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Variability of mineral dust deposition in the western Mediterranean basin and South-East of France

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Abstract

Previous studies have provided some insight into the Saharan dust deposition at a few specific locations from observations over long time periods or intensive field campaigns. However, no assessment of the dust deposition temporal variability in connection with its regional spatial distribution has been achieved so far from network observations over more than one year. To investigate dust deposition dynamics at the regional scale, five automatic deposition collectors named CARAGA ("Collecteur Automatique de Retombées Atmosphériques insolubles à Grande Autonomie" in French) have been deployed in the western Mediterranean region during one to three years depending on the station. The sites include, from South to North, Lampedusa Isl., Mallorca Isl., Corsica Isl., Frioul Isl. and Le Casset (South of French Alps). Deposition measurements are performed on a common weekly period at the 5 sites. The mean Saharan dust deposition fluxes are higher close to the North African coasts and decrease following a South to North gradient, with values from 7.4 $\mathrm{g\,m^{-2}\,yr^{-1}}$ in Lampedusa (35°31′ N–12°37′ E) to 1 $\mathrm{g\,m^{-2}\,yr^{-1}}$ in Le Casset (44°59′ N–6°28′ E). The maximum deposition flux recorded is of 3.2 $\mathrm{g\,m^{-2}\,wk^{-1}}$ in Mallorca with only 2 other events showing more than 1 $\mathrm{g\,m^{-2}\,wk^{-1}}$ in Lampedusa, and a maximum of 0.5 $\mathrm{g\,m^{-2}\,wk^{-1}}$ in Corsica. The maximum value of 2.1 $\mathrm{g\,m^{-2}\,yr^{-1}}$ observed in Corsica in 2013 is much lower than existing records in the area over the 3 previous decades (11–14 $\mathrm{g\,m^{-2}\,yr^{-1}}$). From the 537 available samples, ninety eight major Saharan dust deposition events have been identified in the records between 2011 and 2013. Complementary observations provided by both satellite and air mass trajectories are used to identify the dust provenance areas and the transport pathways from the Sahara to the stations. Despite the large size of African dust plumes detected by satellites, more than eighty percent of the major dust deposition events are recorded at only one station, suggesting that the dust provenance, transport, and deposition processes (i.e. wet vs. dry) of dust are different and specific for the different deposition sites in the Mediterranean studied area. The results also show that wet deposition is the main way of deposition for mineral dust in the western Mediterranean

basin, but the contribution of dry deposition is far to be negligible, and contributes by 15 to 46 % to the major dust deposition events, depending on the sampling site.

1 Introduction

A reliable estimation of the dust content in the atmosphere and of its variability in space and time is needed to assess desert dust impacts on the Earth system. The most convenient tools to conduct this assessment are atmospheric dust models in which the atmospheric cycle of mineral dust is represented: dust emissions by wind erosion on arid and semiarid regions; atmospheric transport which is strongly controlled by the meteorological situations; and deposition of dust along their atmospheric path by dry or wet processes. A validation of the closure of the dust budget in atmospheric dust models needs to quantify precisely the amount of emitted dust, the atmospheric dust load, and the dry and wet deposited dust mass (Bergametti and Foret, 2014).

Significant progress has been made on dust emission modelling during the last two decades (Shao et al., 1993; Marticorena and Bergametti, 1995; Alfaro and Gomes, 2001; Shao, 2004; Marticorena, 2014) and on the dust source monitoring, especially by using satellite observations (Brooks and Legrand, 2000; Prospero et al., 2002; Washington et al., 2003; Schepanski et al., 2012). However, quantitative estimates of dust emissions in atmospheric models are still affected by large uncertainties (Zender et al., 2004; Textor et al., 2006; Huneeus et al., 2011), mainly because a direct and quantitative validation of soil dust emissions at a large scale remains not possible. The spatial distribution and temporal variability of atmospheric dust content has also been significantly improved through the development of aerosol products from spaceborne (e.g. Moulin et al., 1997; Torres et al., 2002; Shi and Cressie, 2007; Remer et al., 2008; Nabat et al., 2013), ground-based (Holben et al., 2001) and ship-borne (Smirnov et al., 2011) remote-sensing instruments. Presently, large available datasets of aerosol optical depth (AOD) have been widely and mostly used to validate dust atmospheric contents simulated by 3-D models at global (e.g. Chin et al., 2002; Ginoux and Torres, 2003; Huneeus et al., 2011) or regional scales (e.g. Cautenet et al., 2000).

Atmospheric dust particles are removed from the atmosphere by dry and wet depositions processes (Duce et al., 1991; Schulz et al., 2012). These two sinks, which counterbalance dust emissions on the global scale, control the atmospheric lifetime of dust particles (Bergametti and Fôret, 2014). However, rather little attention has been paid to dust deposition and few experiments were dedicated to test dust deposition schemes against in situ data. There is an urgent need for further research and measurements of dust deposition. Dust models are mainly validated against proxies for the atmospheric dust load, e.g. AOD, concentrations, dust vertical profiles, or combinations of these. However, at least two of the terms emission, dust load and deposition need to be documented to close the dust mass budget.

In this paper, we will present the results over a three-year period concerning atmospheric mass deposition measurements associated with Saharan dust transport over the Western Mediterranean Basin. The main goal of the sampling strategy is to provide data that can be used directly to test the dust mass budget in dust transport model. This long data set of Saharan dust deposition in the Western Mediterranean region can also be used to identify the transport patterns and the provenance of the dust. To do that, deposition measurements are coupled with satellite observations and air-mass trajectories.

2 Deposition measurement in the Mediterranean region

Atmospheric deposition fluxes have been measured in the Mediterranean region during the last fifty years. Table 1 gathers the direct deposition measurements performed both close to Saharan dust source areas and far away on both sides of the Mediterranean basin. Most of these deposition mass fluxes were obtained directly by weighting the deposited mass, the others being derived from aluminum deposition measurements assuming that this element contributes for about 7 to 8 % of the total dust mass (e.g. Guieu et al., 2002). Note that dust deposition can be also estimated indirectly based on the measurements of atmospheric aerosol concentrations and assuming dust dry deposition velocity and scavenging ratio (e.g. Le Bolloch et al., 1996).

Dust deposition measurements close to the North African dust sources are rare: dust deposition fluxes were only measured in Morocco, Tunisia and Libya. The deposition samples from Tunisia and Morocco (Guieu et al., 2010) were collected at sampling sites located along the Mediterranean coasts and indicate deposition fluxes ranging from 7 to 23 $\mathrm{g\,m^{-2}\,yr^{-1}}$. Dust deposition sampled in Libya (O'Hara et al., 2006), in 24 stations located from the north to the south of the country, provide significantly higher deposition fluxes, ranging from 40 $\mathrm{g\,m^{-2}\,yr^{-1}}$ to 420 $\mathrm{g\,m^{-2}\,yr^{-1}}$.

In the Mediterranean basin itself, dust deposition fluxes exhibit a large spatial variability ranging over more than one order of magnitude, from 2 $\mathrm{g\,m^{-2}\,yr^{-1}}$ to more than 27 $\mathrm{g\,m^{-2}\,yr^{-1}}$ in the western basin and from 4 $\mathrm{g\,m^{-2}\,yr^{-1}}$ to $\sim$ 100 $\mathrm{g\,m^{-2}\,yr^{-1}}$ in the eastern basin (Table 1), but also a strong inter-annual variability with fluxes ranging from 4 to 26 $\mathrm{g\,m^{-2}\,yr^{-1}}$ in Corsica on 11 years. It must be emphasized that most of the temporal variability at a given site is due to the occurrence of very intense but rare events with fluxes of several $\mathrm{g\,m^{-2}}$ that dominate the annual deposition flux when they occur (see for instance Bergametti et al., 1989; Loÿe-Pilot et al., 1996; Fiol et al., 2005).

