. not available

(38%) (Fig. 1). Thus, all the devices underestimated the proportion of particle in the modal class (Fig. 1).

The intercomparison of the devices was conducted on those results. The d50s measured on the CRMs by the four devices were similar, whereas significant differences were observed between the devices for the mixtures (Table 2). For instance, the d50 measured by the Cilas for mixture C ($11.4 \pm 0.5 \mu\text{m}$) was more than twice as high as the d50 measured by the Coulter ($4.6 \pm 2.3 \mu\text{m}$). Modal classes of the CRMs (unimodal) and the mixtures (bimodal) were accurately characterized by all the devices with the exception of mixture B measured by the Coulter. For this mixture, the PSD measured by the Coulter was unimodal and no particles coarser than medium silt was measured, whereas it represented more than 20% for the other device (Fig. 1). Thus, the proportion of clay measured by the Coulter was significantly higher than the other devices for this mixture (54 vs 30 to 33%,—Fig. 1). For the other samples, the proportions measured by the devices were similar (< 10% absolute difference) except for the finest CRM (BCR-066). The proportions of clay measured by the Cilas and the Coulter on this CRM (74% and 70%, respectively) were significantly higher than the two other devices (53% and 58%, Fig. 1).

3.2 Natural samples

The first natural sample, collected directly in the water flow with a Niskin bottle at Barcarin, was mostly silt sized with a non-negligible proportion of clay (Fig. 2, Table 3). The PSD measured by the different devices were similar, so as for the d50 (Table 4). A difference was observed only for the Coulter which measured a significant and higher proportion of clay (+ 11–13%), as observed previously for Mixtures B and C (Fig. 1). The d50 measured by the Coulter was strictly lower than the other devices (Table 4). Sonication was used only on the Cilas and had a negligible impact on this sample (Electronic Supplementary Material).

The sample collected by particle trap at Jons was coarser than the SPM from Barcarin (Fig. 2), with a significant proportion of sand (up to 27%, Table 3). The differences between the devices observed on the measures performed without US remained lower than 10% (Fig. 2) and the d50 s were similar (Table 4). As observed previously, the d50 and the clay content measured by the Coulter were, respectively, lower and higher than the results obtained by the two other devices. The use of US led to significant change in the PSD for the Portable_C (from 20 to 3% of very fine sand), while PSD remained similar for the Coulter and Cilas. Thus, the proportion of silt was

