

IMPACT OF A SAHARAN DUST OUTBREAK ON PM₁₀ GROUND LEVELS IN SOUTHEASTERN FRANCE

Nicolas MICHELOT^{1*}, Wilfried ENDLICHER², Pierre CARREGA¹,
Nicolas MARTIN¹, Olivier FAVEZ³, Marcel LANGNER⁴

¹ University of Nice Sophia-Antipolis – Geography Department, Management and Valorization of the Environment team – UMR 7300 ESPACE CNRS

98 Bd Herriot, 06204 Nice Cedex 3 – France

nmichelot@gmx.fr / pierre.carrega@unice.fr / nicolas.martin@unice.fr

² Humboldt-Universität zu Berlin – Geography Department, Climatology section

Unter den Linden 6, 10099 Berlin – Germany

wilfried.endlicher@geo.hu-berlin.de

³ National Institute for Industrial Environment and Risks (INERIS)

Central Laboratory for Air Quality Monitoring (LCSQA)

60550 Verneuil-en-Halatte – France

olivier.favez@ineris.fr

⁴ Umweltbundesamt (UBA), the Federal Environment Agency

Wörlitzer Platz 1, 06844 Dessau-Roßlau – Germany

marcel.langner@uba.de

* Author to whom correspondence should be addressed; E-Mail: nmichelot@gmx.fr; Tel.: +33 (0) 6 64 36 05 13

× Current affiliation: French Ministry of the Ecology, Air Quality Office, 92055 Paris La Défense

Abstract: Southeastern France is often subject to thermal breezes and inversions that are partly responsible for the dispersion behavior of air pollutants in this region. Generally, the coastal urban zone is the main contributor to PM₁₀ emissions. However, a southerly wind, commonly known as Sirocco, occasionally generates dust advections from the Sahara desert, resulting in poor air quality in the study area. This work demonstrates the quick rise of PM₁₀ levels on the French coastline under the influence of such a weather outbreak. Measurements were performed during a Saharan dust episode which occurred end of April 2013 and caused the tripling of PM₁₀ daily averages at the regional scale in about 24 hours. In Vence, located in the Alpes-Maritimes department, the highest daily average was 7 times greater during the peak than before the dust outbreak. In Venaco (Corsica) off-line chemical characterizations for filter samples show that about 50% of the PM₁₀ mass was composed of terrigenous dust, which confirms that they played a central role in the degradation of air quality and in the exceeding of the EU daily limit value of 50 µg/m³ at a regional scale.

Key-words: Saharan dust, PM₁₀, Sirocco, air pollution, Southern France.

Résumé :

Impact d'une advection de poussières sahariennes sur les niveaux de PM₁₀ dans le Sud-Est de la France

Dans le Sud-Est de la France, les Alpes-Maritimes connaissent une ventilation essentiellement assurée par le jeu des brises et une fréquence élevée d'inversions thermiques. Ces topoclimats conditionnent la dispersion des polluants atmosphériques, en majorité émis au sein de l'aire urbaine côtière. Au-delà de cette singularité climatique, la ventilation occasionnelle d'échelle dynamique, si lorsqu'elle est d'origine continentale ou d'altitude s'avère favorable à la dispersion des polluants (mistral, foehn par exemple), peut également devenir néfaste à la qualité de l'air. Dans ce dernier cas, le flux évoqué est communément appelé Sirocco. Il est facilement identifiable car il colore l'atmosphère de teintes orangées, et est orienté principalement au sud, sud-est.

Au printemps 2013, une advection d'air subtropical a transporté des poussières sahariennes au-dessus du bassin occidental de la mer Méditerranée, allant d'Afrique du Nord à l'Italie en passant par l'Espagne et la France. C'est l'impact de ce vent sur les niveaux de PM₁₀ dans les Alpes-Maritimes et en Corse qui est étudié dans cet article.

Dans le cadre d'une thèse de doctorat, un poste de mesure de particules (TEOM-FDMS prêté par l'Université Humboldt de Berlin) et des appareils de relevés météorologiques ont été installés durant plus de sept mois sur un terrain privé à Vence (commune située à 10 km au nord-ouest de Nice) à environ 300 mètres d'altitude en contrebas des Préalpes. Pour les faits étudiés ici, une attention particulière s'est portée durant la période du 29 avril au 1^{er} mai 2013 sur plusieurs paramètres météorologiques au moment où soufflait de façon discontinue le Sirocco. Afin de déterminer par spéciations chimiques l'origine des particules, le programme de caractérisation des particules (CARA) créé et géré par le Laboratoire central de surveillance de la qualité de l'air (LCSQA) a été mobilisé durant la thèse, mais aussi ultérieurement pour cette étude en retenant le poste de Venaco en Haute-Corse (sous la responsabilité de l'AASQA Qualitair Corse). Ce poste a été retenu d'une part parce qu'il disposait des seuls filtres disponibles dans l'aire d'étude durant cette fenêtre météorologique, et d'autre part parce qu'il était situé sur la trajectoire du vent et donc pouvant être considéré comme représentatif du phénomène de dégradation de la qualité de l'air dans la région.

Les résultats démontrent bien le rôle joué par le Sirocco qui a occasionné par son apport de poussières sahariennes une hausse des niveaux de PM₁₀ dans la zone étudiée. En effet, les mesures indiquent le triplement des moyennes journalières de PM₁₀ à l'échelle régionale en 24 heures de temps. A Vence, la moyenne quotidienne pendant le pic était 7 fois plus élevée qu'avant l'épisode. A Venaco, la caractérisation chimique des particules montre que près de 50 % de la masse des PM₁₀ était composée de poussières terrigènes, ce qui confirme qu'elles ont joué un rôle central dans la dégradation de la qualité de l'air à une échelle régionale et pour le dépassement de la valeur limite quotidienne de 50 µg/m³ fixée par l'Union européenne.

Enfin, l'originalité de cet article consiste à mettre les résultats en perspective avec l'attente réglementaire visée ci-dessus. En effet, la directive 2008/50/CE dispose que les épisodes d'origine naturelle peuvent être soustraits du nombre annuel de dépassements de la valeur limite quotidienne (50 µg/m³) à ne pas dépasser plus de 35 fois par an (ce qui peut éviter d'être confronté à un contentieux). Aussi, à court terme, vis-à-vis de la gestion des pics de pollution, l'identification de l'origine des particules (en l'occurrence majoritairement des poussières sahariennes) permet d'écarter les coupables idéaux (transport routier et industries par exemple) et de cibler des actions adaptées à la nature de la pollution rencontrée. Cela dit, les autorités locales ne devraient pas non plus négliger d'autres leviers de réduction pour les principales sources d'émission locales et pérennes, même s'il n'y a rien à faire à propos de la contribution majeure du moment, en particulier pour protéger les populations sensibles.

Mots-clés : poussières sahariennes, PM₁₀, Sirocco, pollution de l'air, Sud-Est de la France.

Introduction

The occurrence and impact of Saharan dust outbreaks over the whole Mediterranean Basin have been widely studied in recent years (Goudie and Middleton, 2001; Escudero *et al.*, 2005, 2007; Engelstaedter *et al.*, 2006; Querol *et al.*, 2009; Gómez-Amo *et al.*, 2011; de la Paz *et al.*, 2013; Pey *et al.*, 2013). However, this type of brief and visible weather event still needs to be studied in southeastern France. Not only because these dust outbreaks are less recorded than those which are more severe in the Iberian Peninsula, Italy or the eastern part of the Mediterranean Basin, but also because of their contribution to the temporary degradation of air quality, especially when PM₁₀ concentrations are not compliant with EU limit values (Directive 2008/50/CE on ambient air quality and clean air for Europe includes special provisions for exceedences due to natural sources).

The department of the Alpes-Maritimes (Southeastern France) is characterized by a Mediterranean climate, modified by its complex terrain, and the region is usually under the influence of breezes and temperature inversions due to the radiative weather. The coastal urbanization area (about 1,000,000 inhabitants) is the main contributor to particulate matters and other pollutants emissions. Air pollution is not distributed equally in time and space. Topoclimates within this mountainous coastal area (Carrega, 1994, 1989; Martin, 2008; Carrega *et al.*, 2010; Michelot and Carrega, 2012a, 2012b) recycle or block air pollutants in the lower layers. Sometimes, synoptic conditions clean the air when the flow comes from the continent or altitude (Mistral or Foehn winds) as evidenced by fieldwork of Michelot *et al.*

(2014), or degrade the air quality by transporting aerosols from the Mediterranean Sea (sea-salts) and North-Africa (Saharan dust).