These measurements provide a picture of the dust deposition in the Mediterranean basin, the heterogeneity of data does not allow to precisely investigate the spatial and temporal variabilities of the dust deposition fluxes. Indeed, most of these dust deposition measurements were performed either in one site during a long time period (e.g. datasets obtained in Corsica between 1984 and 1994 by Loÿe-Pilot and Martin, 1996, or in Crete between 1988 and 1994 by Pye, 1992 and Mattson and Nihlén, 1994), or on a network of several stations but during a much shorter time period. Moreover, the dust deposition fluxes were generally not measured with the same devices. This complicates the comparison of the data collected at the different sites.

Thus, a better understanding of the dust deposition in the Mediterranean basin, and especially of its spatial and temporal variability, requires measurements performed simultaneously on at several stations and with similar devices during a long-time period. With this objective, a new deposition sampler was developed for use at remote sites in full autonomy over several months (Laurent et al., 2015).

3 Experimental setup

3.1 The CARAGA deposition collector

In order to be able to maintain over a long time period a network to measure dust mass deposition fluxes, our strategy was to sample only the insoluble deposition. This simplifies a lot the design of the collector and the on-site operations. More than 80 % of the total Saharan dust deposition mass in the Mediterranean basin occurs in the form of insoluble material (Guerzoni et al., 1993; Avila et al., 2007). As a consequence, an automatic collector called CARAGA ("Collecteur Automatique de Retombées Atmosphériques insolubles à Grande Autonomie") was developed in collaboration between the ICARE Ingénierie Company and the Laboratoire Interuniversitaire des Systèmes Atmosphériques (Fig. 1). The CARAGA was designed to require limited human interventions and to be produced in small series to develop a standardized deposition network in remote areas (Laurent et al., 2015). A collecting open funnel (0.2 m^2), fixed on a steel structure at least 2.5 m above ground level (a. g. l.), allows the collection of both dry and wet deposition. The funnel vibrates and is rinsed automatically with pure water at the end of a sampling time period to drive down the collected particles on a filter. The online passive filtrating system allows collecting the particles on one of the filters which are installed in a motorized rotating unit carrying 25 filter holders. A solar panel insures the power supply of the CARAGA. A new filter is set automatically in the sampling position for each sampling step whose duration can be defined through an electronic interface. The funnel vibration and rinsing can be programmed to operate two times before every filter change. A complete description of the CARAGA collector can be found in Laurent et al. (2015).

3.2 Implementation of the deposition network

The deposition network in the western Mediterranean basin is constituted of five CARAGA instruments installed mainly on Mediterranean island coasts (Fig. 2). This network is thought to allow dust deposition sampling along a South-to-North transect, from near the North African coast to the South-East of France, covering about 1050 km from South to North and 870 km from West to East. The network is constituted of four island sites and one continental site. The first CARAGA was installed in October 2010 on the small elongated Pomègues Island which is part of Frioul Islands, a 2 km^2 archipelago. The Frioul's site is sited in the Gulf of Lion a few km of Marseille under the influence of natural and anthropogenic air masses. The sampler is at ~ 45 m in altitude on the small ridge that crosses the whole island, distant from the sea by less than 150 m on two opposite sides. This site, is also a site of the French Mediterranean Ocean Observing System for the Environment (MOOSE; http://www.moose-network.fr/) instrumented by the Mediterranean Institute of Oceanography. In July 2011, a second collector was installed at Le Casset in the southern French Alps, at ~ 1850 m in altitude. This site is one of the 13 sites of the network MERA (Observatoire National de Mesure et d'Evaluation en Zone Rurale de la Pollution Atmosphérique à Longue Distance, http://ce.mines-douai.fr/pages/observatoire-mera/) by belonging to EMEP (European Monitoring and Evaluation Program, EMEP; http://www.emep.int/). CARAGA samplers were afterward installed: (i) at 7 m in altitude and ~ 70 m from the sea shore at the Ses Salines lighthouse site on the southeastern tip of Mallorca Island (http://imedea.uib-csic.es/icg/Faro/home_en), a research site operated by the Institut Mediterrani d'Estudis Avançats (IMEDEA), in July 2011; and (ii) at 38 m in altitude and ~ 20 m of the border of the cliff of the northwestern coast of Lampedusa Island located in central Mediterranean, at the Global Atmosphere Watch (GAW) regional background station (http://www.lampedusa.enea.it/) operated by the Laboratory for Earth Observations and Analyses of the Agenzia Nazionale per le Nuove Tecnologie, l'Energia e lo Sviluppo Economico Sostenibile (ENEA; supersite of the Chemistry–Aerosol Mediterranean Experiment, ChArMEx, https://charmex.lsce.ipsl.fr/) in September 2011. The last CARAGA was installed at 75 m in altitude and ~ 300 m from the sea shore on the northern tip of Cape Corsica, headland of Corsica Island. It is close to the AERONET sunphotometer station downhill the French Navy semaphore of Ersa and a few km from the Ersa supersite of the ChArMEx program. At the stations, the precipitation amounts are available at least on a daily basis.

3.3 In-situ sampling and lab protocol

Atmospheric total (wet + dry) insoluble deposition is collected at the five sampling sites over the period 2011–2013. For this study, a one-week sampling duration was chosen as a compromise between maximizing the opportunity of sampling single dust plume deposition event and a long autonomy of the collector, i.e. up to 25 sampling weeks. The same sampling duration is programmed at the 5 sites, with an automatic change of the filter every Thursday at 12:00 UTC. 537 weekly atmospheric deposition samples were collected. At least one year of continuous deposition measurements is available from every station, the longest time series being from the Frioul site (January 2011 to December 2013). Depending on the site, the data recovery rate of weekly atmospheric deposition samples ranges from 77 to 91 % for the sampling period.

Atmospheric particulate concentrations measured in the Mediterranean suggest that more than 90 % of dust particles mass is in the coarse mode (diameter larger than 1.2 µm aerodynamic diameter) (Sciare et al., 2005). Thus, after flow speed tests, we choose to use the AA Millipore cellulose ester filter with a 0.8 µm porosity. To determine the mineral mass collected on the filters an ignition and weighing protocol was defined and is presented in Laurent et al. (2015). Briefly, it is based on the ignition of the sampled filters following a progressive increase in temperature up to 550 °C to remove organics and carbon by oxidation. Thus, the material remaining in the ashes is mineral matter. This mineral mass is then determined by a weighing procedure in controlled conditions. This protocol insures a good repeatability and reliability ($\pm 10^{-3}$ g by filter) in the determination of the mineral masses, limiting the loss of particles during filter handling (especially for filters highly loaded with dust particles). Moreover, if small insects, vegetal debris, pollens or other organic matters are collected on the filters, they are manually removed only if this manipulation does not affect the sample. If the removal of these elements could damage the sample, we leave them on the filter considering that samples ignition eliminates the organic matter (Laurent et al., 2015).

4 Results and discussion

4.1 Weekly atmospheric deposition in the western Mediterranean basin

The weekly fluxes of insoluble mineral deposition measured at the 5 sites are reported in Fig. 3. The highest weekly deposition fluxes are recorded at the two stations nearest to the Sahara desert: 3.2 and 2.7 $\mathrm{g\,m^{-2}}$ were measured at Mallorca and Lampedusa. The maximum deposition recorded for the stations located in the northern Mediterranean basin and South of France (Corsica, Frioul and Le Casset) is almost one order of magnitude lower (0.53, 0.34 and 0.17 $\mathrm{g\,m^{-2}}$, respectively). The same trend is observed when considering the deposition fluxes cumulated over a 1 year period without sampling discontinuities. In fact, the highest annual deposition fluxes are: 7.4 $\mathrm{g\,m^{-2}\,yr^{-1}}$ in Lampedusa for 2012; 5.8 $\mathrm{g\,m^{-2}\,yr^{-1}}$ in Mallorca for 2013; 2.1 $\mathrm{g\,m^{-2}\,yr^{-1}}$ in Corsica for 2013; 3.5 $\mathrm{g\,m^{-2}\,yr^{-1}}$ in Frioul for 2012; and 0.9 $\mathrm{g\,m^{-2}\,yr^{-1}}$ in Le Casset for 2012. Thus, a South-to-North decreasing gradient in the intensity of the mineral dust deposition is observed over the western Mediterranean basin with a flux about 2 to 8 times lower in the northern part of the basin and southern France than in the southern basin. Previous observations pointed out a gradient of dust content in the western Mediterranean atmosphere (see for example Barnaba and Gobbi, 2004).