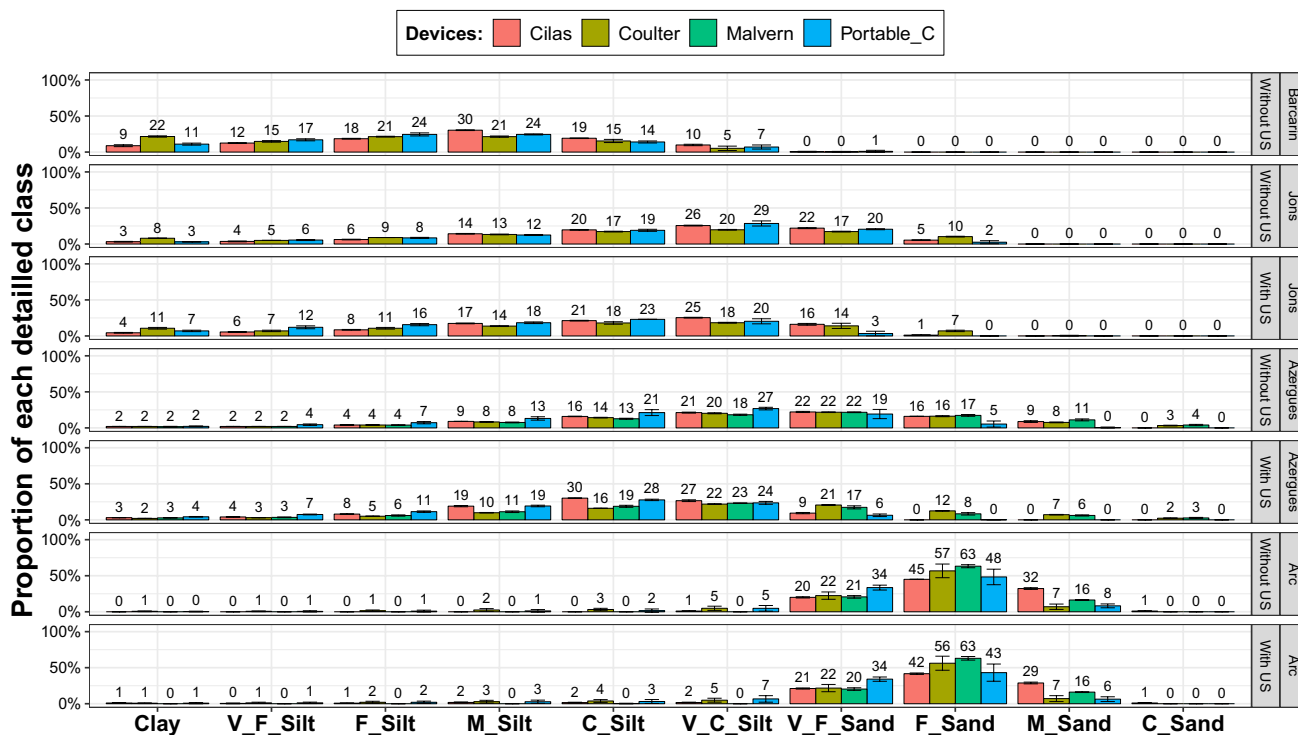


Fig. 2 Detailed classes measured by the different devices on the natural samples with and without US. Values over the bar represent the percentage of the proportion. No measures were conducted on the Malvern for Barcarin and Jons samples

Table 3 Mean proportion (%) and standard deviation of the simple classes measured by the four devices on the natural samples with and without US

Devices	Us	Barcarin			Jons			Azergues			Arc		
		Clay	Silt	Sand	Clay	Silt	Sand	Clay	Silt	Sand	Clay	Silt	Sand
Cilas	Without	8.8±1.3	90.3±0.8	0.5±0.5	3.2±0.4	69.2±0.4	27.4±0.9	2.0±0.1	51.3±0.7	47±0.9	0.7±0.1	1.0±0.1	98.3±0.5
Cilas	With	9.8±2.1	90.0±1.9	0.2±0.4	4.4±0.5	78.3±1.6	17.1±1.5	3.0±0.1	87.6±0.7	9.4±1	1.0±0.1	6.4±2.0	92.8±1.9
Coulter	Without	21.6±0.9	78.0±0.7	0.4±0.5	8.0±0.1	64.8±1.0	27.3±1.0	2.0±0.1	49.0±1.5	49.4±1.4	0.7±0.5	13.0±8.1	86.3±8.6
Coulter	With	n.a.	n.a.	n.a.	10.7±1.2	67.3±3.8	21.7±5.0	2.0±0.1	55.3±1.1	42.7±1.1	0.7±0.5	14.2±8.4	84.9±8.8
Malvern	Without	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	1.7±0.5	44.2±2.1	54±2.2	n.d.	n.d.	100.0±0.1
Malvern	With	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	2.8±0.4	62.3±3.9	34.9±4.3	n.d.	0.4±0.9	99.6±0.9
Portable_C	Without	11.0±1.4	87.5±0.7	1.5±2.1	3.0±0.1	74.0±2.8	23.0±2.8	2.1±0.6	72.6±9.9	25.1±10.7	0.3±0.1	9.6±1.0	90.0±1.0
Portable_C	With	n.a.	n.a.	n.a.	7.0±0.1	89.3±2.1	3.3±3.2	4.2±0.4	89.4±1.7	6.4±2.1	0.7±0.8	15.4±11.8	83.9±12.3

n.d. not detected, n.a. not analyzed

Table 4 Mean D50 (µm) and standard deviation measured by the four devices on the natural samples with and without US

