Aeolian Research xxx (2014) xxx–xxx

Contents lists available at ScienceDirect

Aeolian Research

journal homepage: www.elsevier.com/locate/aeolia

A new generator for mineral dust aerosol production from soil samples in the laboratory: GAMEL

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ARTICLE INFO

Article history:
Received 25 July 2013
Revised 18 April 2014
Accepted 18 April 2014
Available online xxxx

Keywords:
Desert dust
Aerosol generation
Erosion
Emission
Dust cycle

ABSTRACT

A generator has been developed for producing mineral dust from small samples of desert soils. The objective is to perform a thorough characterization of this new tool and show that it is adapted to the future laboratory studies of the relationship between aerosols and their parent soils.

This work describes the principles and operating protocol of the so-called GAMEL generator. A first series of detailed measurements was performed with a Niger soil. During these tests the aerosol size-distribution was monitored in real time with an optical counter and the particles collected on filters submitted to XRF analysis. This allowed characterizing the emission in terms of time evolution of the aerosol production, repeatability of the experiment, and assessing the influence of such generation parameters as the mass of soil and the frequency and duration of the shaking. For this sandy Niger soil, the optimal generation parameters were found to be 1 g of soil agitated 9 min at the frequency of 500 cycles/min, but the effect of modifications of these recommended values have also been quantified.

In terms of size-distribution as well as of elemental composition, the generated aerosol is found to compare well to the one collected in natural conditions during local events. For testing the capability of the GAMEL to produce aerosols from different soils, tests were also performed with 3 other soils from arid and semi-arid areas. Results showed that the GAMEL is able to produce aerosols whose characteristics encompass the regional variability of naturally produced mineral aerosols.

1. Introduction

Mineral dust represents about 40% of the mass of the natural atmospheric particulate matter (IPCC, 2001). It is emitted by the wind-induced erosion of natural soils in arid and semiarid areas. Although some other generation processes have been recently evocated (Shao et al., 2011; Kok, 2011), this natural generation of mineral aerosols is still thought to be mainly due to the combination of the saltation and sandblasting processes (Gillette and Goodwin, 1974). The loose sand-size particles of the topsoil, which are often aggregates made of solid cores with clay-size materials attached to their surface, are set into motion by the wind stress. The movement of these coarse and heavy grains follows a ballistic trajectory and is essentially horizontal: it constitutes the so-called saltation flow. When they hit the soil at the downwind end of their trajectories, the kinetic energy of these saltating soil aggregates allows the detachment of the very thin (of diameters <20 μm) particles they contain. Thus, this "sandblasting" process produces light particles which, because of their very small settling velocity, can be easily entrained into a vertical upward movement. This description shows that besides being submitted to strong enough winds, there are two necessary conditions for the emission of mineral aerosol from a natural soil to be possible: this soil must contain (1) sand-size particles able to be moved by the wind, and also (2) fine particles which are usually contained in the jumping soil-aggregates themselves. The arid and semi-arid areas meeting these minimal wind and surface characteristics conditions are natural emitters of mineral aerosols and grouped under the denomination of "dust source" areas.

The major dust sources on Earth have been identified and studied for years. For instance, Scheuvens et al. (2013) propose an up-to-date inventory of the North African sources. Each of these sources has its own geological, physical, compositional, soil characteristics and is therefore emitting aerosols differing in such physico-chemical properties as their mineralogical composition and size-distribution.

The precise quantification of the impacts of mineral dust on the environment at regional and even global scales requires an accurate description of the dust cycle taking into account the spatial

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http://dx.doi.org/10.1016/j.aeolia.2014.04.004
1875-9637/© 2014 Published by Elsevier B.V.

Please cite this article in press as: Lafon, S., et al. A new generator for mineral dust aerosol production from soil samples in the laboratory: GAMEL. Aeolian Research (2014), http://dx.doi.org/10.1016/j.aeolia.2014.04.004

variability of the dust composition. For lack of available emission models able to link the aerosol properties with those of the parent soils, one usually attributes some compositional characteristics of the soil or at least of one of its fine fraction (generally the clay-sized fraction) directly to the aerosols (Balkanski et al., 2007). However, due to the complexity of the emission process and particularly of the fractionation it involves, the aerosols properties can hardly be considered as reflecting simply those of the parent soil (Lafon et al., 2006).

Our incapacity to describe correctly the mineral dust aerosol cycle and the variability of the emitted dust properties makes it necessary to study the emission processes more in depth, with the focus being set particularly on the initial composition of the emitted aerosol.

Laboratory generation of aerosols from samples of identified natural source soils would be a convenient method for investigating the emission processes and the spatial variability of the emitted dust. Indeed, simulating the emission in the laboratory would allow removing interferences with other processes always possible in natural outdoor conditions, such as mixing with pollution particles or with dust originating from different, sometimes hard to locate, dust-sources and brought by advection. Unlike in natural conditions, the variability of meteorological conditions also does not come into play. Of all the laboratory tools available for simulating dust emission in the lab, wind-tunnels are probably the most faithful ones because they simulate the saltation and sandblasting processes as they occur on the field.

For example, a wind tunnel has been used in the LISA (Laboratoire Interuniversitaire des Systèmes Atmosphériques) to study both the size distribution (Alfaro et al., 1997; Alfaro, 2008) and the composition (Lafon et al., 2006) of "source aerosols", which is to say freshly emitted aerosols coming from a single parent soil. Such wind tunnel devices are particularly appropriate when one has to control and quantify dynamical aspects such as the influence of wind speed. Their main drawbacks are that they require large quantities of parent soil samples-several tens of kilograms-, lengthy preparations, and controlling accurately within them the mass of aerosol collected on filters for subsequent analysis and avoiding ambient air contamination is relatively difficult. For these reasons, simplified aerosol generators may be preferred to wind tunnels when the objectives of the study permit it.

Several kinds of such simplified dust generators have been designed in the past for a variety of purposes. According to Kaya et al. (1996), an appropriate dust generation device should be capable of producing dust similar in particle size, shape and composition to at least some components of atmospheric dust aerosols, and with limited contamination levels. In their review of the technologies developed for the laboratory generation of dust from geological materials, Gill et al. (2006) proposed a classification of the generators responding to this definition and described in the literature of different research fields (e.g., biomedical research, control of fugitive dust for agricultural purpose, dusty manufacturing facilities, etc.). The classification is based on the technique used to produce the suspended dust. Class A corresponds to the gas dispersion and fluidizing bed techniques, which re-suspend pre-existing fine particles into an air flow (the particle size distribution of the aerosol is not different from the one of the sample from which it has been produced). In this class, one can find for example the generator developed by Marple et al. (1978) and based on the fluidized-bed technique, and the Pitt 3 aerosol generator using the acoustic disturbance of air columns produced by a loudspeaker for re-suspending cotton dust samples (Weyel et al., 1984). Class B and C are more appropriately called dust generators because, as in the natural emission process, they use mechanical agitation for providing the parent material with the kinetic energy necessary for breaking the aggregates it contains. The class B devices generate dust by gravitational effect: the parent sample is made to fall through air in a chamber from which the generated dust is evacuated. A typical example is the dust-fall tube of the USDA Agricultural Research Service in Lubbock, Texas (Singer et al., 2003). Finally, class C generators operate by mechanical dispersion/agitation. The rotating-drum generators belong to this category (e.g., Gill et al., 1999). Because they reproduce aggregate collisions similar to the ones resulting from the saltation/sandblasting processes, these class C generators seem to be the more appropriate for the production of artificial mineral dust with characteristics comparing to those of the dust observed in natural conditions. Very few studies have been dedicated to the description and to the complete characterization of the performances of such generators. The class C generators reported in the literature were mainly developed with the objective of assessing the health risk resulting from an exposure to soil dust emitted during agricultural field operations (e.g., the LDGASS, the Southard Laboratory dust generator, and the EDG (Easy Dust Generator), described in Gill et al., 1999; Domingo et al., 2010, and Mendez et al., 2013, respectively). The aim of this work is to present the characteristics, performance, and operating conditions of the mineral dust aerosol generator developed recently in the LISA: the "Générateur d'Aérosol Minéral En Laboratoire" (GAMEL).

2. Methodology

The general strategy of this study consists in comparing the number size distribution and the elemental composition of the mineral dust produced by the GAMEL in various experimental conditions and with field measurements. In this system, natural soil aggregates are shaken artificially and the ensuing collisions are strong enough for the finest fraction contained in these soil aggregates to be released. In natural conditions, the source of energy for the liberation of the fine particles from the aggregates is the kinetic energy transmitted to them by the wind (Gillette and Goodwin, 1974). It depends on the wind speed as well as on the size of the moving sand grains. According to Alfaro and Gomes (2001), these two parameters determine the size-distribution of the emitted aerosol. In the GAMEL, the collisions are due to the shaking of the soil particles inside a small closed vessel. Consequently, the tested parameters which are considered as having a possible influence on the physico-chemical properties of the produced aerosol are the (1) quantity of soil particles introduced into the system, (2) intensity and duration of the shaking, and (3) nature of the parent soil.

Practically, the aerosol generation capacity of the GAMEL has been documented by repeating generation experiments with a first soil selected for the exploratory tests. In a second stage, different natural source soil-samples were also tested. The results of all these tests were used to define an optimal experimental protocol adapted to the production and collection of aerosol samples representative of a given natural source aerosol and meeting the requirements of the analytical protocols applied to them. The sandy soil of Banizoumbou (Niger) was selected for the tests because the laboratory had previously conducted extensive field measurements in this Sahelian area (Rajot et al., 2008; Sow et al., 2009). This also allows a simple comparison of the aerosol generated in the GAMEL with the one produced in natural conditions.

2.1. The GAMEL set-up

GAMEL is a suction type system in which circulates a constant flow of clean air (Fig. 1).