In the framework of a thesis research (Michelot, 2014), a weather and air quality monitoring campaign was implemented in Vence (a city located at the front of the Préalpes mountains, 10 km northwest Nice) during 7.5 months, from 24 November 2012 to 7 July 2013. Its major goal was to understand the temporal behavior of particles under the influence of meteorology. An opportunity to dispose of filters for evaluating PM_{10} sources also came through the nationally-funded CARA program. Initiated late 2007, the CARA network aims at providing a better knowledge about the origins of ambient particulate matter, based on their chemical characterization at a dozen of French monitoring stations (Favez, 2012), as of March 2014.

Over this period, an interesting weather situation occurred from 29 April to 1 May, 2013. During these three days, air pollution was strongly influenced by a subtropical advection carrying Saharan dust across the west Mediterranean basin. This weather phenomenon is also known as Sirocco. It is a warm and dry, or damp, southerly wind crossing the Mediterranean Sea, giving the atmosphere an orange color. Dust deposition can be either dry or wet (gravitational sedimentation or rain-out). Each year, generally in spring, Sirocco causes red rains and red muds on the ground. In France, during a major event in February 2004, this deposit of lithometeors from the Sahara was estimated to 2 million tons on an area ranging from Nantes to Besançon (Masson *et al.*, 2005).

This paper has two aims: the first objective is to study the role of the above-mentioned weather phenomenon on air quality; in other words, to demonstrate the temporal variability of the PM_{10} concentrations, at the Vence-Gaudissard station and others stations in the surrounding area under the purview of Air PACA (the French accredited associations for air quality monitoring: AASQA). The second objective is to characterize the chemical species in order to estimate the contribution of this dust outbreak to the PM_{10} mass measured at Venaco, a station located on the island of Corsica (AASQA: Qualit' Air Corse), the only location that provided its filters samples, and consequently was used to study the chemical composition of particles during this weather event. Furthermore, the implications of the dust outbreak on local air quality policy are discussed.

1. Description of the monitoring sites: Vence-Gaudissard and Venaco

Gaudissard is a residential neighborhood of Vence, a 20,000 inhabitants city located in a coastal suburban area situated 8 km north of the seafront, and 10 km northwest of Nice (county town of the Alpes-Maritimes). The habitat surrounding the city is scattered, composed of individual houses. Beyond the medieval walls, the city is made up of urban collective and individual mixed habitats. The rest of the town is semi-urbanized. The city is built 300 meters above the sea level at the foot of the Préalpes, where the closest summits reach heights of 600 and 800 meters. The topography of the measurement site is characterized by hills and small plateaus, interspersed with small, steep, wooded valleys ranging from 50 to 100 meters deep (figure 1, up). According to Carrega (1994), we know that thermal inversions can be very strong in these places. Northward, the terrain rises rapidly toward the Préalpes, while to the south the relief softens and decreases in the direction of the coastal plain.

The Gaudissard station (PM_{10} and weather) was positioned on a private property on the plateau of Sine-Gaudissard located approximately 1.5 km southwest of downtown Vence. The area is close to the Gaudissard wood that goes down the slopes of the adjacent small valleys. The immediate environment of the measurements stations is partially covered by an olive grove. The field is characterized by broad terraces gently sloping to the west.

Venaco (figure 1, down), located on Corsica Island, is a hinterland village of Haute-Corse department, close to Corte (main town nearby, 7,000 inhabitants), 30 km away from the Tyrrhenian Sea. In a mountainous environment, the station is situated on a ridge at an elevation of 652 meters in front of the “Monte Cardo” (2,453 meters) inside the Regional Natural Park of Corsica. The ridge delimitates two abrupt slopes, oriented northeast-southwest. This rural station is operated by the AASQA Qualit'Air Corse.

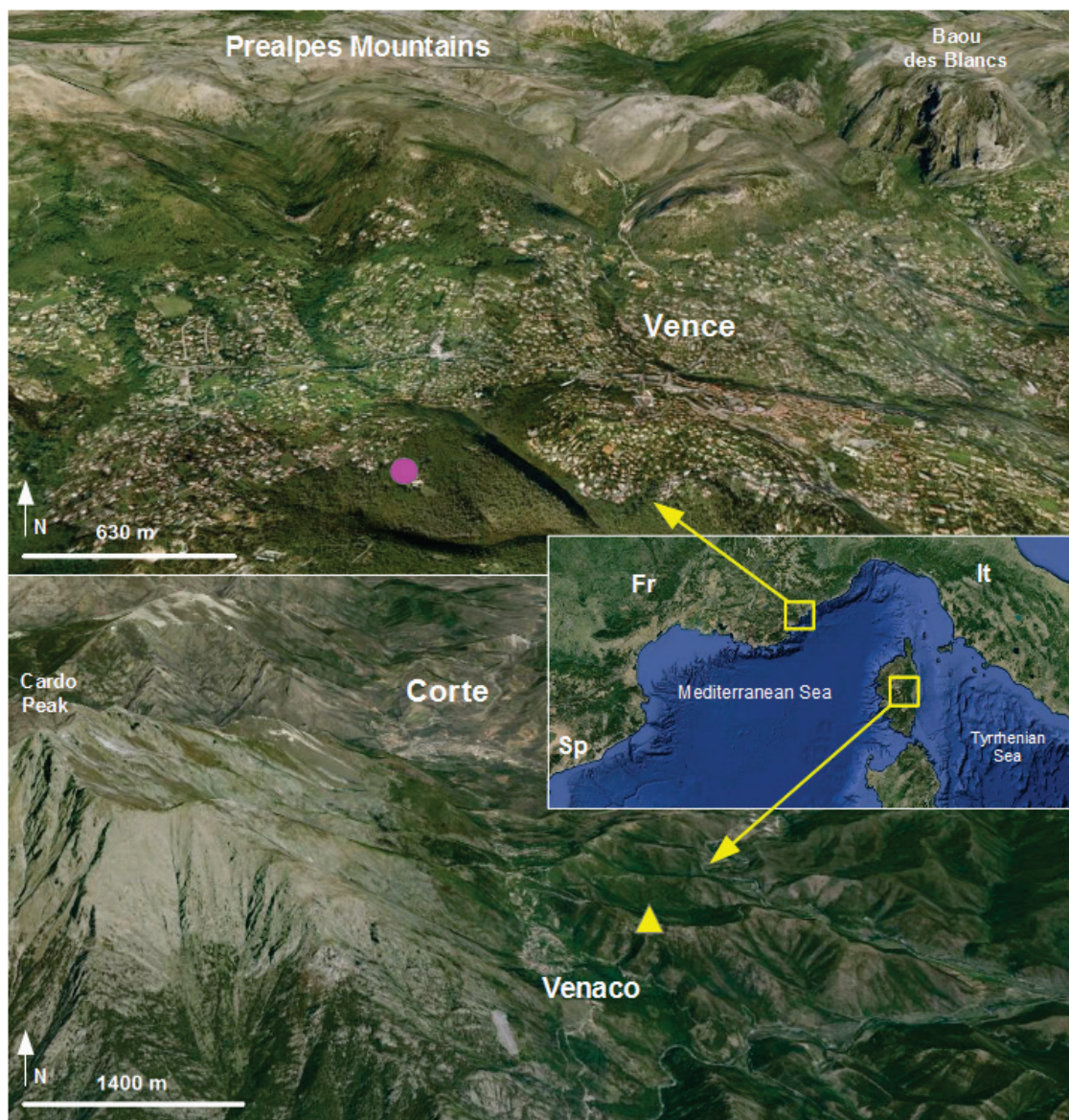


Figure 1. Localizations of measurements stations: Vence-Gaudissard (pink dot, up), Alpes-Maritimes department; Venaco (yellow triangle, down), Haute-Corse department. Base maps: Google Earth.

2. Materials and methods

2.1. Field instruments

At the Vence site, a van converted into a scientific laboratory truck (lent by Humboldt-Universität zu Berlin, Geography Department) was set up with a weather station and an automatic on-line PM₁₀ analyzer TEOM-FDMS (a TEOM 1400 coupled with a FDMS 8500; TEOM: Tapered Element Oscillating Microbalance / FDMS: Filter Dynamics Measurement System).

The TEOM-FDMS unit is a device widely used among the AASQA network for regulatory measurements in France. It continuously and automatically measures the concentration (mass) of suspended particles in the air. The FDMS module has been used in France from 2007 onward to account for semi-volatile material while eliminating the water present on the particles (Wilson *et al.*, 2006). It is equipped with a Nafion[®] dryer membrane.

The meteorological equipment of the van was used to measure several meteorological parameters, which were recorded every minute: wet and dry temperatures (at heights of 0.7 and 2 meters, THIES Clima Pt100 ventilated), wind speed and direction at a height of 6 m and irradiance at the roof of the van. Relative humidity was calculated from wet and dry temperatures.

Some additional parameters were acquired from two Davis weather stations (Monitor2, Vantage) installed during the whole measurement campaign on the same field, a couple of meters away. They measured:

- precipitations (not available in the van);
- atmospheric pressure;
- temperature, humidity.