Devices	US	Barcarin	Jons	Azergues	Arc
Cilas	Without	9.9±0.5	34.6±1.0	57.0±1.3	194.1±3.6
Cilas	With	9.2±0.7	25.0±1.3	23.1±1.1	179.3±5.3
Coulter	Without	6.2±0.4	28.2±0.7	61.1±2.8	146.6±20.0
Coulter	With	n.a.	21.9±3.7	49.7±1.3	145.9±20.5
Malvern	Without	n.a.	n.a.	71.6±5.8	171.9±1.6
Malvern	With	n.a.	n.a.	40.0±5.4	171.5±2.1
Portable_C	Without	7.9±1.0	35.4±0.2	37.9±9.1	143.9±20.1
Portable_C	With	n.a.	15.5±3.4	20.9±1.8	132.9±25.3

n.a. not analyzed

significantly higher (+20%) for the Portable_C with sonication than for the other devices (Table 3).

The bottom sediment collected in the Azergues River was characterized by a mix of silt and sand particles (Table 3), with the main mode observed between the coarse silt and the very fine sand (Fig. 2). In absence of US, the trend was similar for each device and only the Portable_C was characterized by a higher proportion in the silted classes and a lower proportion of sand. The proportion of silt measured by Portable_C (72.6±9.9%) was significantly higher than the other devices (from 44.2 to 51.3%) but characterized by a high standard deviation. The use of US on this sample led to a significant variation for all the devices and to an increase in proportion of silt particles (+6 to 37%) for all the devices (Table 3). However, this variation was greater for the Cilas (Fig. 2) (+37% of silt) than for the three other devices (+6–16%). The proportions of the classes after US were different and separated into two groups: Cilas and Portable_C with almost 90% silt, with coarse silt as the modal class, and Coulter and Malvern characterized by a large proportion of sand (35–43%) and very coarse silt as the modal class. Once again, the differences observed for the d50 were significant whether US was used or not (Table 4). For instance, d50s measured by the Malvern with and without US (respectively, 40.0±5.4 µm and 71.6±5.8 µm) were two times higher than on Portable_C (20.9±1.8 µm and 37.9±9.1 µm).

The last sample, the lag deposit collected in the Arc River, was characterized by fine sands (Fig. 2). For the four devices, the proportion of sand was higher than 80% and reached 100% for the Malvern (Table 3). The Coulter and Portable_C distributions were characterized by higher uncertainty intervals. These devices also measured a higher mean proportion of silt particles compared to the other two devices, resulting in a significantly lower d50 (Table 4). However, despite this difference, the trend was similar between the devices with the maximum proportion observed in the fine sand class (Fig. 2). The PSDs

measured by the Malvern were less spread than for the other device, while the proportion of sand classes measured by the Cilas was more spread with a lower proportion in the modal class (F_Sand) and a higher proportion in the coarser sand class (M_Sand). Finally, as this sample was mostly sandy, US had a negligible impact on the distribution, as observed for the d50 (Table 4).

3.3 Mixtures of natural samples

Measures were then performed on the mixtures of the Arc and Azergues samples. As expected, the increase of the proportion of the coarser sample (from mixture 1 to 5) led to a higher d50 (Fig. 3) and higher proportions in the coarser classes (Electronic Supplementary Material)

whether sonication was used or not. However, breaks were observed for the Coulter (Mix_1), the Malvern (Mix_4 and Mix_5) and the Portable_C (Mix_3 and Mix_5).

The differences observed between the devices were similar to the difference observed for the Azergues sample as it represented more than 70% of the mixture. Thus, the parameters were different both with and without US (Fig. 4). Significant differences were observable for d50 and with the proportion of silt and sand, whereas the proportion of clay remained lower than 5% for each device (Fig. 4). The position of the main mode was also different, both without (simple and detailed classes) and with US (detailed classes). Using US led to the strong variation for simple classes measured by the Cilas and the Portable_C (+35% silt), whereas it had no effect for the Coulter (Figs. 3,

