Saharan dust deposition effects on production in the Mediterranean Sea

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A mi pequeña gran familia

Preface

“The mind that opens to a new idea, never returns to its original size”

Albert Einstein

“In all things of nature there is something of the marvelous.”

Aristotele

“Why not go out on a limb? That’s where the fruit is”

Mark Twain

Abstract

The potential capacity of fertilization of surface oceans by atmospheric deposition is of scientific interest. This is especially true in the oligotrophic Mediterranean Sea. Its surface waters are extremely poor in nutrients necessary for plankton growth. At the same time, it borders with the largest and most active desert area in the world and the atmosphere over the basin is subject to frequent injections of mineral dust particles. Moreover, future scenarios foresee increases in the region's aridity, thus increasing the dust load, as well as changes in ocean stratification that will increase the potential impact of dust deposition on surface waters. Thus, it is important to study the links between deposition and plankton stimulation.

In this context, the present thesis addresses relationships between desert dust deposition and phytoplankton dynamics in the Mediterranean Sea. It places especial emphasis: (1) on the seasonal and geographical patterns of both dust deposition and chlorophyll variability, (2) on the correlation between dust deposition and chlorophyll concentration, (3) on the analysis of the large deposition events and (4) on the effects on chlorophyll concentration due to very large dust deposition events occurred between 2000 and 2007.

Broadly, dust deposition seasonal dynamics shows highest values in late autumn and winter in the Central and Eastern Mediterranean. While, the high dust deposition occurs close to Africa with a decreasing gradient from South to North in the basin. By contrast, the distribution of chlorophyll shows decreasing gradients both from West to East and from North to South. In addition, its broadest variability was found in the Western Mediterranean, coinciding with the highest chlorophyll concentrations on average.

Positive correlations between dust deposition and chlorophyll concentration were found in large areas of the Mediterranean Sea, with a clear South to North decreasing gradient in correlation coefficient. This is especially true for the Central and the Eastern Mediterranean, where dust deposition dynamics matches with chlorophyll annual dynamics. Areas with positive correlations can be found during all seasons, although it is in spring when we see the largest correlation coefficients mainly in the Central, Eastern and Southwestern Mediterranean.

Finally, 153 large dust deposition events were identified between 2000 and 2007. May of them occurred in the years 2000 and 2004, especially in autumn and winter even if they showed a high variability. The Eastern Mediterranean was more affected by the extensive events especially in winter. About the very large dust deposition events, 31 were identified during this period. They were distributed unequally over seasons and over the Mediterranean sub-basins, occurring mainly in autumn and winter in the Central and eastern Mediterranean.

Chlorophyll concentration increases significantly after very large dust deposition events showing peaks in concentration between days 1 and 6 after the event, with chlorophyll increments ranging from 13% to 345% for the different outbreaks. The impact of these large events on chlorophyll shows a decreasing trend from west to east and seems to be related to the eastward increasing importance of heterotrophic bacteria with respect to phytoplankton.

Resumen

La potencial capacidad de fertilización de la superficie de los océanos por deposición atmosférica es de interés científico. Esto es especialmente cierto en el oligotrófico Mar Mediterráneo. Sus aguas superficiales son extremadamente pobres en nutrientes necesarios para el crecimiento del plancton. Al mismo tiempo, limita con la mayor y más activa zona desértica en el mundo y la atmósfera sobre la cuenca está sujeta a frecuentes inyecciones de partículas de polvo mineral. Por otra parte, los escenarios futuros prevén aumentos en la aridez de la región, lo que aumenta la carga de polvo, así como cambios en la estratificación del océano que aumentará el impacto potencial de la deposición de polvo en las aguas superficiales. Por lo tanto, es importante estudiar los vínculos entre la deposición y la estimulación de plancton.

En este contexto, la presente tesis aborda las relaciones entre la deposición de polvo del desierto y la dinámica del fitoplancton en el Mar Mediterráneo. Pone especial énfasis: (1) sobre los patrones estacionales y geográficos tanto de la deposición de polvo cuanto de la variabilidad de la clorofila, (2) en la correlación entre la deposición de polvo y la concentración de clorofila, (3) en el análisis de los grandes eventos de deposición y (4) en los efectos sobre la concentración de clorofila debido a los grandes eventos de deposición de polvo producidos entre 2000 y 2007.