In the late 1980s and 2000s, the mean annual deposition measured in Corsica by Loÿe-Pilot et al. (1986), Bergametti et al. (1989) and Loÿe-Pilot and Martin, (1996) and Ternon et al. (2010) were of 14.0, 11.0, 12.5 and 11.4 $\mathrm{g\,m^{-2}\,yr^{-1}}$, respectively. These deposition fluxes are significantly higher than the one we measured at the Corsica station from January to December 2013. These studies also point out a high inter-annual variability of the atmospheric deposition in the western Mediterranean basin. Investigating 11 years of atmospheric deposition in Corsica, Loÿe-Pilot and Martin (1996) reported deposition fluxes ranging from 4 to 26.2 $\mathrm{g\,m^{-2}\,yr^{-1}}$. These authors indicate that no deposition event greater than 1 $\mathrm{g\,m^{-2}}$ was recorded in the years for which the annual atmospheric deposition was low in Corsica, suggesting that the interannual variability of dust deposition in Corsica was driven by the annual occurrence of very intense Saharan dust deposition events. The contribution of such dust pulses to the annual atmospheric deposition flux in the western Mediterranean has been underlined many times in the literature. For example, Bergametti et al. (1989) reported that a deposition event of only 3 days occurring in March 1986 has contributed for more than 30 % of the annual atmospheric deposition measured in Corsica for elements such as Al or Si. Guerzoni et al. (1997) also mentioned that a single Saharan dust outbreak can account for 40–80 % of the annual deposition flux. In February 2004; Ternon et al. (2010) measured an extreme case of dust deposition in the Ligurian sea area (22 $\mathrm{g\,m^{-2}}$ for two weeks). This Saharan dust event represented almost 90 % of the Saharan inputs reported for 2004 at the Cap Ferrat site (Ternon et al., 2010).

The annual deposition flux we measured in Corsica corresponds to a low dust deposition year. This is also supported by the fact that none of the weekly deposition fluxes we measured exceeds 1 $\mathrm{g\,m^{-2}}$, the higher deposition flux measured in Corsica being 0.53 $\mathrm{g\,m^{-2}\,wk^{-1}}$. However, compared to previous deposition measured in Corsica, even the annual deposition fluxes measured at the south stations of the western Mediterranean basin (Lampedusa and Mallorca) are lower, suggesting that the years 2011, 2012 and 2013 probably correspond to a low atmospheric deposition period. This observation is in agreement with the analysis performed by Pey et al. (2013) from $\mathrm{PM_{10}}$ measurements performed in the Mediterranean region over the period 2001–2011 which concluded to a decrease of the contribution of Saharan dust events to the $\mathrm{PM_{10}}$ load. As mentioned by these authors and according to the study of Moulin et al. (1997), this decrease in the Saharan dust transport over the western Mediterranean Sea is probably due to the continuous decrease of the North Atlantic Oscillation (NAO) index in winter during the period 1990–2012 and in summer since 2006.

4.2 Selection of the most intense weekly deposition fluxes

The previous studies mentioned above indicated that the largest deposition events observed in the western Mediterranean basin are strongly associated with Saharan dust transports (Bergametti et al., 1989; Loÿe-Pilot and Martin, 1996; Guerzoni et al., 1999). We decided to give a particular attention to the most intense weekly deposition flux (MID) measured at each station and to verify if these high deposition events were linked to Saharan dust transport towards the Mediterranean Sea. Among the MID, the most intense Saharan dust deposition samples (MIDD) and the corresponding dust deposition events (DDE) will be discussed.

To select the MID, we first merged all the weekly samples collected at the different stations (537 samples) and then selected the weekly deposition greater than the last sextile of the dataset, i.e. the 16.67 % highest deposition values (i.e. 90 samples). This leads to a threshold weekly deposition flux of 9.3 $10^{-2}\,\mathrm{g\,m^{-2}\,wk^{-1}}$. The 90 highest deposition values were thus retained by using this threshold, independently of the station where they have been measured. However, as shown before, the intensity of the atmospheric deposition is higher at the stations located the nearest to the North African dust source areas. This means that even a relatively high deposition event occurring in the northern part of the basin may be excluded from the selection because its deposition flux is lower than the overall threshold value. To correct this bias, we added a second criterion which consists in also retaining, for a given station, all the samples for which the weekly deposition flux is greater than the geometric mean (m_g) plus the geometric standard deviation (σ_g) calculated for each station, i.e. $m_\mathrm{g} \times \sigma_\mathrm{g}$, statistical tests having shown that the deposition fluxes measured at each station fitted either a log-normal or a Gamma distribution (values below 10^{-4} g for a sample were assumed to be equal to this weighing detection limit). This second criterion allows the minimum selection of the upper sextile of a lognormal distribution. It leads to select 18 additional samples

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at the 2 stations where the average fluxes are the lowest: 4 more samples from Corsica (fluxes $> 0.050\,\mathrm{g\,m^{-2}\,wk^{-1}}$) and 14 from Le Casset ($> 0.036\,\mathrm{g\,m^{-2}\,wk^{-1}}$). Table 2 reports the 108 MID distribution by station, 54 % of them having been sampled in the southern stations of Lampedusa and Mallorca.

The MID events being selected, we verified whether these deposition events were or nor associated with air-masses originating from the Saharan desert. To point out the provenances and the main transport pathways of deposition events at each station, model air-mass trajectories and satellite aerosol observations were jointly analyzed.

Air-masses trajectories were computed using the HYSPLIT model (Draxler et al., 2003; https://ready.arl.noaa.gov/HYSPLIT.php) based on the operational Global Data Assimilation System (GDAS) 3 hly meteorological archive from the U.S. National Centers for Environmental Prediction (NCEP), and with the vertical velocity model. HYSPLIT is commonly used to trace the origin of the air masses transporting mineral dust (Escudero et al., 2011; Meloni et al., 2008) or atmospheric pollutants (Jorba et al., 2004; Pongkiatkul and Kim Oanh, 2007) studies. The statistical analysis of numerous backward trajectories from receptor sites has turned out to be a valuable tool to identify sources of atmospheric trace substances (Stohl, 1998; Scheifinger and Kaiser, 2007). Since the transport of Saharan air masses towards the western Mediterranean basin can occur at various altitudes in the troposphere (Bergametti et al.,1989; Martin et al., 1990; Hamonou et al., 1999; di Sarra et al., 2001; Israelevich et al., 2002; Meloni et al., 2008; di Iorio et al., 2009), 4 days backward air masses trajectories starting every day at 12:00 UTC were computed starting at 0, 500, 2000, 3000, 4000 and 5000 m a.g.l. for each of the five sampling stations. When the circulation of the model air masses was to slow over the western Mediterranean basin, the duration of the backward trajectories was extended to 6 days. Among the 108 MID samples, only one sample collected at Le Casset was not associated with at least one air mass trajectory having crossed Northern Africa during the sampling week.

To point out the origins of the dust deposition events for each station, we combined air-mass trajectories and the Aerosol Optical Depth (AOD) products provided by the NASA's Moderate Imaging Spectrometer (MODIS, http://modis.gsfc.nasa.gov/). To track, as best as possible, the location of the dust sources and to follow the dust plume, we used two products: the daily MODIS deep blue AOD at 550 nm over land and MODIS AOD at 550 nm over the oceanic surfaces (Levy et al., 2013). When MODIS AOD was unavailable due to cloud cover, we also examined the EUMETSAT MSG/SEVIRI dust false-colour composite product available from ICARE Geo Browse Interface (http://www.icare.univ-lille1.fr/), which gives the opportunity to check the transport of the dust plume every 15 min (e.g. Schepanski et al., 2007).