This flow is assured for the entire system by 3 pumps, each controlled by an individual flow-meter. The room air enters the GAMEL through a HEPA (High Efficiency Particulate Air Filter)

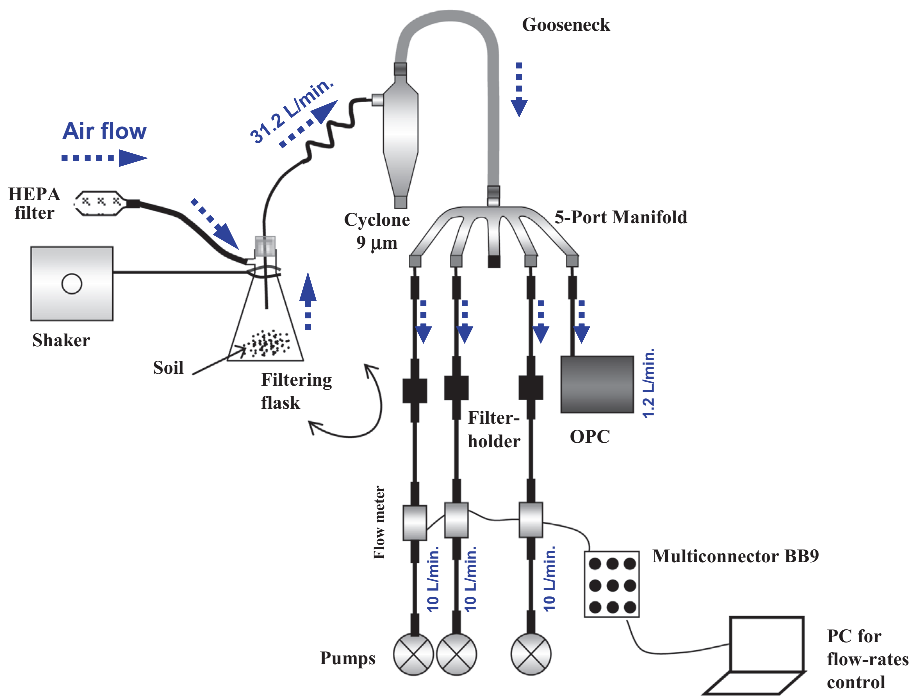

Fig. 1. Set up of the GAMEL.

capsule used to filter it and eliminate the local atmospheric particles. The correct performance of this HEPA filter was tested at the beginning of each generating sequence. This ensures that the only particles present inside the system are those produced by the shaking of the soil. For this purpose, a 500 mL filtering flask containing the soil sample is directly connected to the HEPA capsule and moved by a mechanical wrist-action shaker (Agitest®, 8 mm-amplitude). The collisions taking place between the particles themselves or against the flask walls liberate the fine fraction of the soil aggregates. This fine fraction is entrained through copper and flexible tubes out of the flask and towards the cyclone device. The role of the cyclone is to retain the coarsest particles, in particular the saltating ones, and separate them from the aerosol-sized particles which are the only ones liable to be long range transported in usual natural conditions. The cyclone used in the GAMEL is of the URG type (URG-2000-30EA, URG CORPORATION-Chapel Hill, NC, USA) and has a nominal aerodynamic diameter cut-off of 10 µm for a flowrate of 28.4 L min^{-1}. Here, it is used at a slightly larger flow rate (31.2 L min^{-1}) for which the constructor calibration curves indicate a cut-off size of 9 µm. At the exit of the cyclone, an adapted URG splitter is used to divide the particle-laden air flow into four distinct streams. The first three splitter's outlets are connected to three filter holders equipped with polycarbonate filters (pore size 0.4 µm). The flowrate (10 L min^{-1}) of the three filters is assured by an ALICAT flowmeter (MC-100SLPM-D). The fourth outlet is connected to a Grimm (model G1.108) optical particle counter (hereinafter called the OPC) and operated at a controlled flow rate of 1.2 L min^{-1}. The OPC is used to record the temporal variation of the generated aerosol size distribution (the size being here the scattering cross section equivalent diameter of the particles), over the whole duration of the experiment. Practically, the OPC distinguishes 15 size classes between 0.3 and 20 µm (0.30–0.40/0.40–0.50/0.50–0.65/0.65–0.80/0.80–1.0/1.0–1.6/1.6–2.0/2.0–3.0/3.0–4.0/4.0–5.0/5.0–7.5/7.5–10/10.0–15.0/15.0–20.0/>20.0 µm). In order to document the possible fast variations of the dust concentration in the system, the OPC is set up in our experiments to acquire the data at the minimal possible time-step (6 s). Fig. 2 shows a picture of the experimental set-up.

2.2. Testing procedure

The first tests were dedicated to the definition of an optimized geometric configuration for the system. These tests and their results are not shown here in detail but they led to the current choice of a 500 mL flask, a short flexible tube to minimize the loss of dust on its inner walls, a strong fixation of the shaker to the table and of the flask to the shaker for avoiding parasitic vibrations reducing the efficiency of the shaking. It was also found that for a vessel volume of 500 mL the maximum mass of soil sample allowing it to correctly jump in the flask corresponded to 10 g.

After the GAMEL had been set up, the following testing experiments were performed in order to (1) check the stability of the characteristics of the aerosol produced in a given experiment and the reproducibility of these characteristics in repetitions of this experiment, (2) check its ability to produce a PM10 aerosol similar to the one produced in natural outdoor conditions, and (3) define the optimal working conditions for generating and collecting the mineral aerosol.

Table 1 summarizes the experiments which were made and lists the parameters tested in them.

Because the soil sample is shaken in a closed vessel, the quantity of soil introduced into it influences directly the probability of occurrence of the inter-particle and wall-particles shocks. Nine soil quantities were tested to find the best compromise between the minimum mass ensuring proper representation of the soil sample and the maximum mass needed to maintain the efficiency of the shaking process. The shaking frequency could vary on a scale from 0 to 800 corresponding to the number of cycles per minute. Attention was also brought to the shaking duration and the evolution of the characteristics of the aerosol with time.

In order to compare the aerosols produced in the various experiments, the generated aerosols were characterized by the means of their number size distributions (with the OPC) and of their elemental composition after analysis of the particles collected on the bulk filters. Blanks and ambient air tests were also performed to be compared with the dust-producing cases.

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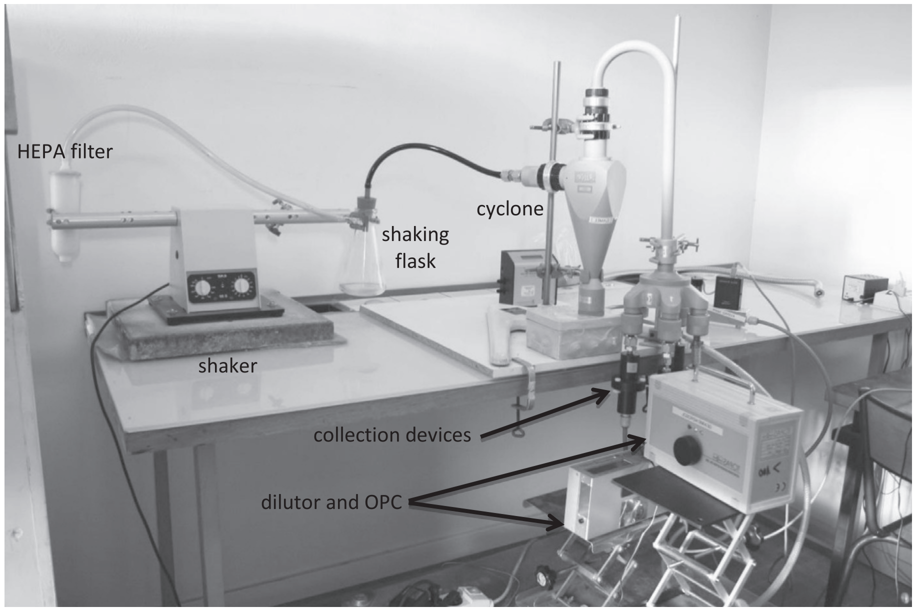

Fig. 2. GAMEL picture.

Table 1
List of generated aerosols and generation conditions. An OPC measurement is performed for each experiment. Sample names are constructed with a letter corresponding to the soil origin, a first number corresponding to the soil quantity in gram, a second number corresponding to the shaking intensity in cycle per minute, and a third number in parentheses corresponding to the number of the repeated experiment.

Soil origin	Soil quantities (g)	Shaking intensity (cycle/min)	Aerosol production period duration (s)	Number of experiments (X)	Sample names
Niger	1	500	180	1	N-1-500(1)
Niger	1	500	540	2–31	N-1-500(X)
Niger	1	500	90	32–39	N-1-500(X)
Niger	3	500	540	1	N-3-500(1)
Niger	3	500	7200	1	N-3-500(2)
Niger	3	500	3700	1	N-3-500(3)
Niger	3	500	4500	1	N-3-500(4)
Niger	1	300	90	1–3	N-1-300(X)
Niger	1	350	90	1–3	N-1-350(X)
Niger	1	400	90	1–3	N-1-400(X)
Niger	1	450	90	1–3	N-1-450(X)
Niger	1	550	90	1–3	N-1-550(X)
Niger	1	600	90	1–3	N-1-600(X)
Niger	1	650	90	1–3	N-1-650(X)
Niger	1	700	90	1–3	N-1-700(X)
Niger	1	800	90	1–3	N-1-800(X)
Niger	0.5	500	90	1–3	N-0.5-500(X)
Niger	0.75	500	90	1–3	N-0.75-500(X)
Niger	1.25	500	90	1–3	N-1.25-500(X)
Niger	1.50	500	90	1–3	N-1.50-500(X)
Niger	2	500	90	1–3	N-2-500(X)
Niger	5	500	90	1–3	N-5-500(X)
Niger	10	500	90	1–3	N-10-500(X)
Spain	1	500	180	1–3	S-1-500(X)
Aïr	1	500	540	1–3	A-1-500(X)
Aïr	1	700	180	1–3	A-1-700(X)
Tunisia	1	500	540	1–4	T-1-500(X)

2.3. Samples and analyses

2.3.1. Measurement sequence

After the filters and the weighted soil sample were installed in the system, the timing of a measurement sequence was as follows:

t_0: the OPC is turned on
t = 1 min: filters' pumps on, checking of the flow rates
t = 2 min: the shaking starts
t = 2 min + collecting time: the shaking is stopped
t = 3 min + collecting time: pumping off
t = 4 min + collecting time: the OPC is turned off: end of the measurement sequence.

Note that before a new soil was studied, the GAMEL was completely disassembled and carefully cleaned to eliminate any trace of remaining dust. This cleaning procedure includes 15 min of sonication, rinsing with deionised water, and drying of the GAMEL elements under a laminar flow hood.