En términos generales, la dinámica estacional de la deposición de polvo muestra valores más altos a finales de otoño y en invierno en el Mediterráneo central y oriental. Mientras, la mas alta concentración de deposición de polvo se produce cerca de África con un gradiente decreciente de sur a norte en la cuenca. Por el contrario, la distribución de la clorofila muestra la disminución del gradientes tanto de oeste a este cuanto de norte a sur. Además, su variabilidad más amplia se encuentra en el Mediterráneo Occidental, coincidiendo con las más altas concentraciones de clorofila en promedio.

Correlaciones positivas entre la deposición de polvo y la concentración de clorofila se encuentran en grandes áreas del Mar Mediterráneo, con un claro gradiente decreciente de Sur a Norte del coeficiente de correlación. Esto es especialmente cierto para el Mediterráneo central y oriental, donde la dinámica de deposición de polvo coincide con la dinámica anual de clorofila. Las áreas con correlaciones positivas se pueden encontrar durante todo el año, aunque es en primavera, cuando vemos los coeficientes de correlación mas altos sobre todo en el centro, este y suroeste del Mediterráneo.

Por último, 153 grandes eventos de deposición de polvo se identificaron entre 2000 y 2007. La mayoría de ellos se produjeron en los años 2000 y 2004, sobre todo en otoño e invierno aunque mostraron una alta variabilidad.

El Mediterráneo oriental se ve más afectado por los extensos eventos, sobre todo en invierno. Acerca de los muy grandes eventos de deposición de polvo, 31 fueron identificados durante este período. Fueron distribuidos de manera desigual durante las estaciones del año y en las sub-cuencas del Mediterráneo, se produjeron principalmente en otoño e invierno en el centro y en el este del Mediterráneo. La concentración de clorofila aumenta significativamente después de los muy grandes eventos de deposición de polvo y muestra picos de concentración entre el día 1 y 6 después del evento, con un incrementos de la clorofila que van del 13% al 345% para los diferentes eventos. El impacto de estos grandes eventos en la clorofila muestra una tendencia a la disminución de oeste a este y parece estar relacionado con la importancia cada vez mayor hacia el este de bacterias heterotróficas con respecto al fitoplancton.

Contents

Introduction

The Mediterranean Sea

The etymology of Mediterranean Sea comes from Latin “Mar Medi Terraneum ”, which means “sea in the middle of the land”. This is because, it is a semi-enclosed sea surrounded by the Mediterranean region that include south Europe, west Asia and north Africa; with a total of 22 countries that overlook on it.