For each MID, we identified by using the Aerosol Optical Depth (AOD) from MODIS or MSG satellite where dust is coming from. For each of these dust provenance area, we used the HYSPLIT model to compute forward air mass trajectories in ensemble mode (i.e., multiple trajectories from the selected starting location by offsetting the meteorological data) or in matrix mode (i.e., starting from the borders of these identified dust sources). These forward air mass trajectories were computed starting at 00:00 and 12:00 UTC at an altitude of 500 m a.g.l. which can be considered as a common dust entrainment altitude (Meloni et al., 2008). We also checked for each forward trajectory the altitude of the air-mass and if precipitation occurred during transport. Figure 4 illustrates the different satellite observations and air mass trajectories used to identify the dust provenance area and transport pattern associated with dust deposition in the western Mediterranean basin. Following this method, 98 samples among the 107 MID present an air mass coming from North African areas with high AOD and reaching the stations. Hereafter, these 98 most intense Saharan dust deposition samples are called MIDD. For the remaining 9 cases, the absence of matching between high AOD observed from satellite images and air mass trajectories linking the dust provenance region to the sampling stations does not necessary mean that these cases are not cases of Saharan dust deposition. The MIDD accounts for 84, 78 and 73 % of the deposition in Lampedusa, Mallorca and Corsica, respectively, while it contributes for around 50 % in Frioul and Le Casset (Table 2).

4.3 Seasonality of the most intense dust deposition (MIDD)

To look at the seasonality of the MIDD occurrence, the fact that each month of the year has not been sampled with the same frequency at each sampling station has to be taken into account. We computed the number of weeks which have been sampled for each month and for each station. A weighting coefficient $a_{(M)}$ was computed for each station by dividing the number of sampled weeks for each month (M) by the total number of weeks of sampling at each station. $F_{(M)}$, the weighted number of MIDD occurrences for a given per month M was then computed as:

$$F_{(M)} = \frac{N_{(M)}}{a_{(M)}} \tag{1}$$

where $N_{(M)}$ is the number of MIDD during a given month.

Figure 5 reports the number of MIDD occurrence per month at each site. Most of the MIDD occurred during spring (March–June): 53 % in Le Casset, 49 % in Frioul, 81 % in Corsica, 38 % in Mallorca and 55 % in Lampedusa. A second maximum is observed in autumn in Mallorca and Lampedusa, and at the end of summer and early autumn in Frioul, Corsica and Le Casset. From their long-term data set in Corsica, Loÿe-Pilot and Martin (1996) also observed the most frequent and intense dust events in spring and autumn. According to Bergametti et al. (1989), the frequency of Saharan inputs in Corsica seems to be the highest during spring and summer, 80 % of the events being observed between March and October 1985. Ternon et al. (2010) observed high deposition in spring and summer with a maximum in June from their measurements performed in the Ligurian area between 2003 and 2007. Avila et al. (1997) showed that the occurrence of red rain episodes in northeastern Spain between 1983 and 1994 were higher in autumn and spring. The dust deposition seasonality can not be directly compared with other atmospheric observations. For instance in Lampedusa, the seasonality of atmospheric dust content and dust deposition are different. Maximum AOD indicate highest dust atmospheric content in summer in 2001–2005 (Meloni et al., 2004, 2008) while the crustal aerosol contribution to PM_{10} measurements performed between 2004 and 2010 does not show any evident seasonal pattern.

Our results point out that the stations of the network are not systematically concerned by dust deposition at the same period. To fully understand the variability of Saharan dust deposition in the western Mediterranean basin, several sampling sites are required to perform direct deposition measurements.

4.4 Origin of the dust deposition events

The number of stations operating when a MIDD was recorded is given in Table 3. This points out that 82 % of the MIDD were recorded when at least 4 stations were simultaneously operating. However, the 98 MIDD have been collected during 75 different weeks of sampling, and only 17 of them were measured simultaneously by more than 1 station during the same sampling week (12 at 2 stations, 4 at 3 stations, and 1 at 4 stations). 75 % of the cases when at least 2 stations recorded a MIDD associated the Mallorca stations or the Lampedusa station with northern stations. Only 2 cases associated at least both Lampedusa and Mallorca stations. The stations the most often associated to a given MIDD are (i) Frioul and le Casset (6 cases for which at least these 2 stations are associated), (ii) Mallorca and Corsica (5 cases for which at least these 2 stations are associated). This suggests that, in the western Mediterranean basin, the MIDD are associated with different dust provenance and transport pathways, and/or the dust plumes are washed out by precipitation during their transport over the basin.

The joint analysis of the HYSPLIT air mass trajectories and MODIS AOD allows us to identify where dust deposited at the stations for the MIDD is likely coming from. During a sampling week, several dust deposition events (DDE) can be identified and contribute to the weekly deposition flux. A dust event contributing to dust deposition during several days at a station is considered as a single DDE. A MIDD can be a combination of several DDE originating from different dust areas. We identified 132 DDE for the studied period: 50 reached Lampedusa, 27 Mallorca, 22 Frioul, 15 Corsica and 18 Le Casset.

The number of events contributing to the dust deposition is greater for the stations close to the North African dust sources.

For each DDE, the localization of the highest AOD southernmost along the modelled air mass trajectory defined a rough region where dust comes from. As mentioned by Meloni et al. (2008), due to the low resolution of the model meteorological fields and transport model intrinsic errors, the dust location can be relatively wide. It should also be kept in mind that other sources located along the pathway of the dust plume can also contributed to the dust uplifts. Thus, we defined 7 large dust provenance areas (DPA) by grouping together the closest dust localizations (Fig. 6): Niger and Chad (DPA1), northern Mali and southern Mauritania (DPA2), Western Sahara and South Morocco (DPA3), Central Algeria (DPA4), Libya (DPA5), Tunisia and East Algeria (DPA6), and North Morocco and north-western Algeria (DPA7).

The number of DDE at each station (weighted as in Sect. 3.3 and expressed in %) originating from the 7 areas are reported in Fig. 6. 73 % of DDE in Frioul and 69 % of DDE in Le Casset come from the western part of the Sahara (DPA2, 3 and 7). The Western Sahara (DPA3) and Tunisia (DPA6) are the most frequent provenance of DDE reaching Mallorca. Dust deposited during the DDE in Lampedusa generally come from the Tunisian (DPA6) and Libyan (DPA5) regions and the Central Algeria (DPA4). DDE in Corsica generally come from the Western Sahara and South Morocco (DPA3), Tunisia and East Algeria (DPA6) and Libya (DPA5), and the same level of similarity can be observed between the dust provenance areas affecting Mallorca and Corsica than between Corsica and Lampedusa. We also noted that provenance areas, even south of 20° N, like Niger and Chad (DPA1), and northern Mali and southern Mauritania (DPA2), could contribute to DDE (DPA1 for Frioul and Lampedusa, DPA2 for all the stations). Tunisian and Libyan sources have been pointed by Salvador et al. (2014) for specific dust outbreak observed in spring in Balearic Islands and central Mediterranean. This dust transport pathway can be due to high pressure systems located over these sources (Salvador et al., 2014). Moreover, Marconi et al. (2014) also pointed out source regions located in Tunisia-Algeria and Libya to explain atmospheric dust content in Lampedusa. Meloni et al. (2008) indicated the Morocco, Algeria and Tunisia as dust loading areas, as well as southern areas in Mauritania–Mali.

These results confirm that the different parts of the western Mediterranean basin are not affected in the same proportion by Saharan dust coming from different regions. It is nevertheless important to keep in mind that what was tracked here is the southernmost occurrence of dust along the trajectory associated to intense dust deposition events. Hamonou et al. (1999) have shown that dust layers of different origins can also be present concurrently over a given station in the northern part of the Mediterranean.