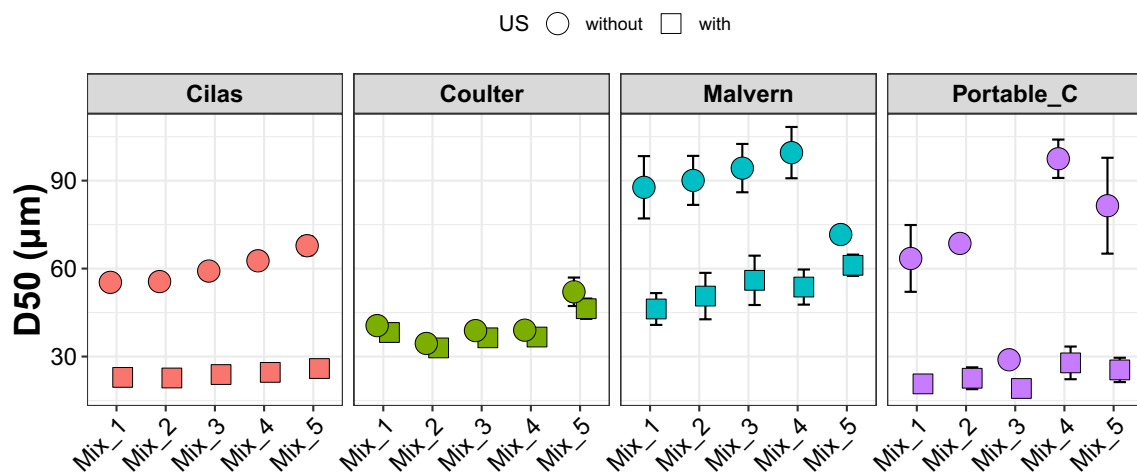


Fig. 3 d50 measured by the different devices on the mixtures of Arc and Azergues samples

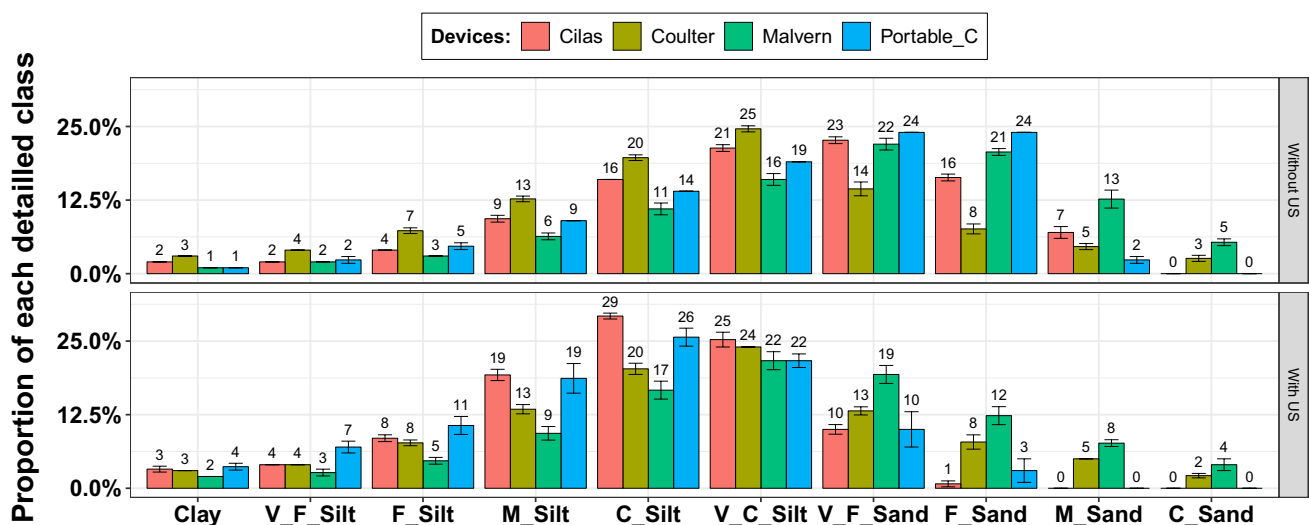


Fig. 4 Detailed classes measured by the different devices on mixture 2 with and without US. Values over the bar represent the percentage of the proportion

4). US also affected the Malvern device (+ 20% silt). The trends and the differences between the devices were similar irrespective of the proportion of sand in the samples.