The Mediterranean Sea is temperate mid-latitude basin with an extension of 2.5 10^6 km2. It is 3900 km long with a maximum width of 1600 km and its greatest depth is 5121 m. It connects with the Atlantic Ocean through the Strait of Gibraltar; with the Black Sea through the Dardanelles and the Bosporus; and with the Red Sea through the Suez Canal. Notwithstanding it occupies only the 1% of the world’s ocean surface it could be considered an isolated oceanic system. Moreover, for its peculiar environmental conditions could be considered also an excellent natural laboratory where processes can be studies at small scale. It is characterized by thermohaline circulation that is determined by the flux of the incoming fresh Atlantic water through the Gibraltar Straits and by the sinking of cold waters formed at the Lions Gulf and at the Adriatic (Pinardi and Masetti, 2000; Malanotte-Rizzoli, 2001; Robinson et al., 2001; Tanhua et al., 2013). The Atlantic incoming waters increase in density during they travel through the basin and this because evaporation exceeds precipitation. On the other hand, occur the formation of new water masses via convection events driven by intense local cooling especially during winter storms. Bottom water is produced: for the western basin in the Gulf of Lions and for the eastern basin in the southern Adriatic (Malanotte-Rizzoli, 2001; Robinson et al., 2001; Tanhua et al., 2013). Another characteristic is its high salinity concentration, which is due to the imbalance between precipitation, river discharge and evaporation. The net evaporation into the Mediterranean Sea is calculated to be of 50 to 100 cm year−1, this is the result between evaporation, precipitations and river run-off. It also loses heat to the atmosphere. Part of these losses is balanced by the inflow–outflow through the Strait of Gibraltar. Enter warmer, fresher and deep Atlantic water and go out colder, saltier and superficial Mediterranean water (Borghini et al., 2014). Also, the Mediterranean Sea is considered an oligotrophic basin, due to its primary production generally weak and because the chlorophyll concentration in the open ocean rarely exceeds 3 mg m-3 (D’Ortenzio and Ribera D’Alcalà, 2009). Here, the phytoplankton follows a typical mid-latitudinal seasonal dynamics with a biomass increase in late winter, early spring and very low values in summer (Marty et al., 2002). The coupling with physical forcing, such as the wind action, seems to controls the phytoplankton biomass increase. During winter, the column water is totally mixed and this allows the transport of nutrient, from deep waters to superficial layer, inducing favorable condition for the phytoplankton growth. On contrary, during summer the Mediterranean is characterized by a marked and extend stratification period during warmer season, which inhibit the nutrient flux toward the superficial layer. This mechanism is the responsible for the oligotrophic and ultra-oligotrophic condition of the basin (Goffredo and Dubinsky, 2014). In addition, the chlorophyll geographical distribution shows an east-west basin gradient which highlighting an ultra-oligotrophic condition of Eastern basin vs the most productive Western side (Morel and Andre, 1991; Antoine et al., 1995; Bosc et al., 2004). The chlorophyll concentration is principally controlled by the nutrient availability into the Mediterranean superficial waters. The basin nutrient budget present a west-east decrease gradient for nitrogen (Ribera d'Alcalà et al., 2003), phosphorous north-south decrease gradient (Lazzari et al., 2016), while the silicon only shows a slight increase (Ribera d'Alcalà et al., 2003). Nutrients values range between 4.75, 0.15, and 5.5 µmol dm-3 for nitrates, phosphates and silicates, respectively, in the east Mediterranean and 8.25, 0.32, and 6.5 µmol dm-3 for nitrates, phosphates and silicates, respectively, in the west Mediterranean (Ribera d'Alcalà et al., 2003). The mean concentration of nitrates, phosphates and silicates rarely exceed 0.5, 0.05 and 1.5 µmol dm-3 respectively. During summer or fall the phosphates especially in the east Mediterranean could be below the detection limit. Through, the analysis of nutrients concentration shows that the Mediterranean Sea have an anomalous Redfield ratio compared with other oceanic province. The $N:P>25$ and $Si:N>1.3$ ratio in the east Mediterranean vs $N:P\sim20$ and $Si:N\leq1$ ratio in the west Mediterranean display a decreasing east-west gradient (Ribera d'Alcalà et al., 2003). Nowadays, this anomaly is an open issue for understanding the Mediterranean internal processes which control its functioning. Other open issue is how external source such as the river or atmospheric inputs could alleviate the Mediterranean waters nutrient starvation and allow the phytoplankton increase.

Atmosphere and ocean interaction

The main connection between atmosphere and oceans is their ability to store and exchange energy between them in form of heat, moisture and momentum. This dynamic balance plays an important role in the Earth climate control. In addition, they could exchange a large number of minerals and chemical compounds, which when present in the atmosphere are commonly named aerosols.
Over the past three decades it has become evident that the atmosphere has a significant role about the contribution of both natural and anthropogenic substances into the surface seawaters. The atmosphere is one of the principal means by which different materials are moved from mainland regions towards the open Ocean (Goudie and Middleton, 2006). When these particles are moved from the continent, they are stored in the troposphere where they spend a relative short residence time. Despite this time, the aerosol particles, thanks to the winds blow, have the capability to travel far away from their origin source. Also, the atmosphere could be considered the most important pathway for the long-range transport of continental particles to open ocean. As well as the atmospheric transport disperses material widely (up to hundreds of kilometers far away), the deposition fluxes decrease as aerosol particles move far away from their source regions. The accumulation due to the deposition of these substances could alter or have an influence (as positive as negative)

on the biological, chemical and geological marine processes. For example, substances as lead and copper could be potentially harmful to the marine biological systems. Others, such as nitrogen, phosphorous, silicon and iron are essential nutrients for the "green" seawater component and they could enhance the marine productivity. For this reason, the aerosol particles could be considered an important environmental modifier (Chester and Jickells, 2012).

Desert dust

Dust is a very significant component of the desert system. It affects not only the local processes (geomorphological or biological) or atmospheric condition but influence the atmosphere, the land and the ocean surface far from its source area.

The aeolian erosion in arid and semi-arid regions is the principal responsible by which atmospheric dust derived. Also Sahara region and Gobi desert are considered to be the most important source. As calculated by Ginoux et al. (2001) the total maximum global emission of dust into the atmosphere is almost 2 billion t yr^{-1} representing about half of the total tropospheric aerosols. Again, the highest dust concentrations over Western Sahara and the Sahel region (above 250 μg m^{-3}) and a deposition rate of 162 g m^{-2} yr^{-1}. Dust emission into the atmosphere occurs as sporadic events that could vary in duration, magnitude, season and particle concentration or size.