4.5 Transport routes of the dust deposition events

The main transport routes associated with the 132 DDE in the western Mediterranean Sea were investigated. We classified the forward air mass trajectories computed for each DDE according to their pathway. The six most frequent types of trajectories (representing 96.3 % of all trajectories) are illustrated in Fig. 7. Note that 4 cases among them were classified as “others”, each of them corresponding to a trajectory observed only one time during the studied period. The air-mass trajectories over the western Mediterranean basin are often transported in high altitude (Escudero et al., 2005; Querol et al., 2009). Low level transport of dust are mostly observed at Lampedusa. Trajectories types (a), (c) and (d) are the most frequent transport ways of Saharan dust towards the western Mediterranean basin, since they all together account for almost 70 % of all trajectories. Trajectories type (a) correspond to a straight transport of dust emitted from sources located in Tunisia and/or Libya towards Lampedusa and the the eastern part of western Mediterranean basin. This type of trajectory is the dominant transport in spring (Fig. 8). Trajectories type (c) correspond to transport from sources located in West Algeria/Morocco and Mauritania/Mali towards the western part of the basin and type (d) to transport in a West to East flow from sources located in western and central Sahara and mainly towards the southwestern Mediterranean Basin. They are the dominant Saharan dust transport pathways during summer (Fig. 8). These trajectories have already been mentioned in previous studies as major transport ways for

Saharan dust over the Mediterranean Sea (Bergametti et al., 1989; Guerzoni et al., 1997; Moulin et al., 1998; Israelevich, 2003; Meloni et al., 2008; Marconi et al., 2014). Even if they are less frequent, Saharan dust transport trajectories of type (b) (straight transport towards the westernmost part of the Mediterranean basin from sources located in North Morocco and West Algeria) and (e) (stagnant air masses and cyclonic flow centered over the Atlas and southern Mediterranean) represent 12.1 and 10.6 % of all trajectories, and often occur in spring (Fig. 8).

HYSPLIT model trajectories and precipitation were used to examine whether the different transport trajectories to the western Mediterranean Basin were systematically associated or not with precipitation during transport. We consider a trajectory with an occurrence of precipitation along its path between the Saharan dust provenance area and a given station as a “wet transport case”, whatever the rainfall rate. Main uncertainties are due to the low spatial resolution of the meteorological data set which prevents, for example, accounting for the summer precipitation due to convective cells (Meloni et al., 2008). Moreover, the precipitation is precipitation rate at the grid cell where the trajectory is located and does not take into account the air mass altitude transport. Figure 9 presents the proportion of Saharan dust trajectories coming from each dust provenance area (identified in Sect. 3.4) and for which precipitation during transport have been computed by the HYSPLIT model. Dust air masses from the western Sahara have the highest probability (> 60 %) to be washed during transport. On the opposite, dust air masses from southern and central Sahara are transported about 2/3 of the time in dry conditions. This suggests a more efficient transport of dust from these Saharan regions to the western Mediterranean basin. Air masses coming from the most eastern source regions exhibits intermediate values, with a 50 % probability to be washed during transport.

Results on dust provenance and transport pathways of the DDE suggest that different parts of the western Mediterranean basin are affected by dust deposition events at different periods and from different dust source regions. Dust masses follow different transport trajectories to reach the Mediterranean, some of them being probably washed by precipitation during their transport. This means that Saharan dust inputs to the different parts of the western Mediterranean Basin do not occur at the same time, and can differ in intensity and in composition.

4.6 Dry and wet deposition

In order to provide information on the relative importance of dry and wet deposition to the MIDD, we examined the daily precipitation measured at the sampling stations in combination with the forward air mass trajectories starting from the identified provenance areas. The air mass trajectories provide for each identified dust event a theoretical date of its arrival at a given sampling station. As mentioned above, there are uncertainties on the computed trajectories due to both the model and the resolution of the meteorological fields. Moreover, in most cases, the starting date of the trajectory from the source regions (as determined by looking at the satellite images) is known with a precision not better than ±12 h. Thus, a dust deposition event for which no precipitation is recorded at the station 24 h before and after the dust plume arrival is defined as "dry". For each of the 98 MIDD, the numbers of dry, wet and mixed (wet + dry) DDE were computed. Obviously, this procedure underestimates the number of dry only deposition cases and provides only a lower estimate of dust dry DDE events relatively to the total DDE identified at each station.

Table 4 reports, for each sampling station, the propositions of the wet-only, dry-only, and mixed DDE to MIDD (in terms of occurrences and mass fluxes). Between 47 and 67 % of MIDD (depending on the sampling stations) occur in only wet conditions. Even if dry deposition events can be underestimated following our approach, dry deposition events are far to be negligible both in frequency of occurrence and intensity, especially in Lampedusa, Mallorca and Frioul. In terms of mass, dry deposition represents at least between 15 and 46 % of the deposited mass, the lower contribution being observed in the remote island sites of Mallorca and Corsica. The highest MIDD measured in Lampedusa (2.7 $\mathrm{g\,m^{-2}\,wk^{-1}}$; 20 % of the total deposition flux, respectively) corresponds to a single wet deposition DDE. The second highest MIDD measured in Lampedusa

($2.1\,\mathrm{g\,m^{-2}\,wk^{-1}}$; 16 % of the total deposition flux) corresponds to 2 successive dry DDE. In this latter case, dust plumes were transported below 2000 m. Such a low altitude dust transport in this area was also reported by Barnaba and Gobbi (2004). The highest MIDD measured in Mallorca at the end of April 2013 ($3.2\,\mathrm{g\,m^{-2}\,wk^{-1}}$; 42 % of the total deposition flux) resulted from 3 DDE (1 dry and 2 wet). For the studied period, our results shows that the most intense dust deposition fluxes can be due to single or several successive DDE and can involve wet deposition as well as dry deposition events.

5 Conclusions

A network of 5 sampling sites was deployed from 2011 on the western Mediterranean region to measure the insoluble atmospheric deposition fluxes. It included from South to North an Italian site (Lampedusa Isl.), a Spanish site (Mallorca Isl.) and 3 French sites (Corsica Isl., Frioul Isl., and Le Casset in southeastern France). The data recovery rate varied between 77 and 91 % according to the station. The deposition dataset acquired between 2011 and 2013 include 537 weekly samples. It allowed us to investigate Saharan dust deposition events in this region.

At the three northern stations of the network, Le Casset (44°59′ N–6°28′ E), Frioul (43°15′ N–5°17′ E) and Corsica (43°00′ N–9°21′ E), the maximum deposition fluxes measured on a weekly basis were 0.17, 0.34 and $0.53\,\mathrm{g\,m^{-2}}$, respectively. For the two southern stations, Mallorca (39°15′ N–3°03′ E) and Lampedusa (35°31′ N–12°37′ E), the maximum weekly deposition fluxes are almost one order of magnitude higher, 3.2 and $2.7\,\mathrm{g\,m^{-2}}$, respectively. Deposition fluxes for 1 yr measurements ranged from $7.4\,\mathrm{g\,m^{-2}\,yr^{-1}}$ in the southern part of the western Mediterranean to $0.9\,\mathrm{g\,m^{-2}\,yr^{-1}}$ at the French northern station. This confirms a strong South-to-North decreasing gradient of the atmospheric deposition mass flux in the western Mediterranean region. These annual deposition fluxes are significantly lower than those previously measured between the 1980s and early 2000s mainly because only few intense deposition events

(> 1 g m^{-2}) were recorded. These results seem to be in agreement with the decreasing trend of PM$_{10}$ concentrations over the Mediterranean region and with variation in large scale atmospheric circulation affecting dust atmospheric contents (lower values of the NAO indices during the last two decades).

We selected the 98 most intense dust deposition events (MIDD). They occurred preferentially in spring whatever the sampling station. However, the southern stations of the network (Lampedusa and Mallorca) exhibit a second maximum in autumn while the northern stations (Corsica, Frioul, Le Casset) exhibit this second maximum in summer. Few dust deposition events were recorded simultaneously on several stations, suggesting that different dust events contribute to the deposition measured in different parts of the western Mediterranean.

By matching satellite observations of MODIS AOD and HYSPLIT air mass trajectories, we defined 7 Saharan dust provenance areas and discussed how they contribute to dust deposition in the western Mediterranean region. The western Sahara is by far the most frequent dust provenance of the intense dust deposition measured in the northern and western part of the western Mediterranean basin. The central and the south-eastern parts of the western basin are equally affected by dust transported from western and eastern Saharan regions. In the same way, we also discussed the main dust transport trajectories leading to the Saharan dust deposition in the different parts of the western Mediterranean. We identified six major dust transport routes. Three of them, corresponding to 70 % of all trajectories, are dominant in spring (one trajectory type, (a)) and in summer (the other two, (c and d)).