Rainfalls were recorded with a rain gauge. Meta-data were also noted (such as types of weather, clouds, *etc.*).

At Venaco, Qualit' AIR Corse used a conventional on-line automatic system for monitoring PM₁₀ (TEOM-FDMS).

2.2. Filters sampling and chemical analyses

Venaco is a rural station chosen as representative of regional background conditions. At this site, filter samples have been collected on an occasional basis (in peak or original situations such as the one under consideration), using a *Digitel* high volume sampler (DA80 type). Chemical analyses performed on these sampled gave the contributions of the major chemical species and the origin (sources) of the PM₁₀. The synoptic air mass being the same between Corsica and the continent, it is highly probable that the contribution of the Saharan dust to the PM₁₀ mass on the French Riviera should be comparable to that in Corsica. This is why Venaco served as a reference.

As proposed by the dedicated European guidance (2011a), off-line chemical characterization is used here to confirm the main origin (Saharan dust) of the studied episode. This approach has already been used within the framework of the CARA program for natural contributions such as volcanic emissions (Colette *et al.*, 2011) and sea spray (Bhugwant *et al.*, 2013). Results are expressed here through “chemical mass closures” which consist in approaching the total mass of aerosols, measured by TEOM-FDMS, as the sum of its analyzed chemical components (Guinot *et al.*, 2007).

Quartz fiber filters (Pall, Qat-Up) were chosen for their low blank levels and their suitability for different types of chemical analysis to be performed. PM₁₀ were collected continuously on 150-mm diameter pre-fired filters at a flowrate of ± 100 l/h. Filters are burnt in an oven for 24 hours at 500°C. The filter sampling interval was 24h. A total of 6 filter samples were collected during this dust outbreak.

2.2.1. Chemical species

In the present work, the classification of species follows that of most current off-line studies focusing on PM chemical composition (Puteaud *et al.*, 2010), where a limited number

of major species form the majority of the particle mass. These species are divided into carbonaceous fractions (elemental carbon and organic matter), secondary inorganic species (nitrate, sulphate and ammonium), sea-salt and mineral dust. They are obtained here from the chemical analysis of:

- water-soluble ions: NO₃⁻, SO₄²⁻, Cl⁻, NH₄⁺, Na⁺, K⁺, Mg²⁺, Ca²⁺;
- metal species: Ti, Pb, Ni, Zn, Ba, Sr, Al, Fe, Ca;
- organic carbon (OC) and elemental carbon (EC).

The determination of these major species enables one to access basic informations about the contribution of natural causes and anthropogenic activity to the mass of PM₁₀ recorded.

Carbonaceous matter: Elemental Carbon (EC) and Organic Matter (OM)

EC is similar to pure graphite, a primary compound. It is exclusively emitted during a combustion process. Organic Matter (OM) is formed by a wide diversity of compounds, difficult to analyze and comes from many sources. In our study, only the organic carbon (OC) is measured (thermo-optically, as described below). The estimation of OM from OC requires the use of a conversion factor, which accounts for non-C atoms present in the OM. The more organic aerosol is oxidized, the higher this factor is. Primary emissions from road traffic produce poorly-oxidized organic aerosol (corresponding to a conversion factor of about 1.4) while secondary organic aerosol (from the conversion of VOC gases to particles) will be highly oxidized (factor up to 2.2) (Aiken *et al.*, 2008). In our case, a ratio of 1.8 was adapted from Turpin and Lim (2001), which does not lead *a priori* to an overestimation of OM.

Mineral dust and sea-salts

Mineral dust can be of natural or anthropogenic origin if it is linked to the remobilization of natural dust by anthropogenic activity. For Saharan dust, it has been considered here that the dust is constituted mainly of clay minerals, quartz, oxides (SiO₂, Al₂O₃, Fe₂O₃, TiO₂), carbonates (CaCO₃, MgCO₃) and Na₂SO₄. As SiO₂ cannot be measured on quartz filters, it has been considered that SiO₂ = 2.5 x Al₂O₃ (Escudero *et al.*, 2011).

Sea-salt particles are commonly estimated based on the concentration of specific tracers, such as sodium and/or chloride. In the present case, a significant part of sodium is assumed to originate from mineral dust, so that the mass of sea-salt has been obtained from measurements of chloride (Cl⁻). Following recommendations of the related European guidance (2011b), the following equation has been used:

$$[\text{Sea salt}] = 1.8 \times [\text{Cl}^-]$$

Chloride concentrations have also been used to determine (non-)sea-salt contributions of sodium, calcium and magnesium (respectively ssNa⁺, ssCa²⁺, ssMg²⁺ for sea-salt sodium, sea-salt calcium and sea-salt magnesium, and nssNa⁺, nssCa²⁺ and nssMg²⁺ for non-sea-salt sodium, non-sea-salt calcium and non-sea-salt magnesium), such as:

$$\begin{aligned} [\text{nssNa}^+] &= [\text{Na}^+] - [\text{ssNa}^+] = [\text{Na}^+] - 0.5558 \times [\text{Cl}^-] \\ [\text{nssCa}^{2+}] &= [\text{Ca}^{2+}] - [\text{ssCa}^{2+}] = [\text{Ca}^{2+}] - 0.0215 \times [\text{Cl}^-] \\ [\text{nssMg}^{2+}] &= [\text{Mg}^{2+}] - [\text{ssMg}^{2+}] = [\text{Mg}^{2+}] - 0.0669 \times [\text{Cl}^-] \end{aligned}$$

It should also be noted that ssCa²⁺ represents less than 5% of total Ca for each sample, so that calcium can be approximated as totally present within mineral dust particles. The concentration of the latter ones could then be estimated following:

$$[\text{Dust}] = 6,6 \times [\text{Al}] + 1,4 \times [\text{Fe}] + 1,7 \times [\text{Ti}] + 2,5 \times [\text{Ca}] + 3,5 \times [\text{nssMg}^{2+}] + 2,7 \times [\text{nssNa}^+]$$

Concentrations of mineral dust particles obtained from these calculations are assumed to be a reflexion of reality.

Secondary inorganic species

Secondary inorganic species which mainly correspond to ammonium sulphate ((NH₄)₂SO₄), and ammonium nitrate ((NH₄)NO₃), are formed in the atmosphere from gaseous precursors such as ammonia (NH₃), nitric acid (HNO₃) and sulfur dioxide (SO₂).

2.2.2. Analytical methods for major species

Anions and cations were analyzed by ion chromatography, after extraction of filter punches in ultra-pure water, according to NF EN ISO 10304 (anions) and NF EN ISO 14911 (cations); and EC/OC by thermo-optical method using the EUSAAR2 protocol (Cavalli *et al.*, 2010). The recommendations of the technical reports of the European Committee for Standardization (CEN/TC 264, respectively TR 16269 and TR 16243) were followed.

The analysis of metal elements was performed by optical spectroscopy and mass spectrometry (ICP-MS) in accordance to NF EN 149022.

2.2.3. PM₁₀ mass balance

The major species account for almost all of the total mass of the PM₁₀. In our study, this mass balance or “chemical mass closure” takes the form of the following equation:

$$[\text{PM}_{10}] \approx [\text{EC}] + [\text{OM}] + [\text{ammonium nitrate (NH}_4\text{)NO}_3] + [\text{ammonium sulfate (NH}_4\text{)}_2\text{SO}_4] + [\text{sea-salts}] + [\text{dust}] + [\text{undefined}]$$

The mass of the undetermined components of PM₁₀ is assumed to represent the sum of water adsorbed on the hydrophilic particles and other unknown major chemical components. In addition, it includes different measurement uncertainties (from PM₁₀ TEOM/FDMS measurement, filter sampling and chemical analyses) or underestimation of conversion factors such as calcium carbonate to mineral dust (Guinot *et al.*, 2007).

3. Synoptic evidence of dust outbreak and study of air quality

3.1. Meteorological conditions / PM₁₀ concentrations

During springtime, specific atmospheric disturbances passing through Southeastern France can be observed. In these situations air masses travel through the western Mediterranean Basin from one sea-shore to the other under the effect of an Iberian or Balearic Islands disturbance and a high pressure area over North Africa. This causes the advection of subtropical North African air masses loaded with Saharan dust to the Mediterranean French coasts. The sky turns orange, the atmosphere becomes turbid, and the weather becomes hot and damp, or dry: the Sirocco generally blows from south/southeast. Sometimes showers induce a rain-out. In the Maghreb, meteorological conditions promote the suspension of fine terrigenous materials into the atmosphere, and these particles are then dispersed throughout the Mediterranean Sea about 800 km away, when the transport takes place between Maghreb and the French coasts, or nearly 2,000 km away when dust takes the direction of the Middle-East.