Dust plays many environmental roles, depending on the nature and magnitude of the event, the distance traveled by dust and the chemical compounds that are present. The effects can be divided in local, regional or global scale. At the local scale, it can influence soil formation or composition, affect plant photosynthesis, cause human or animal health problems. At the regional or global scale, it can affect terrestrial and marine ecosystems, soil development, air quality or modify Earth's climatic parameters (e.g., rainfall)..

Mineral dust contributes micronutrients to terrestrial and marine ecosystems that have the potential to act as a fertilizer (Baker et al., 2006; Bristow et al., 2010). In the oceans dust particles can enhance biological production, because provide nutrients to phytoplankton and zooplankton in ocean areas that are far from supplies provided by rivers discharge. Sahara desert dust, e.g, is an important nutrient source for the Atlantic Ocean and the Mediterranean Sea (Goudie and Middleton, 2001). In the terrestrial ecosystems dust deposition benefits have also been shown. The 40 million t yr^{-1} of dust transported from Sahara are able to fertilize the Amazon and enhance the productivity of the rain forest (Swap et al., 1992).

Also, dust events can inject a great quantity of microorganisms and pollen into the atmosphere, playing an important role in transporting pathogens or expanding the geographical range of some organisms (Kellogg and Griffin, 2006). It has shown that Saharan dust play a significant role in Caribbean coral reef vitality decline (Garrison et al., 2003). Furthermore, in Caribbean air samples were determinate, that during African dust events, the number of cultivatable airborne microorganisms was between 2 and 3 times higher than normal atmospheric conditions (Griffin et al., 2001).

About soil development, dust plays an important role in earth surface processes, soil and pavement formation, and dune stabilization. Dust also impacts on climate, weather and air quality. The dust in the atmosphere is capable to affect the radioactive budget of the earth by absorbing and scattering the solar and terrestrial radiation, as well as it is able to perturbing the atmospheric circulation patterns (Miller and Tegen, 1998). Dust storm is capable to reduce the solar incoming radiation more than 60% (Badarinath et al., 2007). Additionally, dust can act as condensation nuclei, facilitating the processes of cloud formation (Maley, 1982). As well as, it reduce the rainwater acidity (Carratala et al., 1996).

The Sahara Desert

Deserts areas take up one-fifth of the Earth's land surface, and the Sahara Desert is considered the largest desert in the world with an extension of about 9,4x106 km2. It is located in the north part of Africa (Fig. 1) and it covers about 1/4 of the entire continent. It stretches from the Red Sea in the east, the Mediterranean Sea in the north and the Atlantic Ocean in the west side, while in the south part it is delimited by the Sahel.

Moreover, the Sahara is the most diverse in terms of landforms found within it. Landforms are considered as the surface expression of the interaction between geomorphic processes and the lithology.

Figure 1: Sahara Desert "blue marble" image by NASA's MODIS instrument (Source: http://visibleearth.nasa.gov/view.php?id=57752).

Beside, the landforms dislocations provide information about where the geomorphic processes dominate.

From a geomorphologic point of view, the desert region is composed by very peculiar and distinctive landscapes (Fig. 2) as result of wind blow action on arid and hyper-arid regions. Most of the Sahara Desert area consists of rocky and sand surfaces and it can be described by three main different landscapes. In the rocky region, the wind blow has removed most of the fine sands and it produces the landscape known as rocky desert or Hamada. Here, the loss of fine-grained and sand, by deflation, generates typical desert scenery consisting of gravel, boulders and bare rocks as well as, of high, largely barren, hard, rocky plateaus (Fig. 2A). Then as it is shown in figure 2B, the rocky mountain give way to a mixture of sand and gravel known as Reg or desert pavement of loose stone. Generally, these structures are the remains of prehistoric seabeds and riverbeds, now drained. Finally, the desert areas where sands are accumulated are known as Ergs. An Erg is a broad, flat area of desert covered with wind-swept sand with little or no vegetative cover, as is shown in figure 2C. Here the sand is amassed in huge dunes, which could reach over 180 meters in height. Nevertheless, the dunes and the sandsheet landform classes are not the predominant.

Figure 2: In panel A it is shown the Hamada; in B the Reg and in C the Erg landscape (source: https://sites.google.com/site/worlddeserts/landforms/regs-hamadas).