Finally, we showed that several dust deposition events (DDE) can contribute to high weekly dust deposition fluxes measured in the stations of the network. The daily precipitation measured at the station allowed us to discuss the relative contribution of wet and dry dust deposition to the weekly deposition fluxes. Even if the procedure we used underestimates the dry deposition occurrences, the results show that dry deposition can contribute significantly for the highest deposition fluxes (MIDD) to the total deposition, from 15 and 46 % of the total deposited mass depending on the station.

The results show that dust deposition in the western Mediterranean region is far to be homogeneous. A high spatial and temporal variability of the deposition is observed. A strong south to north gradient exists and different source regions contribute to dust deposition in the different parts of the central and western Mediterranean. The deposition occurs in different periods, with different transport routes depending on the season. This suggests a seasonal variability within the western Mediterranean basin. This unique dataset will be used to test the dust deposition in atmospheric transport models.

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Table 1. Dust deposition fluxes measured in the Mediterranean basin and Northern Africa.

Region	Location	Period	Duration	Deposition flux ($g\,m^{-2}\,yr^{-1}$)	Reference
Western Mediterranean	Cap Ferrat and 3 sites SE France and NW Corsica	2003–2007	4 years[a]	11.4	Ternon et al. (2010)
	Cap Bear, SW France	2001–2002	12 months	10.6[c]	Guieu et al. (2010)
	Capo Cavallo, NW Corsica	1985–1986	12.5 months	12.5[c]	Bergametti et al. (1989)
		1986–1987	20 months	9.7	Remoudaki (1990)
	3 inland sites, Corsica Isl.	1984–1994	10 years[b]	4–26	Loÿe-Pilot and Martin (1996)
	Ostriconi, N Corsica	2001–2002	12 months	27.4[c]	Guieu et al. (2010)
	Pirio, NW Corsica	1995–1997	27 months	2–4[c]	Ridame et al. (1999)
		1999–2000	13 months[a]	9–14[c]	Loÿe-Pilot et al. (2001)
	Capo Carbonara, SE Sardinia	1990–1992	19 months	12.8[c]	Guerzoni et al. (1999)
	Montseny, NE Spain	1983–1994	11 years	5.2 Dust rains only	Avila et al. (1997)
	Palma de, Baleares	1982–2003	22 years	~ 14 Dust rains only	Fiol et al. (2005)
	Campo de Gibraltar S Spain	1982–1983	12 months	22.8[c]	Usero and Gracia (1986)
	Lanjaron, SE Spain	2001–2002	23 months	11.1	Morales-Baquero et al. (2006)
Eastern Mediterranean	Erdemli, SE Turkey	1991–1992	16 months	13.0	Kubilay et al. (2000)
	Akkuyu, SE Turkey	2001–2002	12 months	10.1[c]	Guieu et al. (2010)
	6 sites, Crete	1988–1994	6 years	11–23	Mattson and Nihlén (1994)
	7 sites, Crete	1988–1990	34 month	10–100	Pye (1992)
	Finokalia, NE Crete	2001–2002	12 months	8.8[c]	Guieu et al. (2010)
	Cavo Greco, SE Cyprus	2001–2002	12 months	4.2[c]	Guieu et al. (2010)
	Mytilene, Lesbos Isl.	2001–2002	12 months	5.4[c]	Guieu et al. (2010)
	Israel	1965–1995	[a]	30–90	Ganor and Foner (2001)

Table 1. Continued.

Region	Location	Period	Duration	Deposition flux ($g\,m^{-2}\,yr^{-1}$)	Reference
North Africa	Alexandria, N Egypt	2001–2002	8.5 months	20.3^c	Guieu et al. (2010)
	14 inland sites, N Libya	2000–2001	12 months	58 (< 20 µm only)	O'Hara et al. (2006)
	Cap Spartel, N Morocco	2001–2002	12 months	7.2^c	Guieu et al. (2010)
	Mahdia, E Tunisia	2001–2002	12 months	23.3^c	Guieu et al. (2010)

[a] Sampling performed in different places and periods, [b] in different places, [c] assuming Al is 7.1 % of total dust (Guieu et al., 2002).

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Table 2. Number of weekly deposition fluxes measured, number (and relative proportion in %) of the most intense weekly deposition fluxes recorded (MID), number of MID events with identified Saharan provenance area (MIDD), and their respective contribution (% in mass) to the deposition fluxes measured, at each station of the network.

	Number of weekly samples	Total deposition flux ($g\,m^{-2}\,yr^{-1}$)	Threshold flux ($g\,m^{-2}\,wk^{-1}$)	MID	MID contribution to total deposition fluxes (%)	MIDD	MIDD contribution to total deposition fluxes (%)
Le Casset	119	2.17	0.036	18 (15 %)	61	15	53
Frioul	123	5.84	0.093	21 (17 %)	55	18	49
Corsica	78	2.59	0.050	11 (14 %)	73	11	73
Mallorca	117	9.74	0.093	21 (18 %)	80	20	78
Lampedusa	100	16.0	0.093	37 (37 %)	87	34	84

Table 3. Number of stations operating and number of stations recording a MIDD during the same sampling week.

	Number of stations recording simultaneously a MIDD					
Number of stations operating	5	4	3	2	1	Total
5	0	1	0	3	23	27
4	–	0	4	7	23	35
3	–	–	0	0	7	4
2	–	–	–	2	2	6
1	–	–	–	–	3	3
Total	0	1	4	12	58	75

Table 4. MIDD during which DDE occur only by dry, by wet or by mixed (wet + dry) deposition.

	Number of MIDD				MIDD cumulated mass			
	Total	Wet deposition	Dry deposition	Wet + dry deposition (mixed)	Total mass ($g\,m^{-2}$)	Wet deposition (%)	Dry deposition (%)	Wet + dry deposition (mixed, %)
Le Casset	15	10	4	1	1.2	58	37	5
Frioul	17[a]	8	8	1	2.7	49	38	13
Corsica	11	7	3	1	1.9	69	15	16
Mallorca	20	11	7	2	7.6	35	17	49
Lampedusa	34	18	15	1	13.5	51	46	3

[a] 17 MIDD plus 1 MIDD for wich no precipitation data was available at Frioul station.

Figure 1. The CARAGA collector operating on Frioul Island.

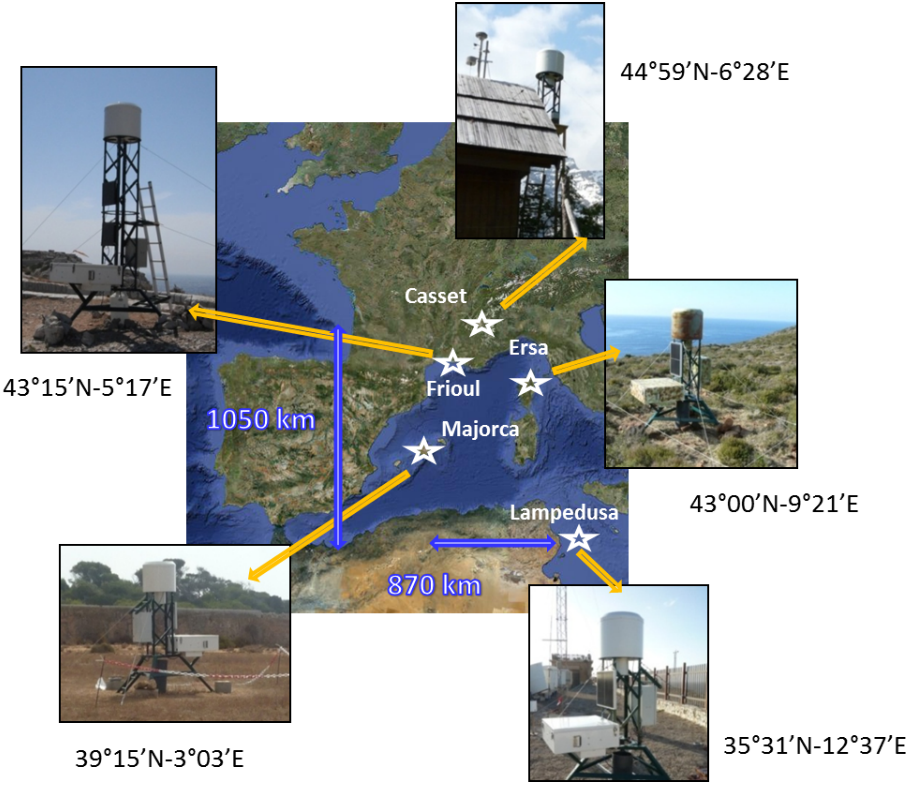

Figure 2. Location of the CARAGA samplers constituting the deposition network deployed in the western Mediterranean basin and southern of France.