2.3.2. Soil samples

The majority of the testing procedures were performed with samples of the surface horizon of a sandy agricultural soil collected near the village of Banizoumbou in Niger (13°31′N, 2°38′E) where many campaigns dedicated to the study of wind erosion have been carried out since the beginning of the nineties (Bielders et al., 2000; Rajot et al., 2003, 2008; Sow et al., 2009). The other soil samples were collected in a different region of Niger (Aïr, 16°31.39′N, 7°41.93′E), in Tunisia (Douz, 33°25′N, 09°02′E), and in northern Spain (Los Monegros, 41°36′N, 0°32′W). Analyses of the 5 fractions' texture and of the mineralogy by X-ray diffraction were performed on the four soils (Table 2). The Banizoumbou soil is typical of cultivated Sahelian dust sources (Rajot, 2001; Abdourhamane Toure et al., 2011). It is a sandy soil in which the coarse sand fraction represents more than 70% of the mass. It is mainly composed of quartz, but also rich in kaolinite and iron oxides in its clay fraction (Lafon et al., 2006). The Aïr soil was selected for being representative of an erodible non cultivated soil with high clay content. Wind erosion was observed during its sampling. Within it, the clay particles are aggregated to form very stable aggregates in the sand size fraction. These aggregates are mixed with typical sand grains mainly composed of quartz. Coarse sand is also the dominant size fraction. The Douz sample coming from an attested dust aerosol source (Guieu et al., 2010) represents the North Africa sources. This is also a sandy soil, but unlike in the case of the Niger samples, the fine sand fraction largely dominates (almost 60% in mass). It also contains a significant proportion (15%) of calcium carbonate as is generally observed in the north Saharan dust sources (Journet et al., 2013). The clayey soils of the Ebro plain area in north Spain do not constitute a dust source as active as the African ones, but the tilled fields emit dust when submitted to the strong Cierzo wind (Gomes et al., 2003). In this case, the sand content is very low (less than 5%) after wet dispersion, but as in the Aïr sample, clay particles form sand size aggregates in dry conditions. Illite is the main clay mineral and this sample is also the richer in calcium carbonate (calcite and dolomite). In each case, only the first centimetre of the layer of loose particles at the soil surface was collected on the field and subsequently sieved under 1 mm in the laboratory to retain only the soil fraction susceptible to wind erosion.

2.3.3. OPC measurements

The standard calibration of the OPC being performed with non-absorbing latex spheres, the measured dust size distributions are given in terms of latex-sphere optical equivalent diameters. As already indicated in the description of the measurement sequences, the size-distribution of the aerosol is monitored for the whole duration of the experiment. The large concentrations achieved in the GAMEL are well adapted to the rapid (a few minutes) collection of filter samples suitable for subsequent chemical analyses, but too large for avoiding coincidence biases in the OPC measurements. Therefore, the OPC is equipped with a ×100 diluter (TOPAS, model DIL-557). Note that the data presented are corrected to account for this dilution.

The following variables were extracted for each experiment:

- TNC, or total number concentration, which is calculated as the sum of the counts performed in each size class. A value was obtained every 6 s during the sampling period and expressed as particles per liter.
- TNC_{max} is the absolute maximum achieved by TNC during the aerosol production period (defined in Section 3.1).
- TSP, or total suspended particle number, is calculated as the absolute number of particles generated along the whole aerosol production period.
- PNP, or percentage number of particles, is the fraction of TSP represented by each size class. A value can be calculated every 6 s during the generation period.
- MPN, or mean number proportion, is the mean of PNP over the whole aerosol production period. A value could be obtained for each size class of the OPC.

2.3.4. Composition measurements

The bulk composition of the produced aerosol was obtained by analyzing the elemental composition of the mineral dust accumulated on 0.4 μm pore size polycarbonate membranes for the duration of the experiment. The quantitative analysis was carried out directly on each filter using a Panalytical PW2404 4 kW, sequential wavelength dispersive X-ray Fluorescence spectrometer equipped with a rhodium tube. Blank polycarbonate filters were also analyzed for each sampling series. Only the filters meeting the thin layer conditions will be considered in this study for avoiding biases resulting from the possible absorption of the less energetic X rays (Losno et al., 1987). For the 28 mm circular filter deposits used in this study, the maximum total mass meeting the thin layer condition was estimated to 1000 μg (Lafon, 2004).

The XRF spectrometer was calibrated with a GS-N standard (Govindaraju, 1995) and MICROMATTER™ standards (vacuum deposition on nuclepore membranes), which allows taking into account the correction of the grain size effect and determining the individual elemental masses. Finally, the total mass present on the analyzed filter was obtained by summing all the elemental masses converted into the weight of their oxide form most commonly found in soil minerals (Na_2O, MgO, Al_2O_3, SiO_2, P_2O_5, SO_3, K_2O, CaO, MnO, TiO_2, and Fe_2O_3). Elemental ratios characteristic of the presence of the various minerals were also studied here. In order to compare the generated aerosols together and with natural

Table 2
Characteristics of the 4 studied soils in terms of 5 fractions texture, total calcium carbonate and major minerals detected by XRD on powder.

Particle size class	Particle size	Banizoumbou (g/kg)	Aïr (g/kg)	Douz (g/kg)	Spain (g/kg)
Clay	(<2 μm)	19	231	98	410
Fine silt	(2–20 μm)	8	171	102	405
Coarse silt	(20–50 μm)	5	46	202	144
Fine sand	(50–200 μm)	235	126	583	37
Coarse sand	(200–1000* μm)	733	426	15	4
Texture		Sand	Sandy clay loam	Fine sandy loam	Silty clay
Total $CaCO_3$	(g/kg)	<1	28	150	331
Major minerals		Quartz	Quartz Calcite Albite	Quartz Calcite	Quartz Calcite Dolomite Gypsum Illite

* The texture analyses were performed on the same sieved soils as the ones that were used in GAMEL.

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atmospheric aerosols, we used the ratios of the elemental masses to Al and Fe. In addition, the total mass concentration (TMC) was sometimes examined: it corresponds to the ratio of the total mass calculated as the sum of all oxides' masses accumulated on the filter to the volume of air pumped through the filter at the flowrate of 10 L min^{-1}. TMC is expressed in μg L^{-1} for each filter. The quantification limits for each element was estimated as being 10 times the standard deviation of the measurements performed on 3 blank filters.These limits are 0.08, 0.41, 0.04, 0.17, 0.09, and 0.77 μg for Al, Si, K, Ca, Ti, and Fe, respectively.

The final results of aerosol composition are the means of at least three repetitions of each experiment. Their standard deviations are computed and discussed to study the reproducibility of the experiments.

Note that in the following discussion of the OPC and XRF measurements, differences will be considered as significant when they are at the 5% confidence level yielded by *t*-test analysis.

3. Results and discussion

3.1. Typical pattern of a generation experiment

In all, more than 80 generation experiments have been performed in the GAMEL. Though the nature of the soil, the mass introduced into the system and the shaking intensity varied from one experiment to the other, the generation phase can always be characterized by common patterns. Fig. 3, which displays the evolution with time of the total number concentration (TNC) measured by the OPC during a typical experiment, N-1-500(30), can be used for illustrating this point. Before the start of the shaking, the particle concentration in the GAMEL is always close to zero, which emphasizes the efficiency of the HEPA filter at stopping the ambient air particles from entering the system. Conversely, a dust production burst is observed shortly after the beginning of the shaking. It corresponds to a concentration maximum observed in all the size classes of the OPC within the first seconds of the measurements. The moment coinciding with the end of the 6 s-period during which this maximum is reached is chosen as the origin of time. Subsequently, the concentration decreases with time all along the experiment until the shaking is finally stopped. At this moment, the concentration falls almost immediately down to zero. Hereinafter the time period separating the initial concentration peak from the end of the shaking will be referred to as the "aerosol production period".

The general shape of this typical emission curve obtained with GAMEL is similar to the ones obtained in the experiments made with other dust generators such as the EDG (Mendez et al., 2013), the LDGASS (Gill et al., 1999), and the Southard Laboratory Dust Generator (Domingo et al., 2010).

In order to document the dust production phase more precisely, we studied first the characteristics of 30 repetitions of the same experiment. Then, we analyzed separately the influence of the generation conditions (mass of soil and shaking intensity) on the characteristics of the dust produced in the GAMEL.

3.2. Analysis of the standard experimental conditions

In this section, we analyze a detailed case corresponding to the optimal "standard" conditions of use (1 g of parent soil shaked at 500 cycles/min.) defined after examining the results of the series of tests performed on the Niger soil and dedicated to the study of the influence of the generation conditions, as described later (Section 3.4).

3.2.1. Variability of the dust production

Even when the same generating conditions (type of soil, soil sample mass and shaking intensity) were used, the maxima and therefore the general level of number concentrations can differ from an experiment to the other. In order to document this variability more precisely, the TNC_{max} measured using the optimal "standard" conditions in 30 repetitions of the same experiment are reported in Table 3. The average value of these maxima is 3.60 10^6 part./L, with a relative standard deviation (RSD) of 38%. This value of the RSD shows that the level of reproducibility in terms of absolute values of the concentration is rather low. A part of this variability lies in the fact that the OPC measurements are performed over periods of 6 s, which implies in turn an uncertainty of 6 s on the definition of the time origin of the experiment. Another part of this variability could lie in the soil sampling uncertainties, themselves linked to the splitting of the bulk soil samples into much smaller subsamples.

3.2.2. Size-distribution and its evolution with time

After the initial period, a supply-limitation in fine particles seems to be denoted by the progressive decrease with time of the efficiency of the dust production. This can probably be explained by the limited size of the soil samples used in the experiments. In each size-class of the OPC below the diameter cut-off of

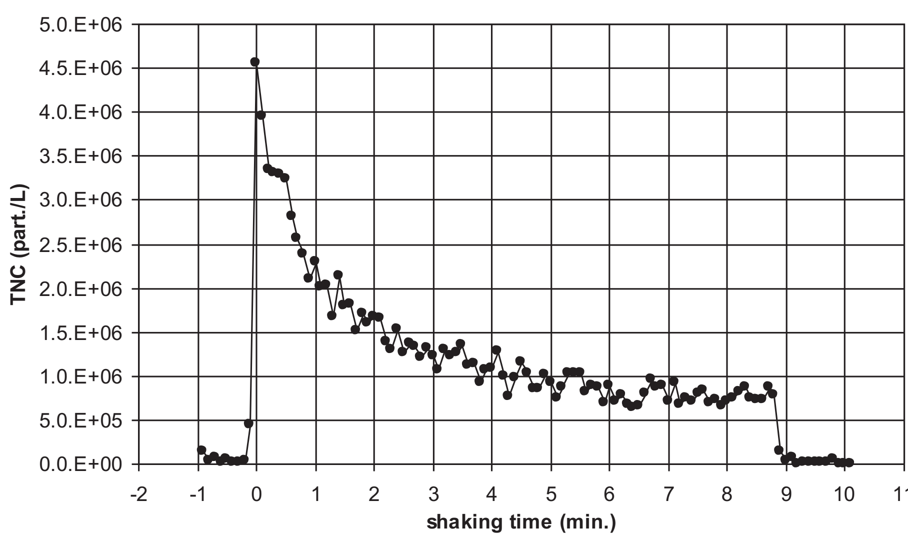

Fig. 3. Total number concentration (part./L) measured by the OPC during a typical generation experiment.