On 29 and 30 April, the general situation promoted the presence of polar air at low altitudes on the northern part of France (figure 2, left). The contrast of air masses along a diagonal from the northwest of Spain to the Baltic Sea is evident (figure 2, right).

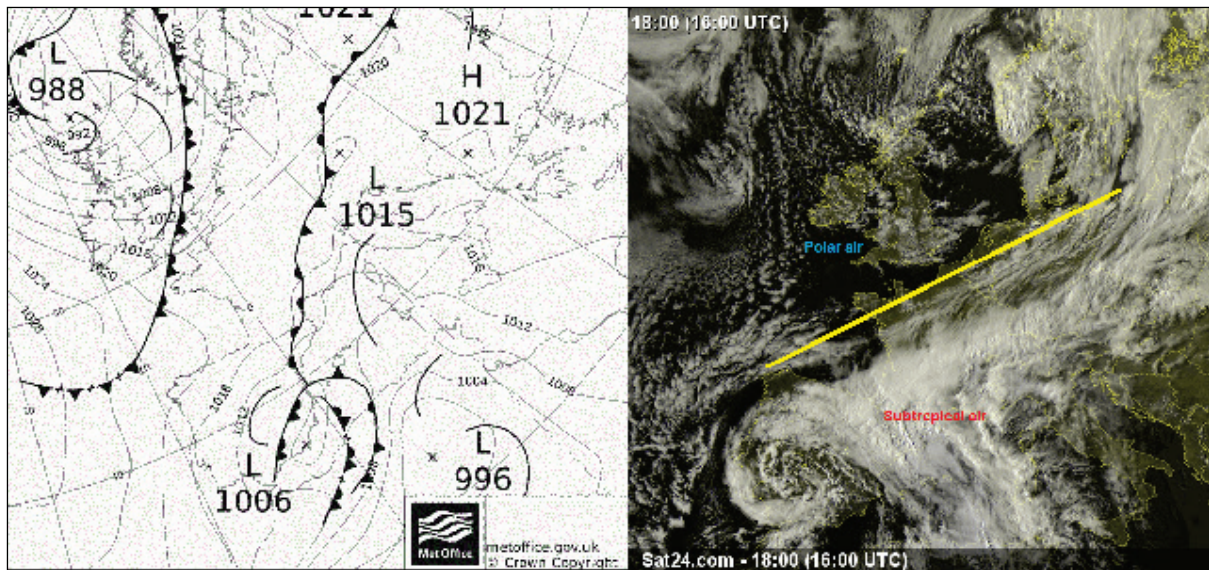


Figure 2. Left: Met Office surface analysis, 04/29/2013 at 00:00 am UTC (source: www.wetterzentrale.de). Right: satellite picture in the visible channel, 04/29/2013 at 04:00 pm UTC. The yellow stripe marks the back of the occlusion line of the Anglo-Scandinavian disturbance descended in France the same day; as well as the demarcation between the polar air mass coming from the north-west and subtropical air mass arriving from the southeast (source: www.sat24.com).

Under a cyclonic system, supported by a cold-drop (cut off) over the Iberian Peninsula lasting a few days, warm air from Africa (figure 3) crossed the Mediterranean Sea on a southeast/northwest axis loading humidity above the sea, then the flux tilted on the surface to the east-northeast in the Gulf of Genoa, bringing dust and orange rains.

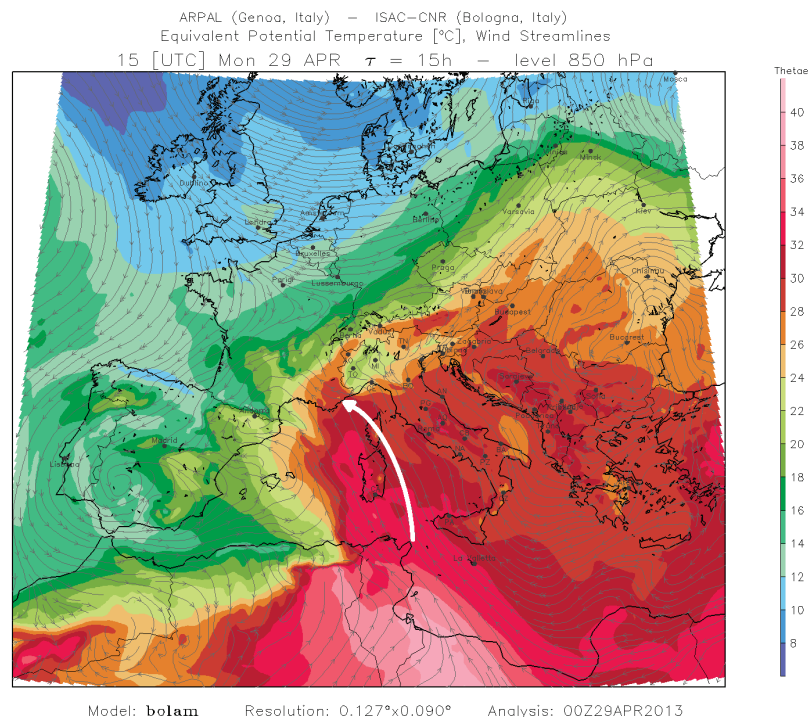


Figure 3. Bolam model forecasts for Theta-e and wind streamlines at 850 hPa level, 04/29/2013 at 03:00 pm UTC. (Source: www.arpal.gov.it).

We used AERONET data to compute one-day back-trajectories starting from the Montesoro site, Cap Corse (figure 4). AERONET is a worldwide network of sun photometers

that measures the optical properties of aerosols (Holben et al. 1998). Air masses clearly originated from the Sahara desert during this period. The air masses observed during this period showed a south-southeast influence over the Mediterranean Sea. The residence time of these air masses above ground level was 7 days (cross step time on figure 4), and a relatively slow flux could be seen.

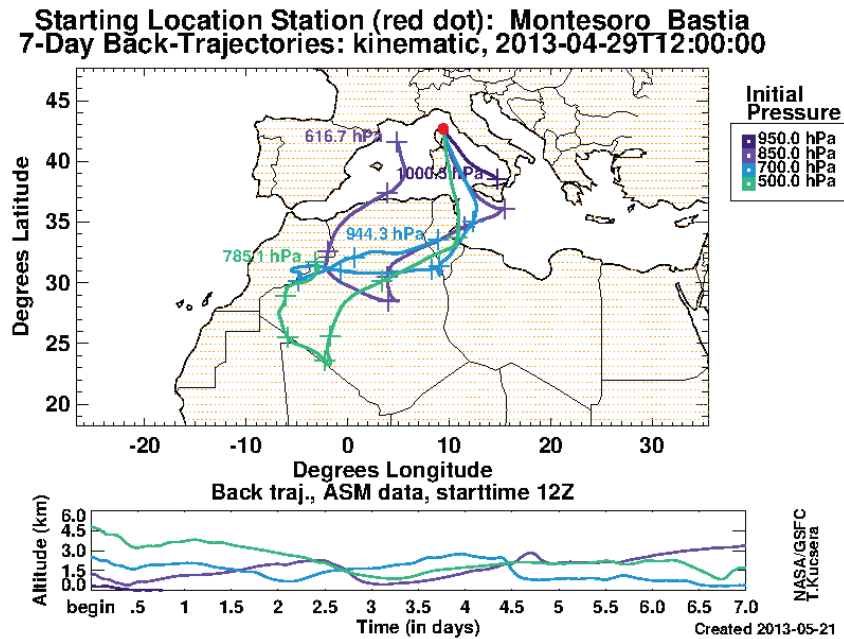


Figure 4. Back-trajectories, 04/29/2013 in Cap Corse (source: <http://aeronet.gsfc.nasa.gov>).

At the beginning of this dust outbreak, model outputs based on the chemistry-transport model CHIMERE (Bessagnet *et al.*, 2009) used in the forecasting system PREV'AIR (www.prevair.org) (Rouil *et al.*, 2009), display PM₁₀ concentrations at the European scale (figure 5), indicating high levels over the western Mediterranean basin because of Saharan dust. High particle concentrations could then be found in the central Mediterranean, the Tyrrhenian Sea and the Gulf of Genoa, which then moved on and affected mainly Italy, the Adriatic and finally on to the Balkans.