As it is possible to appreciate in figure 3, they cover only the 37% of the Sahara Desert. They are the areas with high sediment availability but it is limited by wind transport capacity. Then, the 21% of desert is occupied by Regs. These are areas of potentially high sediment storage, but due to the armoring of the surface, there is low sediment availability. Finally, the rest of the desert is formed by rocks (Ballantine et al., 2005; Laity, 2009).

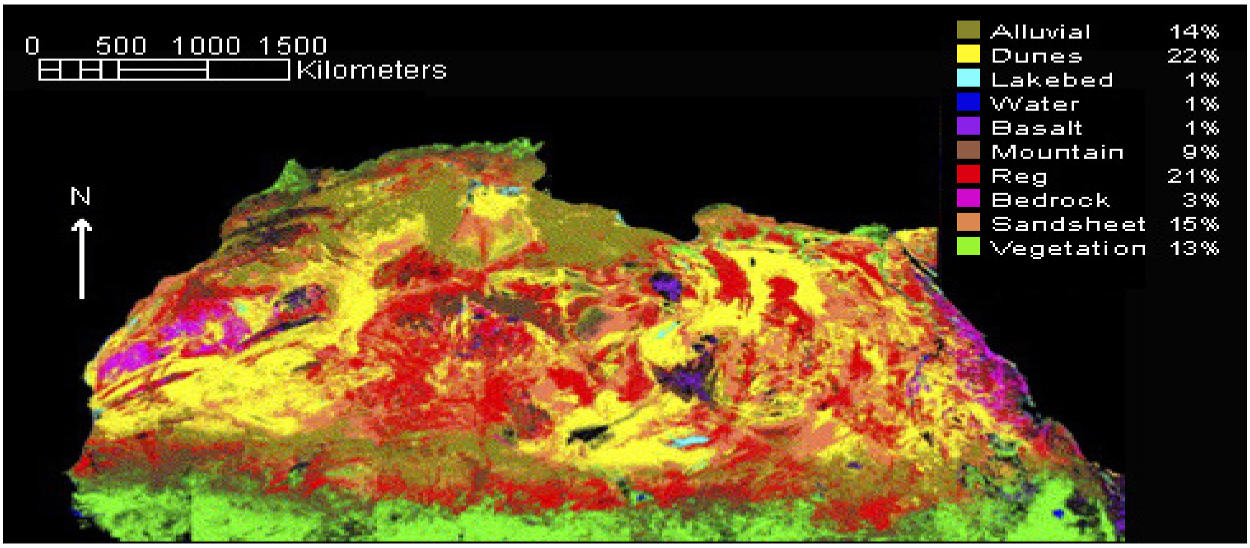

Figure 3: Landform map of Sahara desert and the cover percentage for each landform class (from Ballantine et al. (2005)).

The Sahara has been recognized as the world's largest desert region as well as the most important aeolian desert dust source in the world (Schütz et al., 1981). Most of the world's atmospheric aerosol consists in dust eroded from Sahara (Prospero et al., 2002). It is estimated that the Sahara annual dust production is of 840 Tg·yr-1 vs. the global desert dust production of 1536 Tg·yr-1; this means that the Sahara account for 55% of the total global dust emissions (Ginoux et al., 2012).

Transport and deposition

The importance of some regions as dust sources is linked to different factors; a high content of fine-grained particles is not sufficient to qualify a desert area as a source of dust. Moreover, the topography of the area, the weather conditions and the presence of sediment surface of high-speed wind are very important factors, which depends on if the particles are put in movement or not. Dust transport involves three stages: entrainment, dispersion and deposition. While the nature of sediment transport is controlled both by nature of wind near ground and by the ground surface properties over which the flow occurs (Pye, 1987).