Table 3
Total number concentration maximal (TNC_{max}) and mean particle number percentages (MPN) of two size classes as measured in 30 repetitions of the same experiment.

Sample	TNC_{max} (part/L)	MPN 0.30–0.40 μm (%)	SD (%)	MPN 2–3 μm (%)	SD (%)
N-1-500(2)	2.372×10^6	42.10	7.31	10.76	3.84
N-1-500(3)	3.01×10^6	42.84	6.60	10.85	2.87
N-1-500(4)	6.941×10^6	45.84	7.78	8.39	3.81
N-1-500(5)	1.475×10^6	38.82	6.62	12.22	2.76
N-1-500(6)	2.585×10^6	39.12	6.11	11.63	2.83
N-1-500(7)	2.373×10^6	37.83	6.34	11.87	3.10
N-1-500(8)	3.615×10^6	38.69	5.37	12.13	2.72
N-1-500(9)	2.53×10^6	38.01	6.10	12.43	2.93
N-1-500(10)	2.737×10^6	38.04	6.06	12.41	2.81
N-1-500(11)	2.995×10^6	45.38	6.53	8.50	2.46
N-1-500(12)	3.44×10^6	46.88	6.65	8.10	2.52
N-1-500(13)	1.57×10^6	47.58	5.75	7.85	2.19
N-1-500(14)	2.994×10^6	43.49	5.87	9.25	2.65
N-1-500(15)	3.513×10^6	43.08	6.79	9.59	1.97
N-1-500(16)	2.53×10^6	41.74	5.45	9.92	2.54
N-1-500(17)	2.732×10^6	38.80	6.25	11.35	2.50
N-1-500(18)	2.509×10^6	38.57	5.82	11.51	2.86
N-1-500(19)	2.11×10^6	37.67	6.32	12.27	2.63
N-1-500(20)	3.928×10^6	48.21	4.19	7.31	1.71
N-1-500(21)	3.545×10^6	47.29	6.13	7.84	1.76
N-1-500(22)	5.784×10^6	44.03	5.37	10.05	1.90
N-1-500(23)	4.308×10^6	44.16	5.26	10.34	1.87
N-1-500(24)	5.097×10^6	42.35	5.02	10.70	2.05
N-1-500(25)	3.791×10^6	40.32	5.20	11.75	2.00
N-1-500(26)	4.922×10^6	40.00	4.10	11.28	1.67
N-1-500(27)	4.696×10^6	39.93	4.70	11.63	1.82
N-1-500(28)	3.546×10^6	38.74	5.05	12.16	1.96
N-1-500(29)	5.684×10^6	40.53	4.05	11.25	1.90
N-1-500(30)	4.562×10^6	39.90	4.18	11.95	1.95
N-1-500(31)	5.603×10^6	40.34	3.75	11.44	1.64

the cyclone (9 μm) the general pattern of Fig. 3 was observed. This shows that the soil disaggregation produces particles in all the size classes <9 μm, including the submicron size range. In particular, the most abundant size class is the 0.3–0.4 μm one with an average proportion of 41.6% (SD = 3.5) (Table 3). In the supermicron range of sizes, the 2–3 μm channel represents on average 10.6% (SD = 1.7) of the total number of particles.

The fact that the relative standard deviations of the relative size-distributions calculated from the 30 MPN values remain smaller than 15 % shows that the variability of the size distribution between experiments performed in the same conditions is far from being as important as the variability of the concentration. The initial (in the sense that it is calculated as the mean number percentage on the first minute of the aerosol production period) relative size distribution of the aerosol generated using the optimal "standard" conditions with the Banizoumbou soil is reported on Fig. 4.

We already mentioned that after the initial peak, the concentration in each size class tends to decrease according to the general pattern of Fig. 3. However, a closer examination of the rates of decrease shows that these rates are not exactly the same for all the size-classes. In order to quantify this effect, a mathematical law of the power type can be fitted to describe more precisely the progressive decline of the concentrations after the initial maximum.

$$C_i/C_{0,i} = t^{-a} \quad (1)$$

Note that this equation, in which $C_{0,i}$ is the initial concentration of the particles in the ith size-class and t is time expressed in seconds after the obervation of TNC_{max}, is not valid for $t = 0$ and can be applied only after the first 6 s-period of the generation. As denoted by the large value of r^2 (>0.8 in all cases) the adjustment obtained in these conditions is fairly good.

The fact that the value of the exponent a increases with size (Table 4) shows that the production of the coarsest particles in the GAMEL slows down faster than the one of the finest particles. This suggests that there might be a stronger supply-limitation of the coarse particles than of the submicron ones in the sand grains used for the generation experiments. Due to this progressive depletion of the largest size-classes, the relative proportion of fine (up to 1 μm approximately) particles increases with time as can be seen on Fig. 5.

It can be noted that even if the system generates an aerosol whose size distribution evolves slightly with time, the repetition of the same generation sequence replacing the soil sample by a new one yields aerosols with nearly identical size characteristics. This implies that, in the case of time-integrated measurements such as the collection of aerosols for ulterior analyses, the duration of the sampling must be carefully controlled if one wants to collect comparable aerosols. Based on three long-time experiments (N-3-500(2), N-3-500(3) and N-3-500(4)) performed for checking the continuity of the generation process, we examined how the concentration and size evolution of the generated aerosol along the experiment could affect the bulk characteristics of the time-integrated aerosol. The decrease of the concentration was found to be continuous for at least the first two hours of an experiment performed with the same sample. Over this particularly long duration, the dust concentration is reduced by one to two orders of magnitude but the disaggregation process does not really stop.

Calculating the absolute number of particles accumulated after a given duration, we found, as expected from the results presented in Fig. 3, that the increase of the accumulated number of particles was not simply proportional to the collection time but slowed down as the duration increased. For example in the case of the N-3-500(2) experiment, if one pumps through the filter for 1 min, 4.4×10^7 particles will be collected as compared to only 6.7×10^7 particles after 2 min. The filtering would need to be continued for 3.5 min for collecting 8.8×10^7 particles, which is to say twice the number of particles sampled during the first minute. This general decrease has to be taken into account when trying to estimate the quantity of aerosol produced by the GAMEL during a given experiment (for which the maximum TNC is known).

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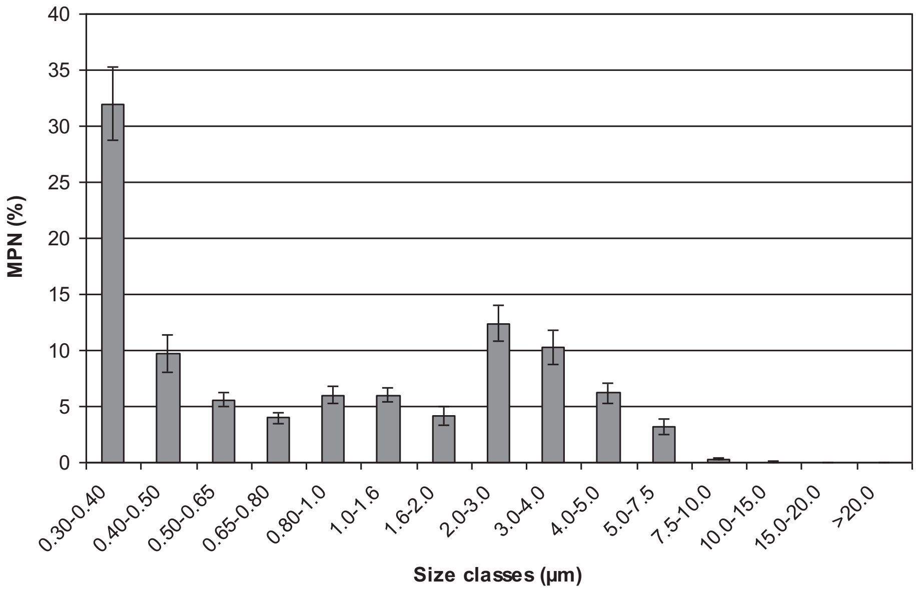

Fig. 4. Relative mean particle number (MPN) by particle size class of an aerosol generated with the Banizoumbou soil. The error bars represent the standard deviation of the 30 experiments.

Table 4
Values of the exponent *a* of Eq. (1) for each size class.

d (μm)	0.30–.40	0.40–0.50	0.50–0.65	0.65–0.80	0.80–1.0	1.0–1.6	1.6–2.0	2.0–3.0	3.0–4.0	4.0–5.0	5.0–7.5
a	0.358	0.430	0.410	0.419	0.386	0.480	0.479	0.704	0.838	0.986	0.917

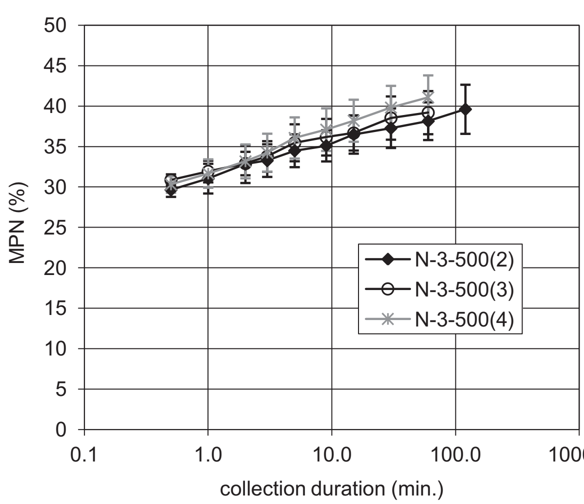

Fig. 5. Mean number proportion of 0.3–0.4 μm size class calculated as a function of collection duration along the three long-time experiments N-3-500. The error bars represent the standard deviation of all the OPC measurements for this size class along the considered collection duration.