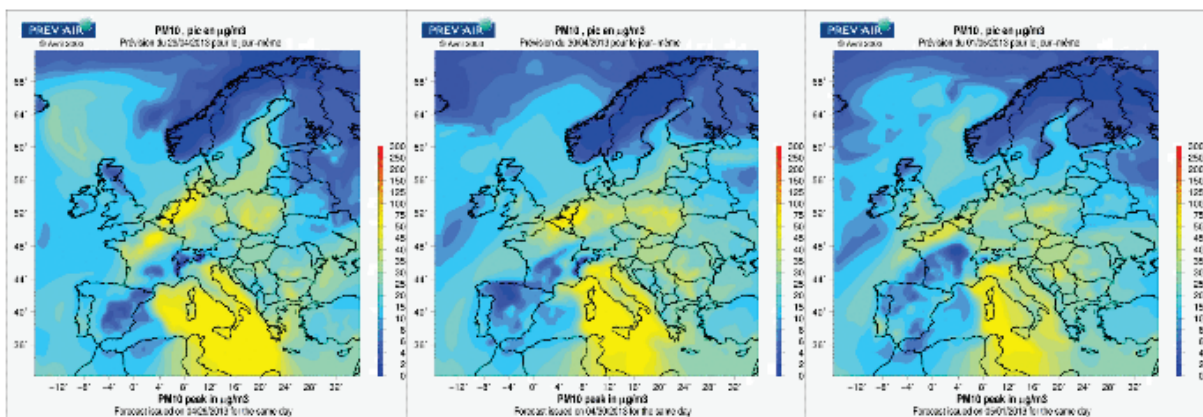


Figure 5. PREV'AIR forecasts of daily maximums PM₁₀, April, 29-30 and May 1st, 2013 (source: www.prevair.org).

The southeasterly flux (main stream) and the influence of the Gulf of Genoa caused the dust to revolve from east to southeast, except on 1 May, when the flow came from the

southwest during the day or even northeast due to the flexion of the flux by the coastal mountains. Also, the eastern part of Corsica was more affected by the orographic effect, where the surface stream was blocked by southeast slopes, and by the movement of the low pressure to the east.

In addition to the initial contribution of terrigenous materials, the dust that was not rained-out or deposited was recycled at the end of the outbreak. Indeed, for this last point, the atmospheric circulation in the Gulf of Genoa and the Tyrrhenian Sea (particular by the fact of the coastal relief curve) offered a Saharan dust stock that arrived more by the southeast. This explains the relatively high final levels of PM₁₀ in the Gulf of Genoa and the Tyrrhenian Sea.

3.2. Results from the measurements in Vence-Gaudissard

During the last decade of April 2013 (figure 6), after a brief anticyclonic thrust raising average levels of PM₁₀ on 23-24 and 25 April 2013, the situation returned to low pressure on 26 April. The flux change was followed by two days where showers reduced the daily PM₁₀ average from 20.5 to 6.6 $\mu\text{g}/\text{m}^3$ respectively on 27 and 28 April.

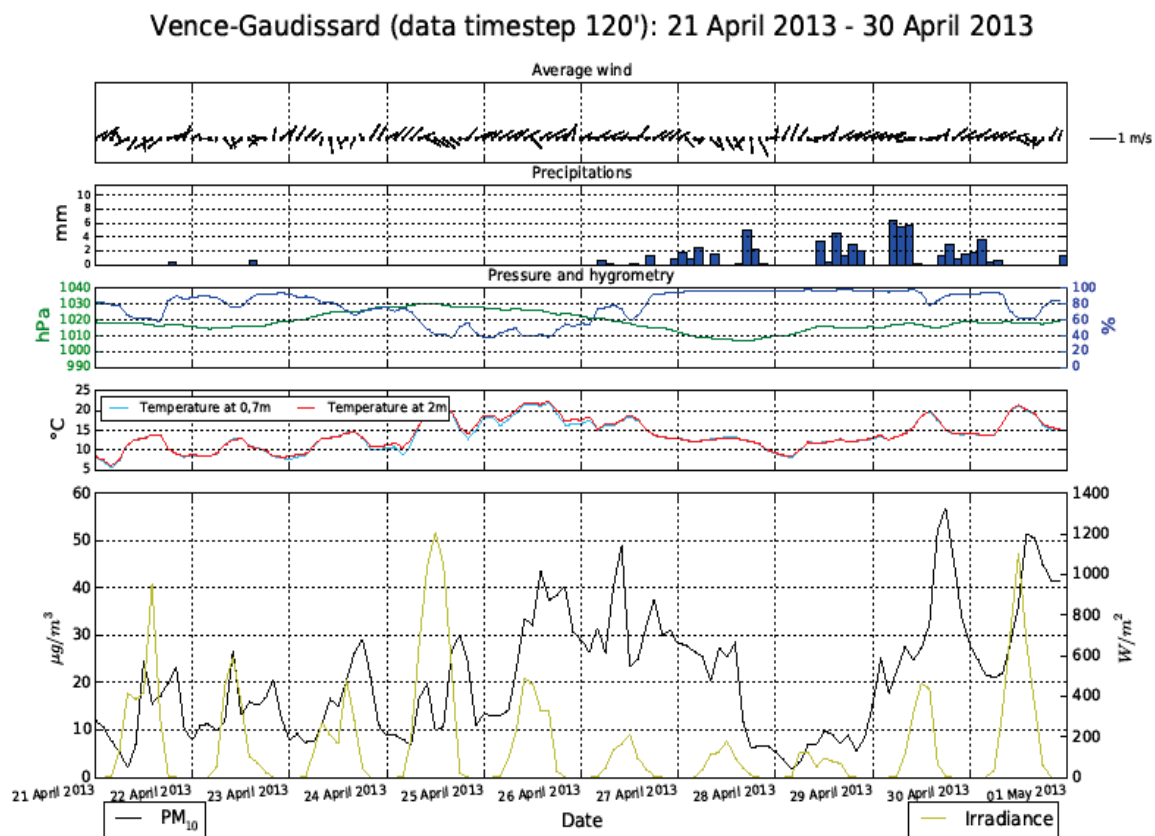


Figure 6. Bi-hourly PM₁₀ concentrations and meteorological parameters evolution during the last decade of April 2013, in Vence-Gaudissard (each 24h period starts from the intersection of the ordinate and abscissa to the right of the annotation of the day. For example, it reads the highest PM₁₀ value for this decade, *i.e.* 57 $\mu\text{g}/\text{m}^3$ on 04/29/2013 at 06:00 pm).

Weather conditions previously presented marked the beginning of PM₁₀ fluctuations on a hourly level on 29 and 30 April 2013, where PM₁₀ peaks were interspersed with Sirocco rains-out (figure 6, third vertical bloc from the right). Period of 28-30 April indicates the presence of a Sirocco as a diurnal southwesterly flux in Vence-Gaudissard on 29 April (tilted by site effect), then as a southeasterly to southwesterly wind the next day. Nocturnal flux of these two days was not a mountain breeze, but a low synoptic northeasterly wind.

At the beginning of May 2013, the episode continued because of favorable weather conditions supplying and recycling Saharan dust. PM₁₀ levels increased to a maximum of 68.6 µg/m³ at 12:00 pm on 2 May, 2013, when a daily average of 48.2 µg/m³ was measured. During the next day, average levels started to decline and this day marked the end of the outbreak. Simultaneously, high pressures and high radiative fluxes were present and took over the nycthemeral cycle of thermal breezes (note the superior bloc on figure 7: diurnal thermal southeasterly wind, nocturnal thermal northeasterly wind). It is a factor of air pollution in the Alpes-Maritimes (Michelot, 2014), and in this case interspersed with rains contributing to declining PM₁₀ concentrations until 6 May (daily average: 21.5 µg/m³) (figure 7).