4 Discussion

The measures performed by the four devices on the different samples (CRMs, sediments and associated mixtures) allowed determining their respective capacities in terms of measurement quality. The results demonstrated that several differences were observed despite the good and reproducible characterization of the main/modal classes and their distribution by the devices. These differences can be separated into three groups: intra-variation, variation with the certified materials and variation between the devices.

4.1 Intra-variation

The intra-variation is the difference observed for the aliquots of the same subsample measured by the same device. For the CRMs, this difference was observed only in mixture C measured by the Coulter (Fig. 1) and is linked to the operator subsampling operation. Subsampling appears to be highly operator-dependent and can infer variations on the results of the same sample that are even higher than the analytical differences observed [5]. The intra-variation of the natural samples was observed mostly for coarse classes (very coarse silt to fine sand) measured by the Coulter and the Portable_C devices (Fig. 2). This was observed both without and with US and was mainly related to the heterogeneity of the sample particles. Indeed, during the subsampling some samples contained coarse organic particles such as tree leaves that modified the proportions between modes. For the Portable_C, it was also related to the presence of bubbles as explained in the chapter 2.4. To improve the characterization of sand particles, the pump speed of this device was increased for coarse samples and triggered the formation of bubbles. It is therefore relevant to perform measures on multiple sub-samplings of a sample (at least three) to characterize a more precise mean PSD and to be careful with the presence of coarse organic materials and bubbles. Also, the sieving of coarse particles could be a solution for decreasing the pump speed.

4.2 Variation with the CRMs

The measures performed on the CRMs for all the devices presented differences with the certified values. First, they underestimated the proportion of clay in the finest reference (BCR-066) and the different associated mixtures

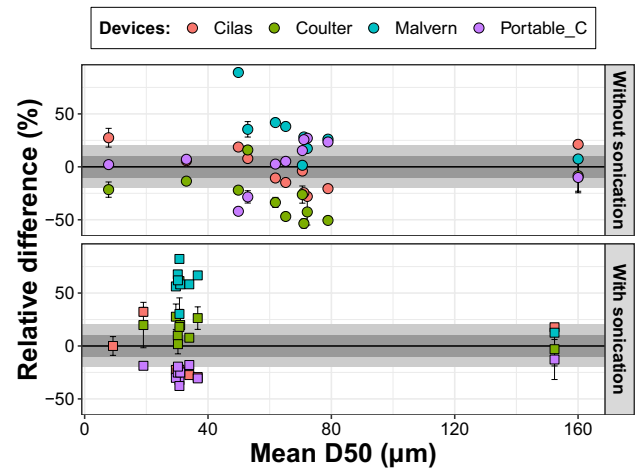
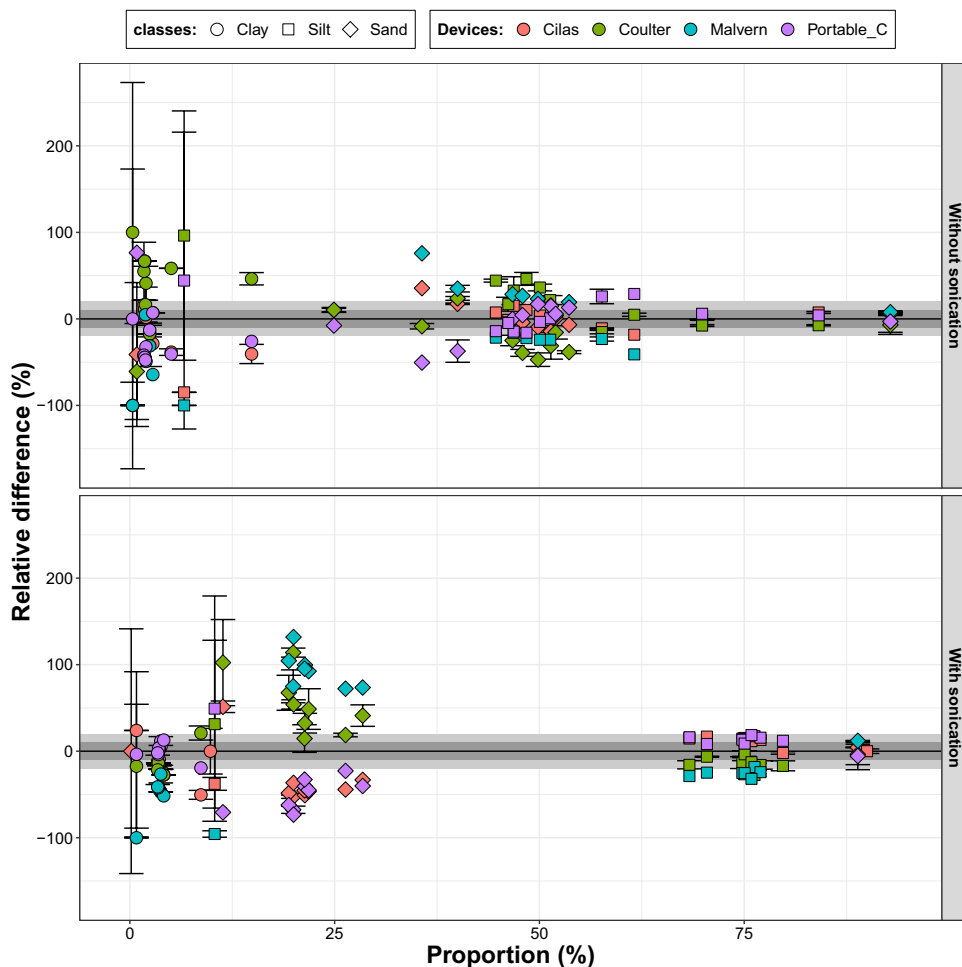