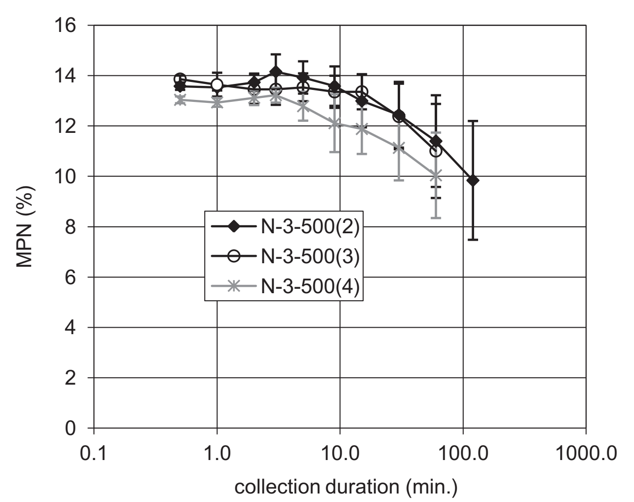

Fig. 6. Mean number proportion of 2–3 μm size class calculated as a function of the collection duration along the three long-time experiments N-3-500. The error bars represent the standard deviation of the all the OPC measurements for this size class along the considered collection duration.

In order to better understand the implications of this evolution of the size-distribution during the generation experiment on the characteristics of the aerosol collected on the filters, the proportion of the number of particles in the class 0.3–0.4 μm and 2–3 μm can be plotted as a function of the collection duration (Figs. 5 and 6, respectively).

The evolution of the proportion of 0.3–0.4 μm particles clearly showed again, that, in addition to the evolution of the absolute concentrations, the size distribution of the aerosol produced by the GAMEL changed along the production period. This evolution was relatively slow as compared to the one of the number concentration. Nonetheless, it is not possible to neglect this variability when an accurate quantification of the characteristics of the aerosol generated in the GAMEL is needed.

The production of the two sizes classes was affected by the generation time (Figs. 5 and 6), but for the 2–3 μm particles two regimes could be observed: in the first 9 min of generation the number proportion of 2–3 μm remained stable between 13% and 14%, then this proportion declined regularly to reach 10% after 100 min which again suggests the existence of a supply-limitation for the supermicron particles available in the soil samples.

As a matter of consequence, the collection time needs to be taken into account in the generation protocol. In particular, it is advisable to limit the generation to 9 min, in the case of Niger soil. Should the accumulation of a large aerosol mass on a filter be needed, the production process could be repeated as many times as necessary with fresh soil samples.

3.2.3. Mass concentration and elemental composition of the generated aerosol

Seven filters were collected during the series of 30 repetitions performed in the experimental conditions of Table 3 (shaking 500, soil quantity 1 g, collection duration on filters of 9 min). In addition, three other filters connected to the three sampling lines were loaded in parallel for a unique 3 min experiment.

First, we checked if the three parallel sampling lines provided similar results. The total mass collected (TMC) on the three filters varied between 180 and 199 μg, with a mean of 187 μg and a relative standard deviation of 6%. The mean ratios of Si, K, Ca, Fe, and Ti to Al, and the associated uncertainty (RSD) are given in Table 5. The very low Ca content in the Niger dust aerosol explains the large relative uncertainty on the quantification of this particular element. In all the other cases, the RSD is low on both the measured total mass and the elemental ratios and of the same order of magnitude as the instrumental uncertainties (10%). This shows that the three parallel sampling lines collect similar aerosol samples in terms of mass as well as in terms of elemental composition.

For the 7 repetitions, the mean TMC on the filters was 292 μg with a relative standard deviation of 27%. Based on the OPC measurements, the TSP for these 7 repetitions had a mean value of 1.14×10^7 part. (RSD = 43%). Consistent with the variability of the TSP variability, the one of the TMC was also rather large. Conversely, the elemental ratios calculated with Al chosen as a reference (Table 5) did not significantly differ from those measured for 3 min and had the same low RSD. Since the elemental ratios present a low variability from one filter to the other, we conclude that with the Niger soil the aerosol production is well reproducible in terms of elemental composition in the 9 min during which the size distribution remained practically constant (Section 3.2.2).

In summary, with the GAMEL it is possible to generate from a very small soil sample an aerosol whose relative number size distribution and elemental composition are reasonably well repeatable. The repeatability of the generation only requires that the "generation conditions", namely the combination of shaking intensity, soil quantity, and shaking duration used in the experiments are well controlled. The concentration levels produced by the shaking are more variable but they remain in any case compatible with a collection of the particles on filters for subsequent analyses.

For the sake of comparison with other generators, the average PM10 concentration has been calculated from the measured mass on filter. This concentration is found to be 6.2 and 3.2 mg m^{-3} respectively for the 3-min and the 9-min optimal "standard" with the Banizoumbou soil. This order of magnitude compares to the one obtained in previous studies (Amante-Orozco, 2000) using the LDGASS generator. The GAMEL also produces about 10 times more aerosol than the EDG one for comparable soil masses.

3.3. Comparison with a natural aerosol

In the frame of the AMMA (African Monsoon and Multidisciplinary Analyses) international campaign (Rajot et al., 2008), the emission of mineral aerosols have been extensively studied in Banizoumbou (Niger). Therefore, it is possible to compare the results obtained in the laboratory with those obtained on the field in natural conditions. During AMMA, the size-distribution of the local source aerosol was monitored with the same OPC as the one connected to GAMEL and particles were also collected on filters for subsequent XRF analysis in the laboratory. Four events belonging to 3 different types of typical situations are chosen to perform the comparison: (1) an event of intense local erosion, corresponding to a convective situation (hereinafter referred to as LocEr-C), (2) an event of moderate local erosion associated to monsoon wind (LocEr-M), and (3) two events during which dust was not produced locally but advected from relatively close sources (CloEr). In these last two cases, the dust comes from Sahelian sources similar to the local one but a part of its coarsest particles has been removed through deposition during the short range transport. For more details on the local erosion cases the reader is referred to Sow et al. (2009) and to Rajot et al. (2012) for the transport ones. Note that the size cut-off of the sampling system used on the field was larger (ca. 40 μm) than the one of GAMEL (9 μm). For making the comparison of the field and laboratory measurements possible, the raw field OPC measurements performed in each size class were corrected using the efficiency of the URG cyclone provided by the manufacturer and reported in Table 6. This transformation yields an OPC dataset which corresponds now to the size distribution of the natural atmospheric aerosol as it would be seen if it were collected through the collecting part of the GAMEL. Finally, the laboratory and field cases number measurements are converted into a volume distribution on the basis of a spherical hypothesis. The relative size distributions obtained for the field cases can be compared (Fig. 7) to the mean of the GAMEL measurements performed in standard conditions with the Banizoumbou soil.

Large similarities are observed between the GAMEL and the field size distributions. The general shape of these distributions is the same, displaying the same supermicron mode of particles with an absolute maximum between 4 and 7 μm. In the submicron range, the proportion of particles produced in the GAMEL is generally smaller than the one observed on the field but closer to the one measured in the energetic convective conditions. This can be

Table 5
Results of XRF analyses (mean and relative standard deviations) on filters collected during N-1-500 experiments as total mass on filter and elemental ratios for 3-min parallel samplings (3 filters for 1 generation) and 9-min series of sampling (1 filter for 7 generations).

	Mass on filter in μg (RSD)	Si/Al (RSD)	K/Al (RSD)	Ca/Al (RSD)	Fe/Al (RSD)	Ti/Al (RSD)
3-min parallel samplings	187 (6%)	2.42 (3%)	0.10 (6%)	0.03 (30%)	0.56 (5%)	0.13 (2%)
9-min series of sampling	292 (27%)	2.42 (4%)	0.11 (6%)	0.03 (18%)	0.56 (6%)	0.14 (3%)

Table 6
Efficiency of the URG cyclone for each size class.

Mean geometric diameter of the size class (μm)	0.3	0.4	0.6	0.7	0.9	1.3	1.8	2.4	3.5	4.5	6.1	8.7	12.2
Transmission (%)	100	100	100	100	100	100	100	100	100	100	96	60	24

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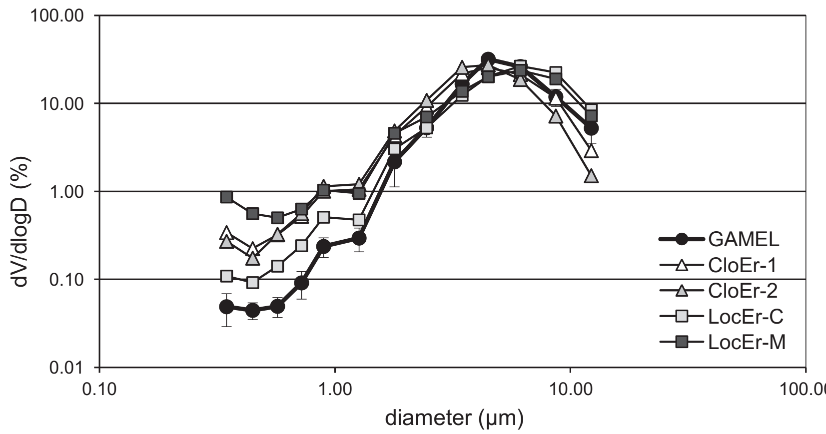

Fig. 7. Aerosol volume relative size distributions measured (1) on the field in Banizoumbou (Niger) and (2) in GAMEL during generation experiment performed with the Banizoumbou soil samples. The errors bars for GAMEL measurements represent the standard deviation of the 30 repetitions of the same condition experiments.

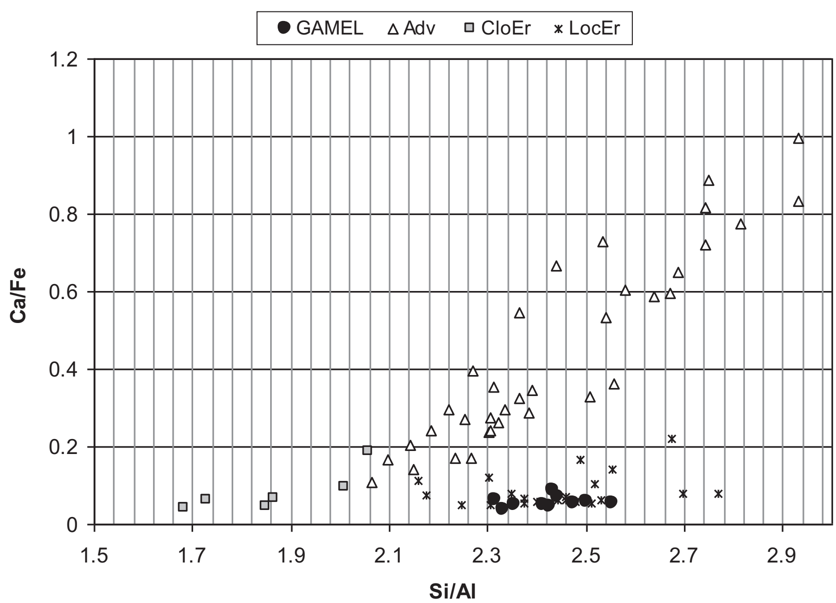

Fig. 8. Ca/Fe ratios in function of Si/Al ratios measured by XRF on filters of aerosols generated in GAMEL with Niger soil and on filters collected in Niger natural atmosphere.

explained by the fact that (1) the presence of a fine pollution component in the Banizoumbou aerosol could enhance the amplitude of the size-distributions in the submicron range, and (2) that this effect is less marked in the strong erosion (convective) conditions when the atmospheric load in mineral dust is so large that the contribution of the pollution particles to the relative size-distributions becomes almost insignificant. In conclusion, the GAMEL is able to generate an aerosol whose size characteristics are quite similar to the ones of the mineral dust measured on the field during local erosion.