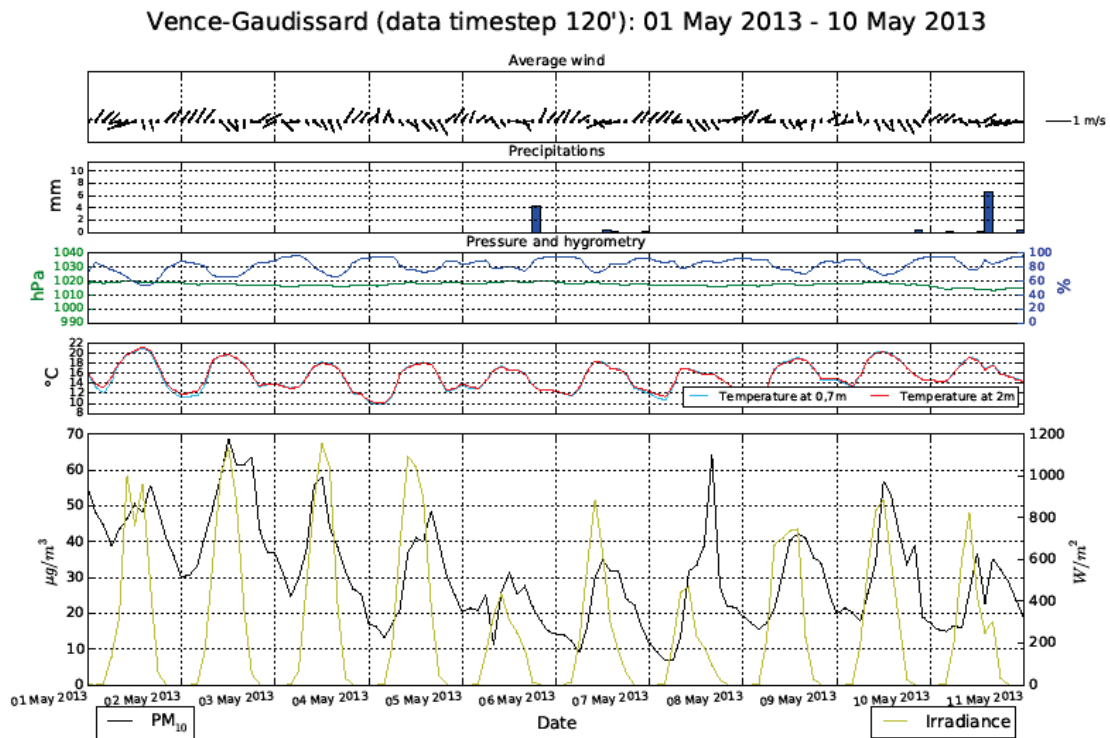


Figure 7. Bi-hourly PM₁₀ concentrations and meteorological parameters evolution during the first decade of May 2013, in Vence-Gaudissard.

The temporal variability of hourly PM₁₀ concentrations at Vence-Gaudissard resembles the short-term variability of PM₁₀ at nearby Air PACA stations (figure 8). Also, figure 8 indicates that the starting point of the dust outbreak was on 29 April for the whole region, and on 3 May the dust phenomenon ended due to a change of the flow regime. Table 1 summarizes the evolution of the PM₁₀ concentrations during the onset of the dust event.

Figure 8. Hourly average PM₁₀ concentrations evolution from April 28 to May 3, 2013, in some stations of Air PACA official network and Vence-Gaudissard (research station).

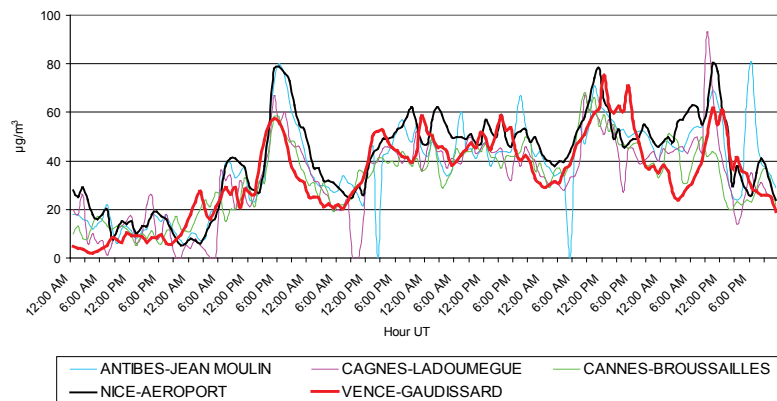
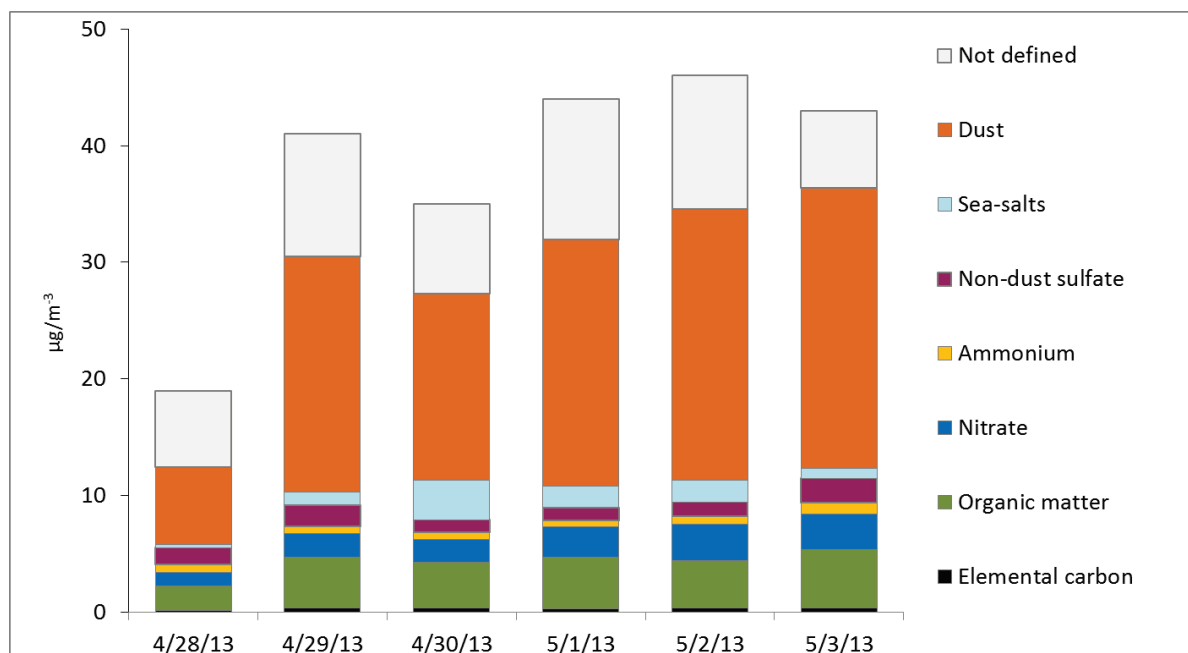


Table 1. PM₁₀ loading-up at the onset of the dust event in some stations of Air PACA network and research site.

	$\mu\text{g}/\text{m}^3$	4/29/2013	4/30/2013	5/1/2013
ANTIBES-JEAN MOULIN	Mean	12.2	23.8	50.7
	Min.	6	7	23
	Max.	18	39	80
CAGNES-LADOUMEGUE	Mean	13	16.7	43.8
	Min.	5	0	21
	Max.	26	36	67
CANNES-BROUSSAILLES	Mean	10	21.3	41.1
	Min.	5	11	21
	Max.	17	33	58
NICE-AEROPORT	Mean	12.7	23.5	54.5
	Min.	5	6	27
	Max.	19	41	79
VENCE-GAUDISSARD	Mean	8.5	23.6	41.6
	Min.	5.4	16	26.4
	Max.	13,0	29,2	57,4

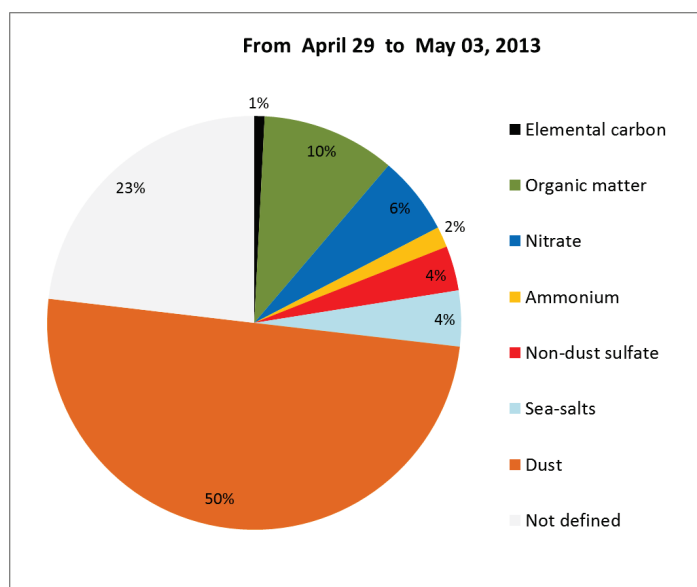
3.3. Major chemical constituents of PM₁₀

Figure 9 characterizes the different dust species in the PM₁₀. As already mentioned, the dust outbreak clearly started on 29 April 2013, where dust levels were three times higher than the day before and then represented half or more of the chemical species composing the PM₁₀ by mass. It is also interesting to note the significant presence of sea-salts during the event which was brought by a southeasterly stream.

**Figure 9.** Evolution of daily average concentrations and major species of PM₁₀ in Venaco, from April 28 to May 3 2013.

During the whole episode, *i.e.* from 29 April to 3 May 2013, we can consider that half of the PM₁₀ mass was composed of terrigenous dust (figure 10), from the Sahara desert in this case.

Figure 10. Chemical closure of the major chemical species of PM₁₀. Venaco from April 29 to May 03, 2013.