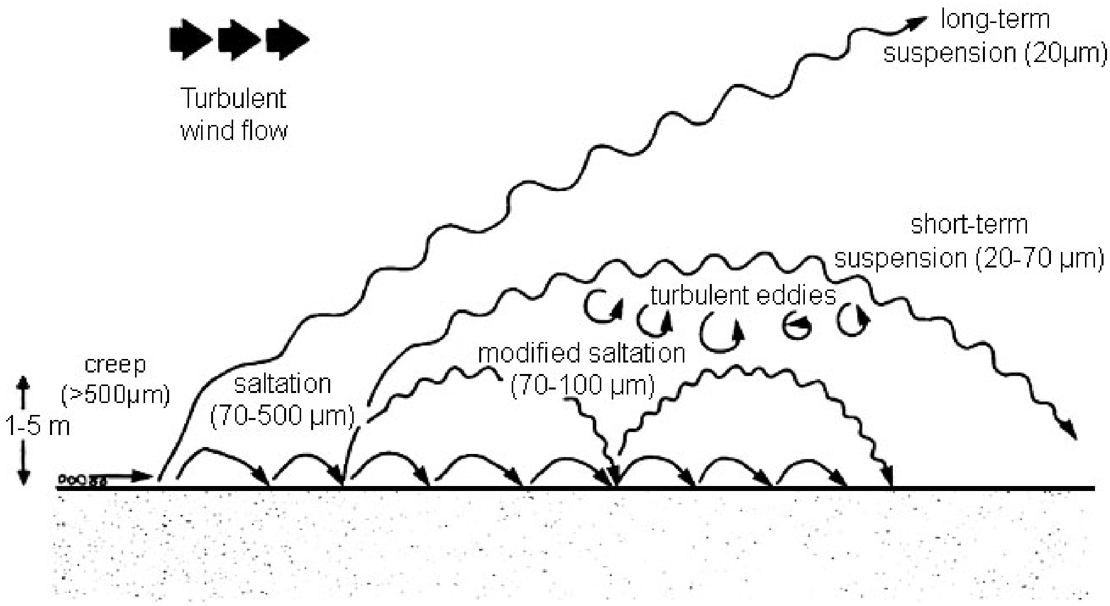

Figure 4: modes of sediment particles transport by wind flow (from Pye (1987)).

Principally, the movement type depends on particles diameter (Fig. 4). Normally, particles with diameter more than 500 μm, due to their size, have a propensity for crawl or rolling, while these between 500 and 70 μm are moved by saltation. Particles with dimensions < 70 μm are transported by suspension, even if due to their weight, they fall down rapidly. Finally, these ones smaller than 20 μm could be transported far away from their origin source; they could travel for hundreds of kilometers. Once the dust particles are mobilized from their desert source they can be transported far away (Fig.5). Thanks to the wind action dust particles are injected into the atmosphere. Here, they could have an effect on the solar radiation, altering the Earth radioactive balance. Furthermore, the dust particles during their transport interacted with atmospheric gases and water so that; they could be subjected to chemical modifications. Additionally, they are subjected to wind selection. The wind selects the particles by granulometry, transporting far away, whose are small and light while the heavy particles fall down immediately. For this reason the particles transported, for example, toward the Atlantic Ocean have dimensions between 1 and 5 µm (Perry et al., 1997; Prospero et al., 2001). The smallest particles are raised at high altitude into the troposphere and travel hundreds kilometers. Later, they are deposited by wet or dry deposition mechanisms.

The dust deposition may give rise to the formation of aeolian deposits, both into earth and oceans. The most evident effect of this phenomenon is the formation of loess and sea-mounts (Stuut et al., 2009). Loess deposits cover about 10% of the Earth's surface (Pecsi, 1968), while it is estimated that in parts of the Pacific Ocean, more than half of the total sediment is of aeolian origin (Rea et al., 1985). And in some ocean areas the aeolian dust comprises up to the 80% of the total accumulated sediments (Pye, 1987).