The elemental composition of the aerosol generated in the GAMEL can also be compared to those of the aerosols sampled on the field. The comparison involves the N-1-500 series of GAMEL filters on the one hand and the field measurements performed during the AMMA campaign (Rajot et al., 2008) on the other hand. Among these, three types of events can be distinguished: (1) local erosion events associated with either convective or monsoon situations, (the LocEr-C and LocEr-M measurements of Fig. 7), (2) short-range advection cases already referred to as CloEr events, and (3) long-range advection (Adv) cases, not considered so far in our size-distribution comparisons.

The 7 N-1-500 filters and 62 filters collected in the field (21 filters for LocEr, 6 for CloEr, and 35 filters for Adv) were analyzed by XRF. Fig. 8 displays the Ca/Fe ratio as a function of the Si/Al ratio for each case.

Note that Si/Al is sensitive to both the relative proportion of quartz and aluminosilicated minerals and to the mineralogy of aluminosilicates. Close to the sources where the proportion of coarse quartz grains is known to be more important than after transport, this ratio is expected to be larger than farther downwind. The Si/Al ratio also reflects the composition of the parent soil. For instance, the relative abundances of the fine, easily transported, clay species (kaolinite, illite, smectite, palygorskite...) particles or the content in diatoms skeletons made of amorphous silica, will have a significant impact on the Si/Al ratio at a distance from the source (e.g., Lafon et al., 2006). The Ca/Fe ratio also depends strongly on the parent soil's composition. This ratio is largely influenced by the presence of iron oxides and calcium carbonates (calcite, dolomite). In the Sahel, soils are richer in iron oxides than in the North Saharan areas and are generally depleted in calcite. This capability of the elemental composition to reflect the origin and transport history of the aerosol has been used to discriminate the different types of episodes (long-range advection vs local and short-range emission) sampled in Niger during the AMMA campaign (Rajot et al., 2008).

In good agreement with the previous observations based on the analysis of the size-distribution (Fig. 7), the composition of the aerosol generated by the GAMEL was reasonably similar to the one of the aerosol produced by local erosion in Banizoumbou (Fig. 8). In particular, it was characterized at the same time by the low Ca/Fe ratio typical of the Sahelian soils poor in calcite and rich in iron oxides, and by a relatively large Si/Al ratio reflecting here the proximity of the source. Note that, as a result of the selective deposition of the coarse quartz particles during transport, the aerosols coming from more distant sources of the Sahelian area display lower Si/Al ratios than the aerosols sampled during local erosion events.

The similarity of elemental composition between the GAMEL-generated and locally-produced natural aerosols was further confirmed by the comparison of other elemental ratios such as Ti/Al, K/Al, and Mg/Al whose means and associated RSD for the GAMEL aerosol were 0.14 (3%), 0.11 (6%) and 0.05 (3%), respectively, to be compared to 0.14 (8%), 0.11 (14%), and 0.05 (20%) for the local erosion aerosol.

In summary, the particle size distributions and elemental ratios of aerosols generated in the GAMEL with samples of the Banizoumbou soil were very similar to the data produced locally under natural conditions.

3.4. Influence of the generation parameters

In the previous experiments, the generation conditions were kept constant. In the following, we want to explore the impact of modifications of the most important generation parameters (shaking frequency and amount of soil sample) on the characteristics of the generated aerosol. More precisely, the influence of the two parameters was explored independently by performing experiments in which the frequency of the shaking varied between 300 and 800 cycles per minute, and the soil mass between 0.5 and 10 g. During these tests, the sampling duration was maintained constant (90 s). The results of these tests are detailed in the two following sections.

3.4.1. Influence of the shaking intensity

3.4.1.1. On the levels of dust concentrations. At increasing step of 50, each shaking rate between 300 and the maximum of 800 cycles per min has been tested three times with 1 g of soil. The movements of the soil grains began to be observable with the naked eye for a shaking rate of 400. Results are presented as mean values of the three repetitions (with standard deviation as error bar). Regarding TNC_{max} (Fig. 9), a strong variability was once again observed: the error bars represent up to 50% of the mean values. The very low values of the TNC_{max} observed for shaking rates <400 is explained by the low kinetic energy of the soil aggregates. The production of particles increased practically exponentially with the shaking rate between 300 and 600 cycles per minute but finally tended toward saturation. Note that the range of TNC covered more than 3 orders of magnitude. The efficiency of the dust production was thus found to be very sensitive to variations of the shaking intensity.

3.4.1.2. On the size distribution. The average relative proportions of the 0.3–0.4 μm, 0.8–1 μm, and 2–3 μm size classes (Fig. 10) did not change significantly for shaking rates increasing between 400 and 600 cycles/min. The large error bars for the weakest shaking frequencies are due to the very low values of the concentrations measured at these frequencies. Above 400 cycles/min, a significant amount of dust was generated and the relative number size distribution integrated over the first minute of the production period was then less dispersed. It was only above 650 cycles/min that a relative decrease in the proportion of coarse (2–3 μm) particles can be observed. Above 650 cycles/min, the proportion of the 0.8–1 μm class becomes greater than the proportion of the 2–3 μm class. This is in good agreement with the fact that at increasing kinetic energies the soil aggregates liberate aerosols enriched in fine particles (Alfaro and Gomes, 2001; Sow et al., 2009). This suggests that the generation of dust aerosol by GAMEL, for this particular soil, needs to be done at shaking rates >400 cycles/min, but also that the size-distribution of the aerosol would not be exactly the same were the shaking rate inferior or superior to 600 cycles/min.

3.4.1.3. On the mass concentration and elemental composition. Only the samples produced at frequencies above 400 cycles/min were studied here because the masses collected on filters at lower frequencies are too low to be properly quantified with XRF. One filter was analysed for each shaking rate. As already observed with the maximum TNC, the total calculated mass concentration increased by more than one order of magnitude with the shaking rate between 400 and 600 cycles/min before it eventually saturated above this last value (Fig. 11). Interestingly, the fact that the aerosol mass collected on the filters strongly depends on the shaking

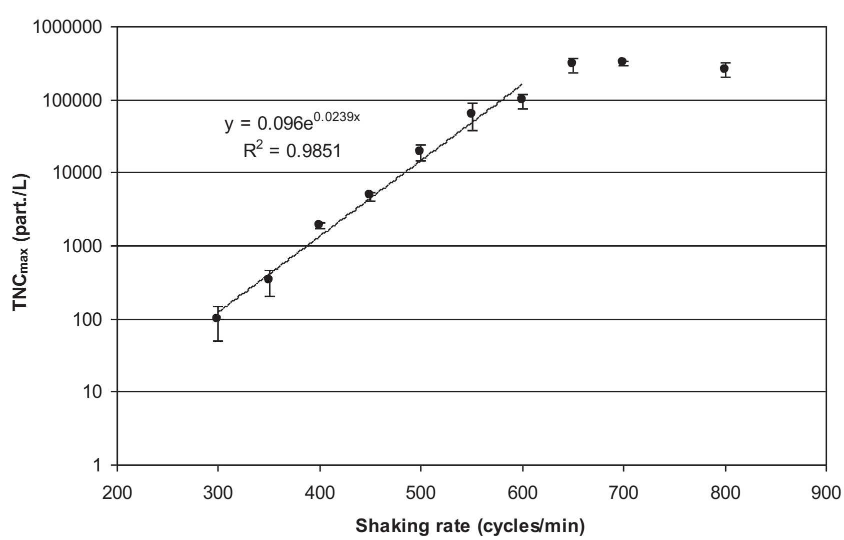

Fig. 9. Dependence of the maximum total number concentration to the shaking rate. The error bars represent the standard deviation of three trials.

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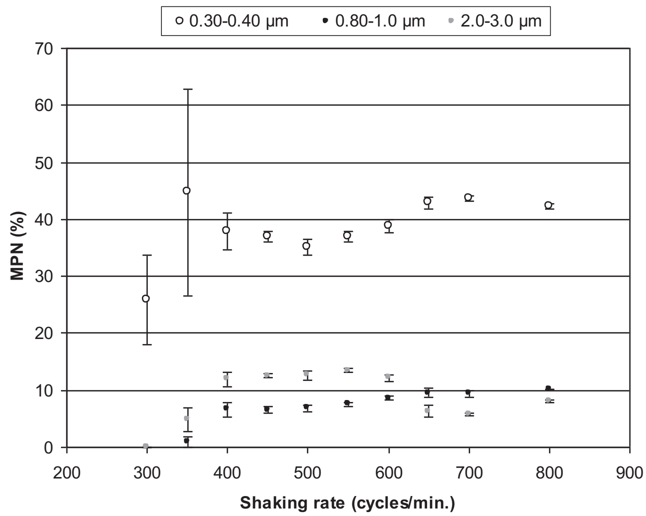

Fig. 10. Mean number proportions of three size classes as a function of the shaking rate. The error bars represent the standard deviation of three trials.

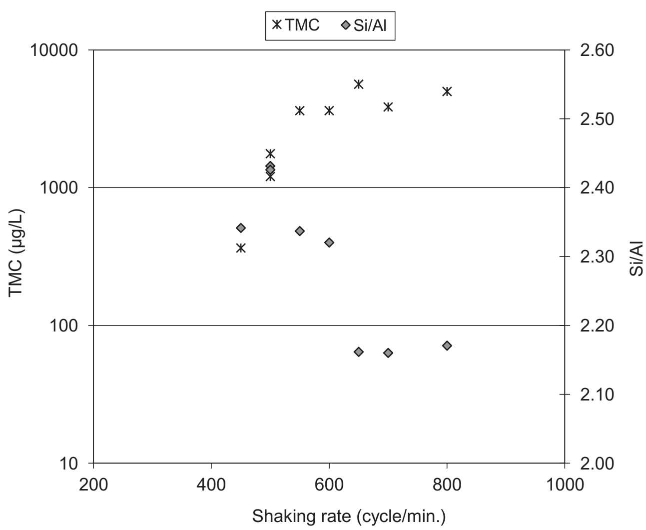

Fig. 11. Total mass concentration and Si/Al ratios obtained from XRF measurements performed on GAMEL filters as a function of the shaking rate.

frequency constitutes a powerful tool for controlling the mass production.