4. Discussion

According to a press advisory from Qualit'Air Corse, on Monday 29 April 2013, the information/recommendation procedure (JORF, 2014) was triggered by the prefect in the department of Corse du Sud (using data from the Ajaccio-Canetto urban station). Levels slid below the daily regulatory threshold ($50 \mu\text{g}/\text{m}^3$) at midday on 30 April. Because of the lasting unfavorable meteorological conditions and the concentration levels remaining very close to the regulatory threshold, the information/recommendation procedure was maintained as a precaution until Thursday, 2 May 2013 at 06:00 pm (local time). From a regulatory perspective, the peak pollution affected only the Southern Corsica department. Nevertheless, concentrations increased also in Bastia and Venaco (Northern Corsica department), but were still below the daily limit value for PM₁₀.

In the department of Alpes-Maritimes, daily averages of PM₁₀ measured at the research station did not exceed the daily threshold, with $46.5 \mu\text{g}/\text{m}^3$ on 1 May and $48.5 \mu\text{g}/\text{m}^3$ on 2 May 2013. However, one station of the Air PACA network near Vence, at the Nice airport exceeded the PM₁₀ daily limit value with 51 and $52 \mu\text{g}/\text{m}^3$ on the same days. These exceedances could have caused the regulatory triggering of an information/recommendation procedure (JORF, 2014). However, it was not triggered since the air pollution was of natural origin, the levels were very close to the daily regulatory threshold and the forecasts indicated a downtrend towards concentrations below the threshold the next day. Indeed, the constraint imposed by the triggering of an information/recommendations procedure was not justified and would have had no effect on lowering the PM₁₀ levels.

This is in the context of a recent change to the management of pollution peaks in France. Indeed, the interministerial order of March 26, 2014 (JORF, 2014) establishes a new management protocol (Michelot, 2015). It involves the implementation by the prefects, of locally relevant measures in various sectors, in very short timeframes. This national order is translated locally through prefectural orders. They memorize the actions to be triggered based on the characteristics of the pollution measured or predicted. For example, according to information/recommendation or alert (in this case prescriptive regulatory measures) levels of regulated pollutants, more health information, best practices or prohibitions are initiated in the sectors of transports (speed reduction on roads, anti-pollution controls, recommendation to use public transportations, carpooling, limiting the use of APU aircraft, banning or alternating vehicles, trucks circumvention of the city, etc.), agriculture (delaying of nitrogen spraying,

farm work and prohibition of burning-off, imposition of a rapid burial of effluents, *etc.*), industry (restricting activities that emit VOC, strengthening of pollution control, *etc.*) and residential/tertiary (limiting the use of open stove wood-burning, full compliance with the ban on burning of green waste in backyards, *etc.*). A technical instruction specifies the methods for implementation of this national order. Lastly, even though it is not possible to reduce the natural source of this type of pollution, some of these measures still prove useful by limiting some additional, anthropic, pollution and help avoid overexposure, especially for sensitive population, and especially if the episode would threaten to be long-term.

This event was not exceptional with respect to PM₁₀ levels. However these minor exceedances must be tracked and characterized precisely. Indeed, their notification makes it possible to remove them from the count of the daily limit exceedances ($50 \mu\text{g}/\text{m}^3$ not to be exceeded more than 35 days a year) reported to the European Commission for air quality monitoring (JOUE, 2008).

An extrapolation of the chemical results (*i.e.* 50% in dust Venaco chemical closure) to the coastline of the Alpes-Maritimes does not appear inappropriate. It is therefore reasonable to say that at the same time the French Riviera has received a major or significant chemical contribution of terrigenous dust from the Sahara, because the air mass was the same. However, this hypothesis must be seen as a rule of thumb, especially with respect to the rural site of Venaco, because the environment of the coastal PM stations of the Alpes-Maritimes is largely urbanized, with a primary, major, contribution of being road traffic (EC), and because of specific household practices in springtime (burning green wastes, especially in backyards), which would seem to indicate (in the absence of a proof) that the proportion of Saharan dust would be lessened by the other significant contributions in Vence, as OM in spring (Michelot, 2014).

Conclusion

The results of this fieldwork improve the knowledge of dust outbreaks in southeastern France. Findings are summarized as follows:

- The influence of a warm and wet disturbance flux from North-Africa to Southeastern France in springtime on the quick degradation of regional air quality has been illustrated.
- Long range dust transport caused the tripling of PM₁₀ daily averages at the regional scale in about 24 hours.
- During the dust outbreak (29 April to 3 May 2013), the maximum daily average concentrations of PM₁₀ on the two research sites were $39.5 \mu\text{g}/\text{m}^3$ in Vence-Gaudissard, and $42 \mu\text{g}/\text{m}^3$ in Venaco.
- In Vence-Gaudissard, the maximum PM₁₀ concentration on 29 April was 22 times higher than at the start of the dust outbreak ($2.6 \mu\text{g}/\text{m}^3$ at 06:00 am 28 April; $57.3 \mu\text{g}/\text{m}^3$ at 06:00 pm 29 April).
- In Vence-Gaudissard, the daily average was 7 times higher at the peak of the dust outbreak on 2 May, than on 28 April, while a pronounced intraday variability was observed, due to rains-out at the beginning of the event.
- In Venaco, a predominance of mineral chemical species characterized 50% of particle mass, consisting of terrigenous Saharan dust, and proved that these play a central role in the degradation of air quality and for the exceedance of the daily limit value ($50 \mu\text{g}/\text{m}^3$), both in Corsica and on the French Riviera.

Such information may have important implications for local policy makers who have to propose efficient abatement strategies aiming at the reduction of PM_{10/2.5} concentrations in the context of EU PM_{10/2.5} limit values, which have been transposed to the French code of the environment (JORF, 2010). Even if there are no possible actions to control air pollution from natural sources on a short-term level, it should be worthwhile to consider possible measures to reduce emissions from major anthropogenic sources in these situations, since the natural contribution is added to them, giving rise to concentration peaks.

The implications of our results for health issues are evident on the PM₁₀ peaks observed during our study, because they were mainly composed of dust. According to InVS (French Institute of Public Health Surveillance), this species may be associated with short-term mortality in southern Europe. Other studies suggest that these particles of natural origin could be dangerous to health (Perez and Künzli, 2001; Deroubaix *et al.*, 2013; Martigny and Chiappello, 2013), even more-so than those from other sources, such as road traffic. Finally, during a dust air pollution event, local authorities should not overlook other means of reduction for the main local emitting sources, even if there is nothing to do about the major contribution, especially to protect the sensitive population.

Acknowledgements: The authors would like to thank the Qualit'Air Corse team who collected filters during this weather event. We are also grateful to the National Institute for Industrial Environment and Risks (INERIS), in the framework of its missions in the Central Laboratory for Air Quality Monitoring (LCSQA) for the chemical analyses.

Author Contributions: The corresponding author conducted the literature search, developed the study design in agreement with Wilfried Endlicher, Marcel Langner and Nicolas Martin, and collected all relevant data under the supervision of Pierre Carrega (Director of Michelot's thesis). These authors have conveyed and installed measurement devices. Olivier Favez performed the chemical analysis (under the missions of LCSQA) and Nicolas Michelot predominantly analysed and interpreted all the data. The corresponding author wrote the manuscript. The authors discussed the findings and interpretations, critically reviewed the manuscript, gave hints for improvement and Wilfried Endlicher advised the whole process.

Conflicts of Interest: The authors declare no conflict of interest.

References

- AIKEN A.C., De CARLO P.F., KROLL J.H., *et al.*, 2008. O/C and OM/OC Ratios of Primary, Secondary, and Ambient Organic Aerosols with High-Resolution Time-of-Flight Aerosol Mass Spectrometry. *Environ. Sci. Technol.*, 12, 4478-4485.
- BESSAGNET B., MENUT L., CURCI G., HODZIC A., GUILLAUME B., LIOUSSE C., MOUKHTAR S., PUN B., SEIGNEUR C., SCHULZ M., 2009. Regional modeling of carbonaceous aerosols over Europe - Focus on Secondary Organic Aerosols. *Jour. Atmos. Chem.*, 61, 175-202.
- BHUGWANT C., BESSAFI M., FAVEZ O., CHIAPPINI L., SIEJA B., LEOZ-GARZIANDIA E., 2013. High contribution of sea salt aerosols on atmospheric particles measured at an urban tropical location in Reunion Island. *Jour. of Environ. Protec.* 4, 828-842.
- CARREGA P., MARTIN N., YOHIA C., 2010. Ozone et flux d'air dans l'arrière-pays niçois : mesures et modélisation à fine échelle spatiale durant un épisode estival (le 10 août 2006). *Pollution atmosphérique*, 207, 297-313.