Fig. 5 Relative difference between the mean d50 measured by all the devices and the mean d50 measured by each device with and without US. Gray areas represent 10% and 20% difference

(Fig. 1). This underestimation was linked to the particle morphology and the clay mineralogy [9] affecting the PSD measured by LDM. Similar observations had already been performed by [10, 21]. Second, the PSDs measured by all the devices were more spread out than the associated references or mixtures. Also, particles were observed in classes where they should not have been present. For example, all the devices measured clay particles with the BCR-067 and SRM1003c (Fig. 1). However, the proportions in these classes were lower than 10%. This difference might be related to the presence of particles not washed during cleaning. Despite the measure of the background PSD with tap or dionized water, some particles might have been trapped in the system and resuspended during other measurements. This difference might also be linked to the different methods used to characterize the particle size for their certification. BCR-066 and BCR-067 were characterized by the Pipette method [39] which might have triggered differences with the LDM, as observed with the proportion of clay.

To summarize, all four devices exhibit the same trends in accuracy from small to coarser particles. However, deviations on the d50 remain roughly lower than 10% for the standard ranging from 4 to 35 μm . As explained previously, the devices do not accurately characterize clay, irrespective of the proportion observed, with the exception of the total absence of clay. Higher relative differences are observed when the proportion of clay is low (approx. 10%). For the silt, a small difference is observed in the CRMs characterized by almost only silt particles. Below 30% silt, the difference can be significant (from 20 up to 300%), especially for CRMs mainly characterized by clay and silt

Fig. 6 Relative differences between the mean proportion of each simple class measured by all the devices and the mean proportion measured by each device with and without US. Grey areas represent 10% and 20% difference. Abscise represent the mean proportion of the associated simple class for all devices



(BCR-066). Finally, the absence of particles in any simple class is accurately characterized by the devices.