Regarding the elemental composition, Fig. 11 shows that for shaking frequencies between 400 and 600 cycles/min, the Si/Al ratio lies between 2.32 and 2.43, which is to say in the same range as the one (between 2.31 and 2.55) of the 30 "same-conditions-experiments" described in Section 3.2. However, the Si/Al ratio achieves smaller values (mean value: 2.16, RSD = 0.3%) in the experiments performed at shaking rates between 650 and 800 cycles/min. This smaller value is consistent with the relative impoverishment in coarse particles (mainly quartz) observed with the OPC (Fig. 10) during the experiments performed at these large shaking rates.

3.4.2. Influence of the parent soil quantity

As already mentioned above, a compromise must be found between the mass of soil used in the experiment and the volume of the flask for allowing the soil aggregates to move relatively freely and to be submitted to sufficiently numerous shocks. For the 500 mL flask used in our experiments, we determined empirically that this condition was met for soil quantities ranging from 0.5 to 10 g.

3.4.2.1. Influence of the size of the parent soil sample on the number concentration. For each soil quantity, the generation experiments were performed three times at a shaking rate of 500 cycles/min. The OPC results are presented as mean values of the three repetitions (with standard deviation as error bar). In spite of a relatively important variability observed in the repetitions, the dependence of the TNC_{max} to the soil quantity was quasi linear up to masses of 2 g (Fig. 12). Above this value, a slower increase was obtained although no clear saturation effect appears, contrary to what was observed in the study of the influence of the shaking rate. Above 2 g of soil, we observed that the error bars were quite larger than below This indicates that the generation experiment below 2 g would be preferable for repetition quality.

3.4.2.2. Influence of the size of the parent soil sample on the size distribution. At the shaking rate of 500 cycles/min used for the tests, the size distributions – Presented as the mean relative proportions of 0.3–0.4 µm, 0.8–1 µm and 2–3 µm size classes over the first minute of production period on Fig. 13-were found to be independent of the soil quantity introduced in the GAMEL.

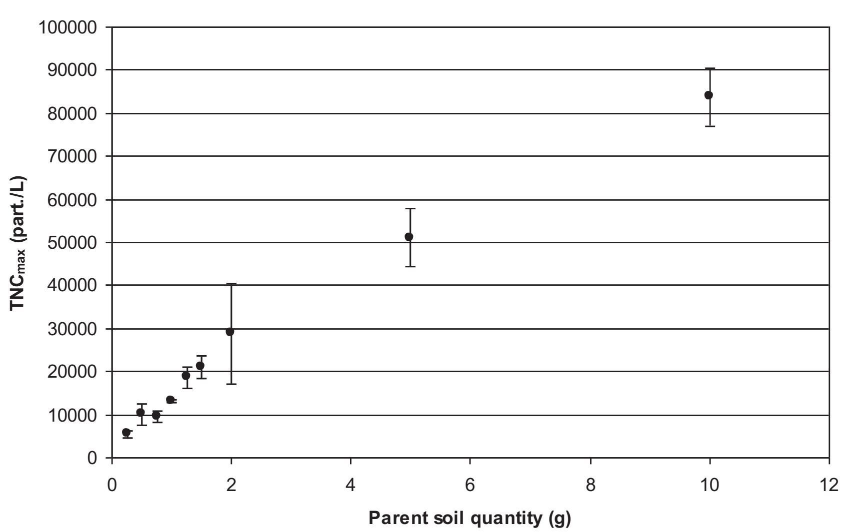

Fig. 12. TNC_{max} as a function of soil quantity. The error bars represent the standard deviation of three trials.

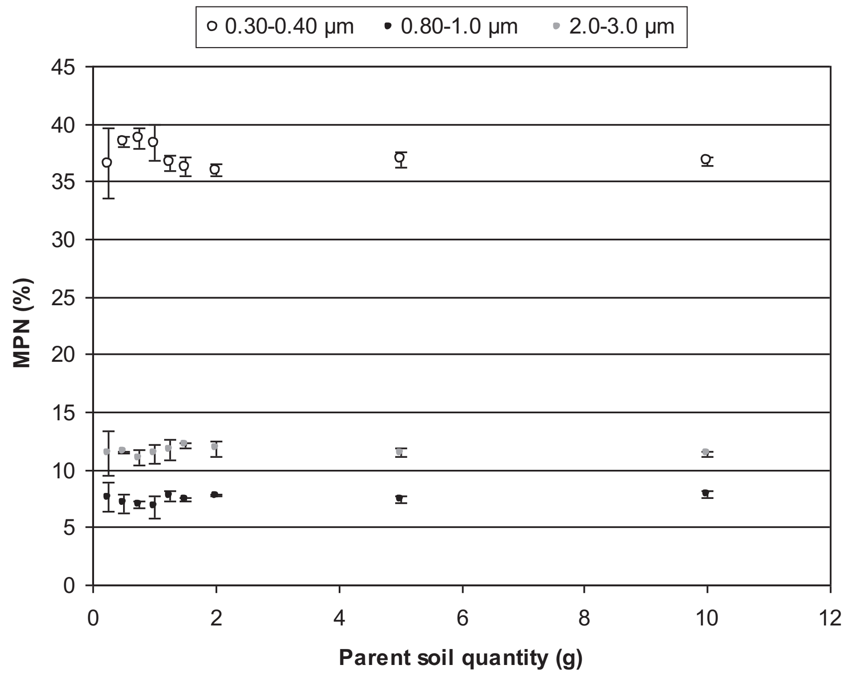

Fig. 13. Relative proportions of three size classes as a function of the soil quantity. The error bars represent the standard deviation of three trials.

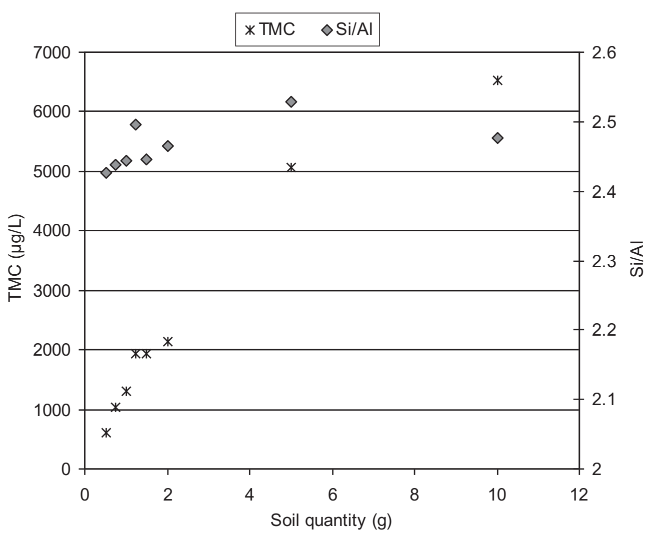

Fig. 14. Influence of the soil quantity on the total mass concentration and Si/Al ratios obtained from XRF measurements performed on GAMEL filters.

3.4.2.3. Influence of the parent soil sample size on the mass concentration and elemental composition. The behavior of the total mass concentration (Fig. 14) is similar to the one of the TNC_{max}. A quasi linear increase is observed for soil masses less than 2 g and a slower increase above this value. No influence of the soil quantity on the elemental composition of the collected aerosol has been be detected. For instance, a mean value of 2.46 (RSD = 2%) was obtained for the Si/Al ratio. This value was not significantly different from the one (2.42 (RSD = 4%) which was obtained in the same-generation-conditions experiments.

Thus, the aerosol generated in the GAMEL at the shaking rate of 500 cycles/min had a size-distribution and a composition independent of the soil quantity initially introduced inside the flask. Therefore, this soil quantity can be considered as a convenient parameter which can be easily adjusted for controlling the aerosol concentration and the mass of particles collected on the filters. As standard conditions for the Niger soil, we recommend choosing 1 g of soil to stay in the lower variability domain.

3.5. Tests performed with soils of different nature

So far, only the Banizoumbou soil has been submitted to all the tests. Because the dust emission process does not only depend on the wind strength but also on the soil properties, we wanted to check the ability of the GAMEL to produce mineral aerosols from soil samples of completely different natures. Thus soil samples were selected to cover the broadest possible range of particle size distributions encountered in soils undergoing wind erosion. These soil samples were also selected for their various carbonate contents, well known to enhance soil aggregates stability. The studied soils have been described in Section 2.3.2.

3.5.1. Size distribution

The relative number size-distributions yielded by the OPC measurements are reported on Fig. 15. Each plot is the mean of three repetitions of generation experiments performed at a shaking rate of 500 cycles/min for one minute. In the four cases, the same type of size distribution with a supermicron population centered around 2 µm and two submicron ones centered on 0.8 µm and on a little less than 0.5 µm was observed. However, on closer examination of this general pattern, slight differences in size-distribution can be observed from an aerosol to the other: (1) the supermicron population was slightly coarser in the Aïr and Spanish cases than in the Banizoumbou and Douz ones, and (2) the aerosol generated from the Aïr soil was substantially less rich in particles of the 0.8 µm population than the other three.

As done before for the Banizoumbou soil, we quantified for the Spain and the Douz cases the decrease with time of the concentrations. Practically, we determined the value of the exponent *a* involved in Eq. (1) for several particle size classes. Note that the case of the Aïr case could not be treated because the concentrations were too low (see below, Section 3.5.2) and variable for the fitting of Eq. (1) to the measurements to be possible. The means of the *a*

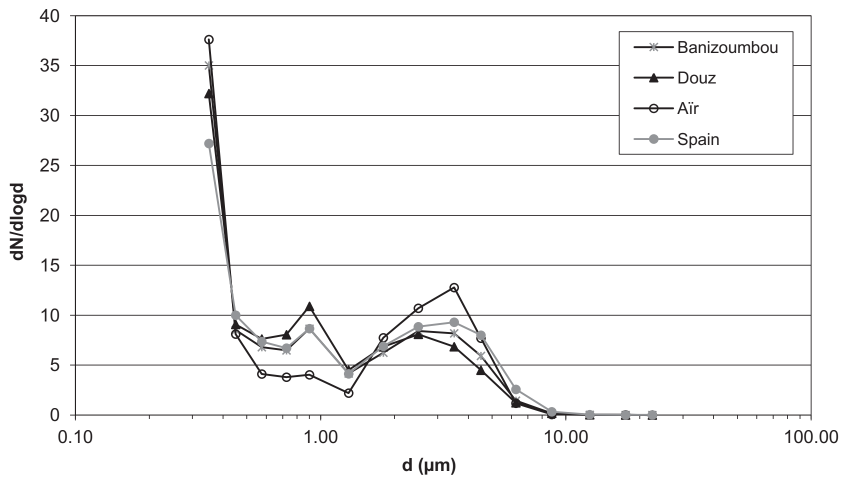

Fig. 15. Number size distributions of the aerosols generated in the GAMEL with different soil types. The results presented correspond to the measurements performed with the OPC during the first minute of the production period.

values obtained from three S-1-500 and four T-1-500 experiments are reported in Table 7.