- CARREGA P., 1994. Topoclimatologie et habitat. (Thèse d'Etat) *Revue d'Analyse Spatiale*, 35-36, 408 p.
- CARREGA P., 1989. Vent et échelles de contraintes géographiques : exemples en région niçoise. *Publications de l'A.I.C.*, 2, 83-88.
- CAVALLI F., VIANA M., YTTRI K.E., GENBERG J., PUTAUD J.-P., 2010. Toward a standardised thermal-optical protocol for measuring atmospheric organic and elemental carbon: the EUSAAR protocol. *Atmos. Meas. Tech.*, 3, 79-89, doi:10.5194/amt-3-79-2010.
- COLETTE A., FAVEZ O., MELEUX F., CHIAPPINI L., HAEFFELIN M., MORILLE Y., MALHERBE L., PAPIN A., Bessagnet B., MENUT L., LEOZ E., ROUÏL L., 2011. Assessing in near real-time the impact of the April 2010 Eyjafjallajökull ash plume on air quality. *Atmos. Environ.*, 45, 1217-1221.
- De la PAZ D., VEDRENNE M., BORGE R., LUMBRERAS J., de ANDRÉS J.M., PÉREZ J., RODRÍGUEZ E., KARANASIOU A., MORENO T., BOLDO E., LINARES C., 2013. Modelling Saharan dust transport into the Mediterranean basin with CMAQ. *Atmos. Environ.*, 70, 337-350.
- DEROUBAIX A., MARTIGNY N., CHIAPELLO I., MARTICONERA B., 2013. Suitability of OMI to reflect mineral dust surface conditions: preliminary application for studying the link with meningitis epidemics in the Sahel. *Remote sensing. Env.*, 133, 116-127.
- ENGELSTAEDTER S., TEGEN I., WASHINGTON R., 2006. North African dust emissions and transport. *Earth-Science Reviews*, 79, 73-100.
- ESCUADERO M., STEIN A.F., DRAXLER R.R., QUEROL X., ALASTUEY A., CASTILLO S., AVILA A., 2011. Source apportionment for African dust outbreaks over the Western Mediterranean using the HYSPLIT model. *Atmos. Res.*, 99, 518-527.
- ESCUADERO M., QUEROL X., AVILA A., CUEVAS E., 2007. Origin of the exceedances of the European daily PM limit value in regional background areas of Spain. *Atmos. Environ.*, 41, 730-744.
- ESCUADERO M., CASTILLO S., QUEROL X., AVILA A., ALARCÓN M., VIANA M.M., ALASTUEY A., CUEVAS E., RODRIGUEZ S., 2005. Wet and dry African dust episodes over Eastern Spain. *Jour. of Geoph. Res.*, 110, D18S08, doi:10.1029/2004JD004731.
- European Commission, 2011a. *Establishing guidelines for demonstration and subtraction of exceedances attributable to natural sources under the Directive 2008/50/EC on ambient air quality and cleaner air for Europe. Sec_2011_0208.*
- European Commission, 2011b. *Establishing guidelines for determination contributions from the re-suspension of particulates following winter sanding or salting of roads under the Directive 2008/50/EC on ambient air quality and cleaner air for Europe. Sec_2011_0207.*
- FAVEZ O., 2012. *Synthèse des travaux 2012 du programme CARA. Rapport LCSQA, 26 p.*
- GÓMEZ-AMO J.L., PINTI V., DI IORIO T., DI SARRA A., MELONI D., BECAGLI S., BELLANTONE V., CACCIANI M., FUÀ D., PERRONE M.R., 2011. The June 2007 Saharan dust event in the central Mediterranean: Observations and radiative effects in marine, urban, and sub-urban environments. *Atmos. Environ.*, 45, 5385-5393.
- GOUDIE A.S., MIDDLETON N.J., 2001. Saharan dust storms: nature and consequences. *Earth-Science Reviews*, 56, 179-204.
- GUINOT B., CACHIER H., OIKONOMOU C., 2007. Geochemical perspectives from a new aerosol chemical mass closure. *Atmos. Chem. Phys.*, 7, 1657-1670.

HOLBEN B.N., ECK T.F., SLUTSKER I., TANRÉ D., BUIS J.P., SETZER A., VERMOTE E., REAGAN J.A., KAUFMAN Y.J., NAKAJIMA T., LAVENU F., JANKOWIAK I., SMIRNOV A., 1998. AERONET: A federated instrument network and data archive for aerosol characterization. *Remote sensing. Env.*, 66, 1-16.

JORF (Official journal of the French Republic), 2014. *Arrêté du 26 mars 2014 relatif au déclenchement des procédures préfectorales en cas d'épisodes de pollution de l'air ambiant. N° NOR: DEVRI400449A.*

JORF (Official journal of the French Republic), 2010. *Décret n°2010-1250 du 21 octobre 2010 relatif à la qualité de l'air.*

JOUE (Official journal of the European Union), 2008. *Directive 2008/50/CE du Parlement européen et du Conseil du 21 mai 2008 concernant la qualité de l'air ambiant et un air pur pour l'Europe.*

MARTIGNY N., CHIAPELLO I., 2013. Assessments for the impact of mineral dust on the meningitis incidence in West Africa. *Atmos. Environ.*, 70, 245-253.

MARTIN, N., 2008. *La pollution par l'ozone et la climatologie dans une espace méditerranéenne : les Alpes-Maritimes.* Thèse de doctorat, Université de Nice Sophia-Antipolis, 298 p.

MASSON O., POURCELOT L., GURRIARAN R., PAULAT P., 2005. *Impact radioécologique des retombées de poussières sahariennes.* Rapport IRSN, 37 p. + annexes.

MICHELOT N., 2015. Le nouveau dispositif français de gestion des pics de pollution : l'arrêté interministériel du 26 mars 2014 relatif au déclenchement des procédures préfectorales en cas d'épisodes de pollution de l'air ambiant. Numéro spécial *Pollution atmosphérique*, 20-24.

MICHELOT N., 2014. *L'influence des topoclimats sur la pollution de l'air aux particules dans le sud-ouest des Alpes-Maritimes.* Thèse de doctorat, Université de Nice Sophia-Antipolis, 416 p.

MICHELOT N., MARTIN N., CARREGA P., 2014. L'impact d'un changement soudain du type de masse d'air sur les concentrations de PM₁₀ au front des Préalpes de Vence. *Actes des Journées de Climatologie de Caen, Commission Climat et Société du CNFG*, 61-70.

MICHELOT N., CARREGA P., 2012a. Le rôle des inversions et des brises thermiques sur la variabilité temporelle des particules à Auribeau-sur-Siagne (Alpes-Maritimes) : description et interprétation des mesures fixes. *Actes des Journées de climatologie de Liège (Belgique), Commission Climat et Société du CNFG*, 21-30.

MICHELOT N., CARREGA P., 2012b. Le rôle des brises thermiques sur la variabilité spatio-temporelle des PM₁₀ en pays vençois (Alpes-Maritimes, France). *Actes du 25^{ème} Colloque de l'Association Internationale de Climatologie* (Grenoble, France), 535-540.

PEREZ L., KÜNZLI N., 2001. Saharan dust: no reason to exempt from science or policy. *Occup. Environ. Med.*, 68(6), 389-390.

PEY J., QUEROL X., ALASTUEY A., FORASTIERE F., STAFFOGIA M., 2013. African dust outbreaks over the Mediterranean Basin during 2001-2011: PM₁₀ concentrations, phenomenology and trends, and its relation with synoptic and mesoscale meteorology. *Atmos. Chem. Phys.*, 13, 1395-1410.

PUTEAUD J.-P., Van DINGENEN R., ALASTUEY A., *et al.*, 2010. An European aerosol phenomenology – 3: Physical and chemical characteristics of particulate matter from 60 rural, urban, and kerbside sites across Europe. *Atmos. Environ.*, 44, 1308-1320.

QUEROL X., PEYA J., PANDOLFI M., ALASTUEY A., CUSACK M., PEREZ N., MORENO T., 2009. African dust contributions to mean ambient PM₁₀ mass-levels across the Mediterranean Basin. *Atmos. Environ.*, 43, 4266-4277.

ROUÏL L., HONORÉ C., VAUTARD R., BEEKMANN M., BESSAGNET B., MALHERBE L., MELEUX F., DUFOUR A., ELICHEGARAY C., FLAUD J.-M., MENUT L., MARTIN D., PEUCH A., PEUCH V.-H., POISSON N., 2009. PREV'AIR. An operational forecasting and mapping system for air quality in Europe. *Bull. Amer. Met.*, 90 (1), 73-83.

TURPIN B.J., LIM H.J., 2001. Species contribution to PM_{2.5} mass concentrations: revisiting common assumptions for estimating organic mass. *Aerosol Sci. Technol.*, 35, 602-610.

WILSON W.E., GROVER B.D., LONG R.W., EATOUGH N.L., EATOUGH D.J., 2006. The measurement of fine particulate semivolatile material in urban aerosols, *J. Air Waste Manag. Assoc.*, 56, 207-215.