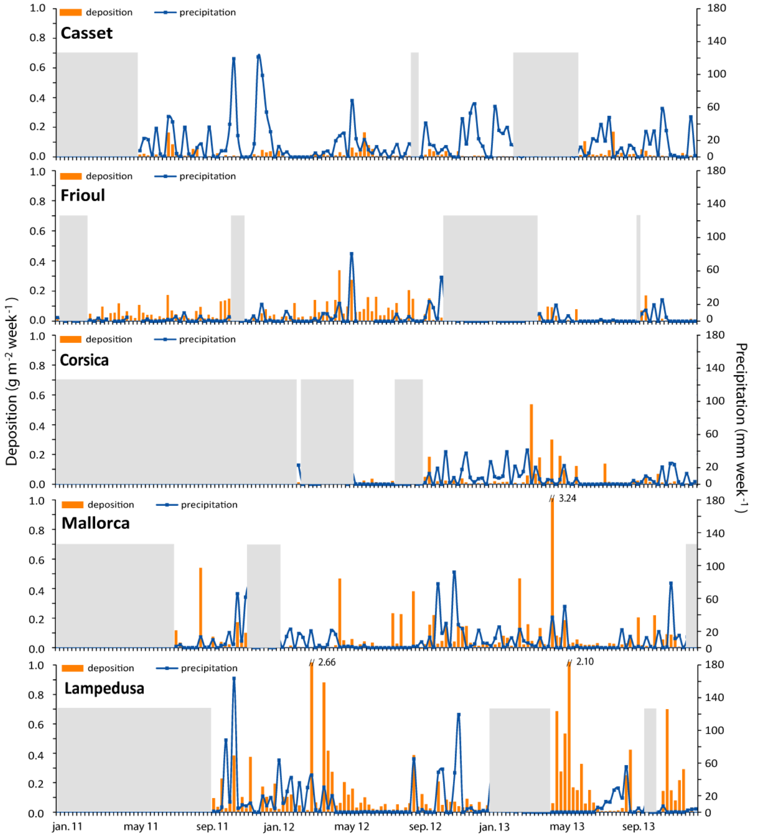

Figure 3. Weekly insoluble mineral deposition fluxes (orange bars) and precipitation amount (blue line) for Lampedusa, Mallorca, Corsica, Frioul and Le Casset from January 2011 to December 2013. The grey areas correspond to periods without sampling. The number of most intense dust deposition for each station as described in Sect. 4.3 are: 34 in Lampedusa, 20 in Mallorca, 11 in Corsica, 18 in Frioul and 15 in Le Casset.

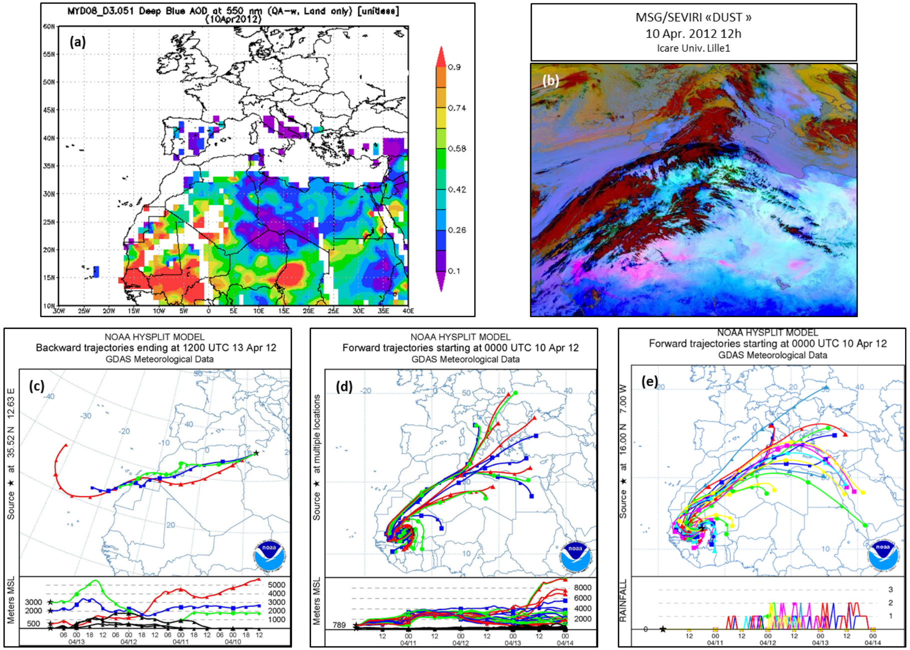

Figure 4. Illustration of the data used jointly to identify a dust transport event and its origin which leeds to high deposition in Lampedusa between 12 and 13 April 2012. **(a)** MODIS deep blue AOD, **(b)** MSG/SEVIRI dust false-colour composite product, **(c)** HYSPLIT backward trajectories for 3 starting heights (500, 2000 and 3000 m), **(d)** HYSPLIT forward trajectories in matrix mode with corresponding altitudes, **(e)** HYSPLIT forward trajectories in ensemble mode with corresponding precipitation.

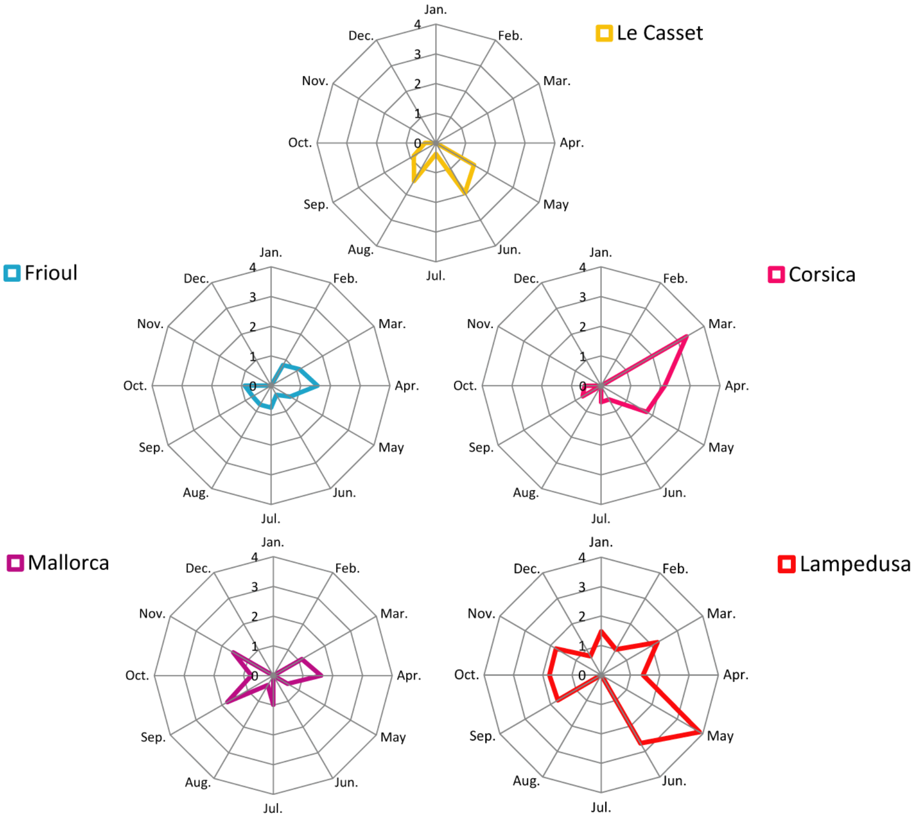

Figure 5. Weighted number of occurrence of MIDD per month over the whole sampling period at each site.