It can be observed that, as compared to the Spanish case, the supply-limitation is more pronounced with the Tunisian soil. However, the dust production with these two soils does not decrease as fast as in the Banizoumbou case which, probably due to its smaller clay content, was characterized by larger *a* values (Table 4).

3.5.2. Concentration levels

Even if the four produced aerosols have similar size distributions, in the range of desert dust sizes, the resulting concentrations differed by almost three orders of magnitude. The Aïr soil releases a very small amount of aerosol particles whereas the Douz and Spain samples liberated much larger quantities of aerosol. In this series, the Banizoumbou soil constitutes an intermediate case. This is illustrated by the values of the mean of the total number suspended particles collected along the first minute of the production period for the repetition of the experiments performed with each soil sample (Table 8).

There is no clear relationship between the soils characteristics and the quantities of aerosols produced in the GAMEL. Gillette (1979) suggested that the vertical emission flux is low for sandy clay loam soils such as the Aïr one, but also for soils with a clay content greater than 35% such as the Spanish one. This observation seems to be confirmed by the measurements performed on the Aïr sample, but not by those performed on the Spain one. Very sandy soils, such as the Banizoumbou one, are supposed to have an intermediate production capacity as was observed in our experiments. Because no soil similar to the one of Douz was tested in the study of Gillette (1979), the comparison is not possible in this case.

3.5.3. Influence of source soil on elemental composition

Fig. 16 displays the Ca/Fe vs. Si/Al plot for all the aerosol filters collected and subsequently analyzed by XRF. The grouping of the points clearly shows that the composition of the dust depends on the composition of the soil sample from which it was produced.

Table 7
Values of the exponent *a* of Eq. (1) for each size class for Spain and Tunisian (Douz) experiments.

d (µm)	0.30–.40	0.50–0.65	0.80–1.0	1.6–2.0	3.0–4.0	5.0–7.5
S-1-500	0.146	0.186	0.174	0.164	0.206	0.219
T-1-500	0.203	0.321	0.279	0.419	0.583	0.330

Table 8
Total number of suspended particles generated in the GAMEL for the first minute of the production period. Four different soil types are tested. The standard deviations are calculated using all the experiments performed with each soil (30 experiments for Banizoumbou, 4 for Douz, 3 for Aïr, and 3 for Spain).

	Banizoumbou	Douz	Aïr	Spain
TSP (part./L)	1.707×10^6	7.991×10^7	1.759×10^5	8.565×10^6
RSD	31	11	19	55

It is not possible to relate the Si/Al values to the main mineralogy of the sample which is always dominated by quartz, because the dust composition mainly depends on the mineralogical composition of the clay fraction. Nevertheless, the Saharan/Sahelian aerosols (Niger-Banizoumbou and Niger-Aïr) has the lowest Ca/Fe ratio, whereas the one of the Spanish and Tunisian samples is much larger in good agreement with the large content of these two soils in calcium carbonate (Alfaro, 2008; Guieu et al., 2010, and Table 2).

3.5.4. Influence of the generation parameters on the dust production from other soils

Additional experiments were performed by increasing the soil quantities (results not shown here) with other soils than Niger and showed that the effect of modifying this generation condition is similar to the one observed in the Banizoumbou case. In other words, the aerosol concentration increased with the soil quantity but the size distribution remained constant.

For the less "productive" Aïr soil, we also performed an experiment in which we increased the shaking rate from 500 to 700 cycles/min. As a result, the TSP increased from 1.76×10^5 part./L to 2.52×10^6 part./L. In good agreement with what we already observed with the Niger soil, the aerosol size distribution was finer at 700 cycles/min than at 500: the relative proportions of the 0.3–0.4 µm size class and 2–3 µm size class were 40% (RSD = 3%) and 16 (RSD = 1%) respectively at 500 whereas they were 43% (RSD = 1%) and 14 (RSD = 1%) respectively at 700. In the same way, we also examined the elemental composition from the XRF analysis of one filter: Si/Al was found to be 2.87 at 700 cycles/min. to be compared to 3.10 obtained at 500 cycles/min. As previously observed for Niger soil, the Si/Al ratio tends to decrease when the shaking rate increases.

These results suggest that, for some samples, such as sandy clay loam, the cohesion forces maintaining fine particles within soil

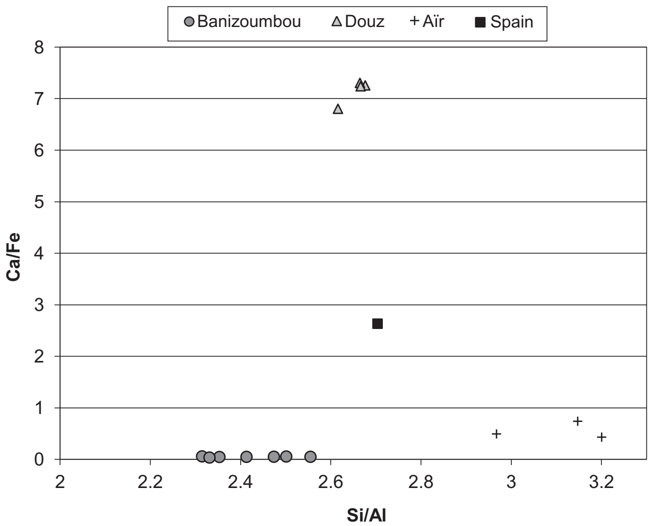

Fig. 16. Ca/Fe ratios as a function of Si/Al ratios measured by XRF on filters from GAMEL generations performed with different soil types.

aggregates can be strong enough to require more energy to be broken than what is provided in our standard generation conditions. This can be achieved by increasing the shaking intensity. Therefore, although our recommendations based on the study of the Niger sandy soil seem to be well adapted to a majority of soils they are certainly not universal. Our suggestion is to start any experiment with a new soil in the standard conditions (shaking 500 for 9 min from 1 g of soil) and, depending on the results of the size and composition analysis of this first experiment, adapt it if necessary.

4. Concluding remarks

We have designed an experimental system, called GAMEL, for generating non-mixed mineral aerosols from sieved natural source soil samples. We believe that, though simplified as compared to outdoor conditions, the shaking of these samples inside a closed vessel is able to generate aerosols which can be easily characterized as airborne suspended particles or collected for subsequent analysis. The first advantage of this generator is the very small amount of soil it requires to produce an aerosol, as compared to wind-tunnel or to several other generators such as the Southard Laboratory Dust Generator which works with hundreds of grams (Domingo et al., 2010).

Another advantage is that we could produce aerosols without any contamination, thanks to the efficient pre-filtering system, thus ensuring that the produced aerosol comes only from the used parent soil. When used with arid soils, the GAMEL produces a mineral dust aerosol with a coarse size mode centered around 4–5 μm in the mass distribution and a submicron population. These two modes not observed on the blanks are representative of the mineral dust emission in GAMEL. Moreover, numerous qualifying tests have been performed with the system. The performance of these tests ensures that the characteristics of the generated aerosol are well controlled and can be predicted for given generation conditions.

The repetition of 30 generation experiments performed in the same conditions (1 g of soil grains and a shaking frequency of 500 cycles/min) with the Banizoumbou (Niger) soil sample show that the produced concentrations, either expressed in terms of number or of mass can differ by as much as a factor of 5. However, when examined from the point of view of the size-distribution and of the compositional characteristics of the mineral aerosol, the repeatability of the experiments is adequate if some sources of variability inherent to the system are properly controlled. Among these, the decrease of the concentration and the evolution of the size distribution with time during the generation period have both been studied and quantified.

The impact of generation parameters such as soil quantity, shaking rate, and shaking duration, has been quantified, which allows us to define optimal "standard" conditions of use, and the way the aerosol properties would change should other generation conditions be used. In particular, we showed that because the properties of the aerosol evolve along the production period, the duration of the collection has an impact on the "bulk" aerosol collected on filters. Therefore, the shaking duration must be well controlled for obtained comparable aerosols. Adjusting the shaking rate between 300 and 600 cycles per minute is an efficient way of controlling the amount of produced aerosol but the properties are also sensitive to this generation parameter. In good agreement with the sandblasting theory (Alfaro et al., 1997), and field observations (Sow et al., 2009), creating more energetic particle collisions inside the GAMEL has an influence on the size distribution of the aerosol, by increasing the percentage of submicron particles. A new point is that this also has an impact on the aerosol composition. For instance, in the Banizoumbou (Niger) case, a "strong-shaking-conditions" aerosol is clearly distinguishable from a "medium-shaking-conditions" one, depending on whether the shaking rate is above or below 600 cycles/min, respectively. Increasing the quantity of parent soil introduced into the flask also increases the aerosol concentration (in a more moderate way than the shaking rate) but does not impact the size-distribution and composition of the generated aerosol. Modifying this parameter also appears as a convenient way of tuning the amount of material collected on filters. After the tests we performed, we recommend using 1 g of soil sample, a shaking rate of 500 cycles per minute, and a collection time of 9 min as standard conditions, but if necessary modifications can be brought to these conditions provided one is aware of the impact of these changes on the aerosol properties.

We also wanted to check if the size and compositional characteristics of the GAMEL-produced aerosol compared well to the aerosols studied on the field in Niger during some intense erosion events of the international AMMA campaign. The good agreement observed between the GAMEL-generated and the natural aerosols suggests that the laboratory generator is a reliable instrument for simulating the natural processes. Finally, the generation tests performed with soil samples differing completely from the Banizoumbou sandy soil show that, provided adequate generation conditions are chosen, the GAMEL is able to produce mineral dust from practically any variety of source soil possibly encountered in the arid and semi-arid areas of the World.

Acknowledgement

Under the name of the MIPAD project, this study has benefited from a grant of the LEFE/CHAT (CNRS-INSU) program.

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