4.3 Intercomparison of the devices

The variation between the devices was observed on almost all the samples including the CRMs and their mixtures and was directly related to the specificities of the devices. As explained previously, each device is manufactured differently (water volume, laser cells, etc.). One of the main sources of differences is linked to the use of US and its power. Strong (Cilas, Portable_C) and weak (Coulter) variations were therefore observed after using US for certain samples. The absence of variation for the Coulter demonstrated that the US power might have been insufficient to separate the aggregates. However, it is difficult to investigate US power on CRMs as they are mostly composed of non-cohesive materials and, unfortunately, this parameter was not expressed in Joules by mL [50], so the power used by the different devices was different (Table 1). This observation demonstrates the need to describe the power and

time of US used in future studies to ensure reproducibility, as expressed by Bieganski et al. [5]. Sonication is used to estimate the aggregation index of soil and sediment samples [51], and the results obtained may not be comparable in the absence of such description.

Another strong difference was observed when the main mode was located close to an interval between two classes, a phenomenon also observed for the CRM SRM1003c. The proportion of these two classes could be significantly different between the devices which distribute the mode between them (Fig. 1). Moreover, this distribution can be problematic when the interval is located between clay and silt or silt and sand such as the Azergues sample (Fig. 2, Table 3). Vigilance is crucial when comparing such results.

Results on raw PSD and corrected PSD of the Portable were compared to investigate the impact of the rising tails observed for the finest classes (Electronic Supplementary Material). The results demonstrated that rising tails had a negligible impact on the proportion of the classes. Highest difference was observed on the clay content of the Barcarin sample with 18% on the raw PSD and 11% on the

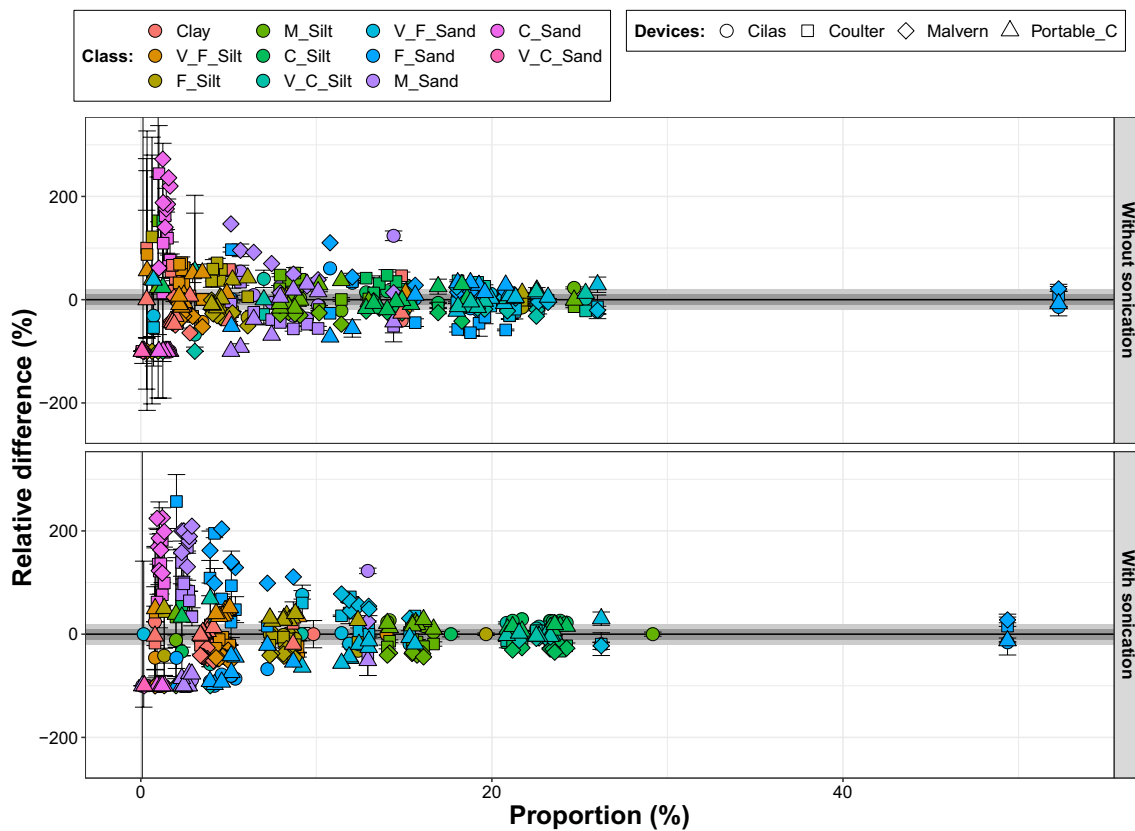


Fig. 7 Relative differences between the mean proportion of each detailed class measured by all the devices and the mean proportion measured by each device with and without US. Grey areas rep-