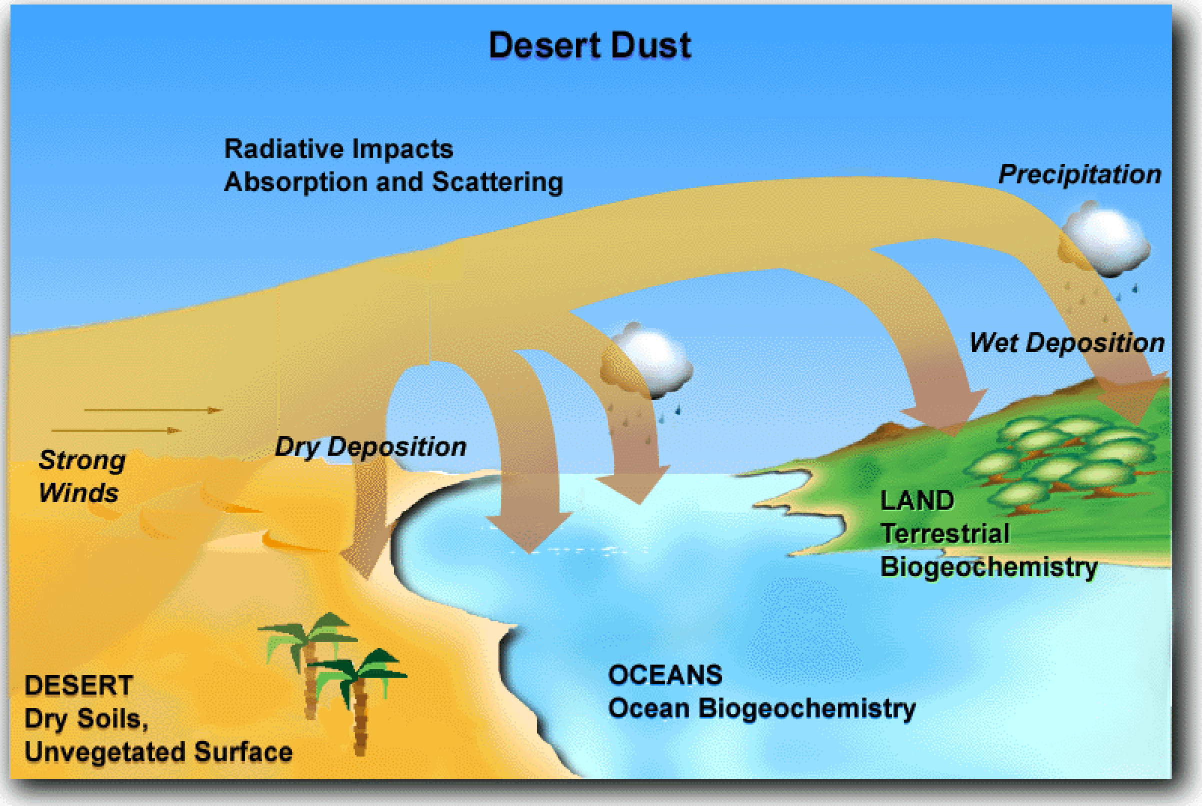

Figure 5: Schematic view of Desert dust deposition types (from http://www.geo.cornell.edu/eas/PeoplePlaces/Faculty/mahowald/dust.htm).

Impacts of dust in the Mediterranean

Dust could have important effects on climate. In the east Mediterranean case, the dust particles are be demonstrated to often coated with soluble sulphate and other soluble materials. These Large insoluble particles (dust) coated with hygroscopic material, permit the large drops formation, acting as cloud condensation nuclei. So the presence of dust particles in the atmosphere could substantially increase the cloud formation and the rain increase (Levin and Ganor, 1996). Also, the marine chemistry may be affected by atmospheric dust deposition. This because the dust constitutes the major source of new nutrients (N, P, Si, Fe and trace metals) in the Mediterranean open surface water(Krom et al., 2004; Bonnet and Guieu, 2006; Pulido-Villena et al., 2010).

As dust could bring nutrient to the surface waters it could also act as efficient mechanism to remove dissolved nutrients from ocean surface waters, by adsorption onto sinking particles (Wagener et al., 2010). For this reason, atmospheric deposition plays an important role on biogeochemical elemental cycling by acting as both a source and a sink for dissolved nutrients in the Mediterranean surface seawater. Furthermore, dust could increase the sinking velocity of organic particles (Ploug et al., 2008) and affected the carbon export due to a ballast effect on POC export (Ternon et al., 2010). Finally, due to the huge quantity of dust deposited every year into the surface Mediterranean waters, it has an important role into the deep marine sediments of the basin. The atmospheric input of mineral dust is the major source of no biogenic sediment in the Mediterranean Sea (Bergametti et al., 1989).

About the stimulation effects of desert dust on phytoplankton, there are some questions which remain opened due to de difficulty to have a wide vision of the phenomenon. They are carried out different experiments, but without a general tendency. In Bonnet et al. (2005) and Laghdass et al. (2011) were investigated both in situ microcosm and mesocosm experiments. They found that the phytoplankton response was positive to dust addition. Moreover, in Romero et al. (2011) was detected the quick response by bacteria and then by phytoplankton community. Mixed response were found by Eker-Develi et al. (2006) and Ridame et al. (2014) while Volpe et al. (2009) did not observed satellite chlorophyll changes after dust depositions events.

Another open issue is the dust deposition data acquisition. The dust deposition samples are generally collected in sporadic stations into the continent (Avila et al., 1997; Guerzoni et al., 1999; Morales-Baquero et al., 2006; Guieu et al., 2010) or inference by the dust source monitoring and transport pathways, especially by using satellite observations (Brooks and Legrand, 2000; Prospero et al., 2002; Washington et al., 2003; Schepanski et al., 2012).