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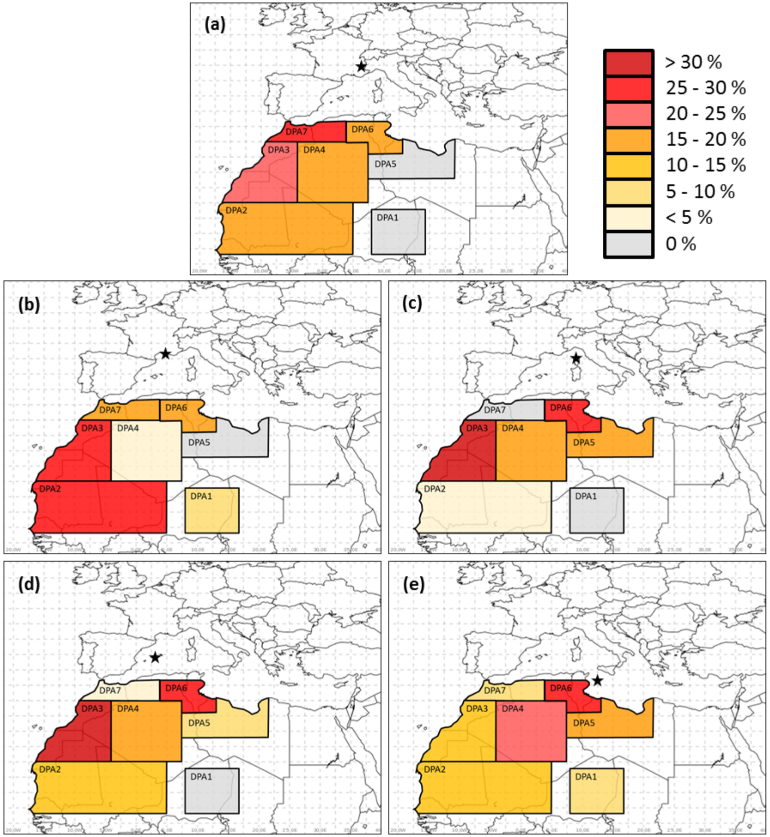

Figure 6. Frequency of dust provenance areas identified using MODIS AOD and HYSPLIT air mass trajectories for the DDE contributing to the MIDD recorded at **(a)** Le Casset, **(b)** Frioul, **(c)** Corsica, **(d)** Mallorca, and **(e)** Lampedusa.

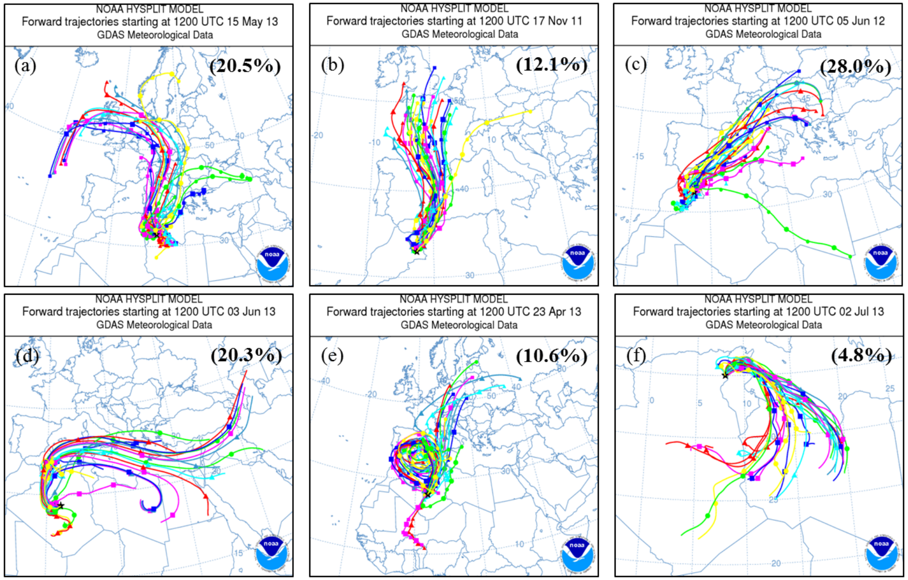

Figure 7. Typical forward air-mass trajectories computed with the HYSPLIT model and corresponding to the different Saharan deposition events collected over the western Mediterranean basin. The number in brackets indicates the relative occurrence frequency for each of the six cases (3.7 % are unclassified).

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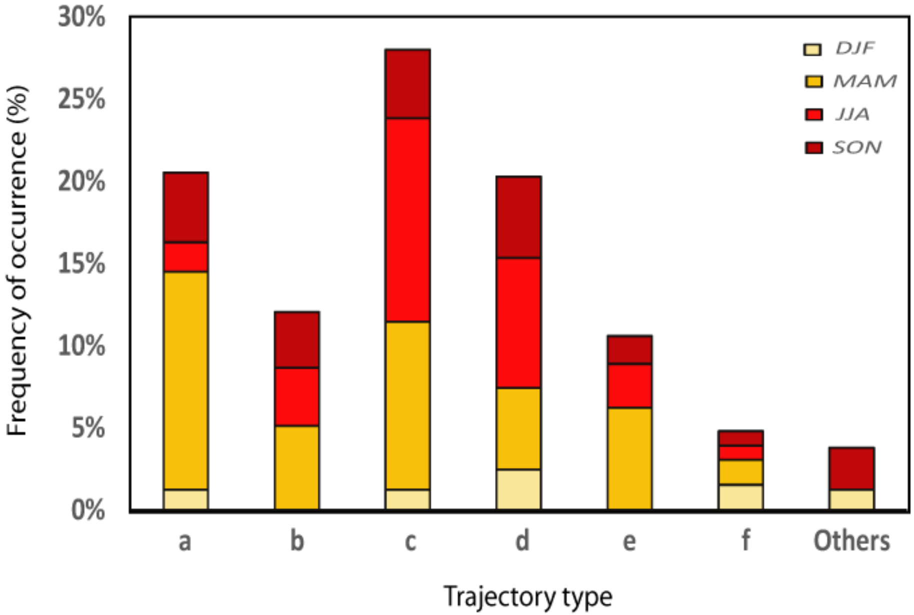

Figure 8. Seasonal occurrence of the different Saharan dust trajectories (see text for details).

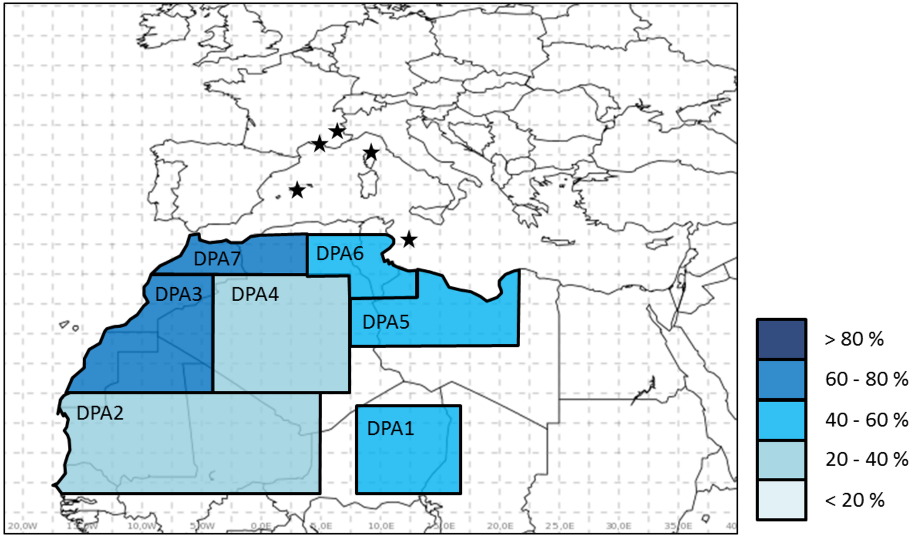

Figure 9. Proportion of HYSLPIT trajectories for the DDE (in %) with precipitation during their transport between the source-regions and the western Mediterranean basin.

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