resent 10% and 20% difference. The abscissa represents the mean proportion of the associated detailed class for all devices

corrected PSD. Moreover, the potential characterization of a lower proportion of clay by deleting the finest classes remained nonsignificant and the values were mainly in the range of the values of the other devices. For this specific study, it was not relevant to remove the finest classes. However, correction remained necessary for the coarse modes induced by the presence of bubbles caused by the pump speed as explained above.

To evaluate the differences and allow users to determine those they can obtain with their devices, Figs. 5, 6 and 7 summarize the observations by grouping all the measures performed on all the natural samples and mixtures for all the parameters studied. Relative differences were then estimated for each device.

For d_{50} (Fig. 5), the larger differences (up to 80% of relative difference with the mean d_{50}) were observed when the sample was a mix of silt and sand particles with a d_{50} ranging from 25 to 50 μm or from 50 to 80 μm , with and without US, respectively. For samples with a high d_{50} (higher than 150 μm), the difference remained lower than 20%, while it could exceed 20% for samples with a low d_{50} .

For the simple classes (Fig. 6), the relative difference was significant ($> 20\%$) when the proportion was lower than 70% and 80% with and without US, respectively, and this difference increased when the proportion decreased and was lower than 25%.

Similar observations were performed for the detailed class (Fig. 7), the relative difference decreased as the proportion increased, but remained mostly higher than 20% whatever the proportion and the US. Therefore, since multimodality was characterized in most cases, detailed classes should be used to describe the trend of the PSD rather than comparing results from different devices on a qualitative basis.

Finally, the specific surface area (SSA) was estimated [52] based on the PSDs of the different references and samples. However, as the proportion of clay was different between the devices, and even nonsignificant, the SSAs estimated by them were all substantially different from each other. It appeared that this parameter is difficult to reproduce on different devices and cannot be compared directly.

5 Conclusion

The tests performed during this study demonstrated that the comparison of particle size measured using different devices can be complicated since differences were observed on certified materials and natural samples. Despite using similar protocols and samples, the results obtained for the selected parameters (d₅₀, simple and detailed classes) were characterized by differences that could be significant. Those differences were directly related to the specificities of the devices, as each device is manufactured differently (water volume, number of laser cells, etc.). In the absence of information on measurement parameters such as US power in J ml⁻¹ and pump speed, the comparison of PSDs from different studies can be erroneous, even for a similar device. It is therefore better to only compare the trend (such as the modal classes or the presence of these types of particle) rather than the measured proportion. Quantitative comparisons should only be performed if the parameters of the different analyses are known.

Differences in d₅₀ were observed even with CRMs. Deviation from the expected d₅₀ observed on all the lasers presented similar trends, with increases in particle size. Although there were differences in the amplitude of uncertainty, they all estimated the d₅₀ for particle-sizes above 10 µm within 10% uncertainty. However, for samples mostly characterized by clay and fine silt particles, the d₅₀ was overestimated. This is important, especially for natural samples where these fractions are often present. Indeed, this information is required in numerous studies. Moreover, on natural sediments mixed with clay silts and sands, the problem of sand is not trivial. Sand is often not seen in these cases when clay size particles are present in quantity. Sieving sediments to remove the sand might be a solution to eliminate the impact of coarse particles.

For the different classes, the main problem observed concerned the position of the modes. Displaying the distribution of the classes greatly depends on the classification chosen relative to the sediment grain size distribution. It is therefore crucial to define the localization of the modes before conducting a comparison with results from different devices. Significant differences can be observed in samples characterized by a PSD with modes between two classes (clay/silt, silt/sand or detailed classes).

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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