The nutrients concentration in the Mediterranean surface waters is normally very low (Ribera d'Alcalà et al., 2003) and the dust deposition is recognized as a significant source of micro- and macro- nutrients to the surface ocean (Jickells et al., 2005; Mahowald et al., 2008; Duce et al., 2009). Nevertheless, when dust addition experiments were carry out , where found that on one hand nutrient dust dissolution can alleviate nutrients starvation but on the other hand the nutrients limitation were shift to another nutrient (Bonnet et al., 2005).

So to deeply analyze the effects of nutrients dust dissolution and their effects on low nutrients waters we need synoptic data in space and time of both, dust nutrient content and water nutrient concentration. Without one of these variable we are no able o determinate which nutrient contained in dust produced the phytoplankton stimulation, but we are able to determinate if dust dissolution inhibit or stimulate the phytoplankton community.

The larger scope

In the last two centuries and half, global change has caused important climate change, as desertification, ocean acidification, ozone depletion, pollution, and other large-scale shifts.
For this reason, a part of scientific community is focused on trying to predict future changes. When the expected systems alterations for the future and the possible scenarios are contemplated, the climate alteration is one of the most important environmental changes. In this context we refer especially to global warming. On the broad scale, when we talk about global warming, we are talking about rise in the average temperature of the Earth's climate system and its related effects.
Some of the consequences in the Earth rising temperature are: the increase in evaporation, the reduction in precipitations, the increase in extent of arid areas and severity of desertification, as well as the increase in temperature of ocean superficial waters with consequent salinity modification and ph decrease. In the IPCC (2014) report were showed data and projection about the weather parameters will be modified due to climate change. As show in figure 6, it is expected that the surface temperature rise in mid-latitude, while the mean precipitation will likely decrease whit an increase in ocean acidification. Furthermore, it is estimated that the desertification currently affects about

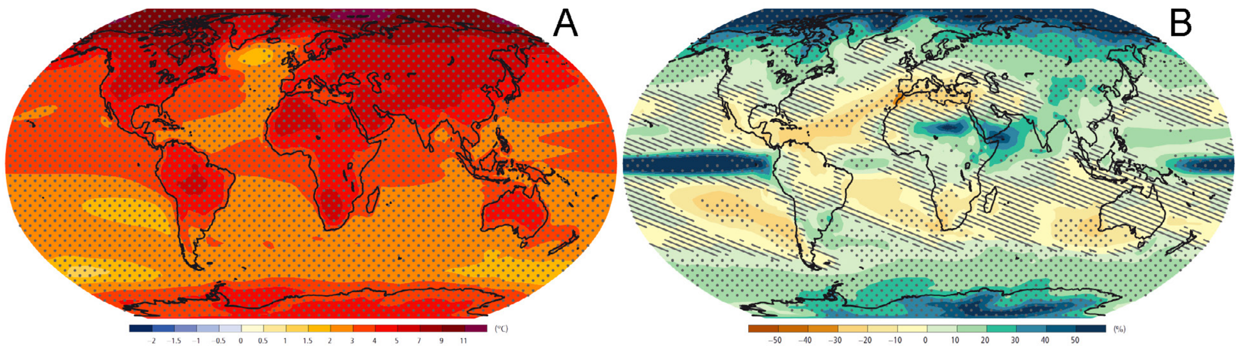

Figure 6: Predicted changes in average surface temperature (A) and changes in average precipitation (B) for the period 2081–2100 (from IPCC (2014)).

the 25% of the world's surface. The global area affected by severe erosion is equal to 1047 Mha-9 of which, 267 Mha-9 are located in Africa and 132 Mha-9 in Europe (Lal, 2003). Also, about 10 Mha-9 yr-1 are lost due to soil erosion (Pimentel, 2006), assuming a conservative rate of erosion expansion of 0.5% yr-1 (Lewis et al., 2013) corresponding to an annual increment of desertified area of 80^103 km2 . If the world's desert areas expand, at the same time the dust particles load in the atmosphere will increase.

The dust particles present into the atmosphere may prompt changes in the atmosphere and may have important consequences in the processes affecting the clime as well as they could affect and alter some ocean properties.
To complicate the situation, according to the climatic models further predictions the Mediterranean basin will be one of the regions most affected by the warming trend and by an increase in extreme events. This makes the Mediterranean a potential model of more global patterns that occur in the world's marine biota. There are reasons to believe that the Mediterranean is already one of the most impacted seas in the world, this because climate change and other disturbances patterns interacts synergistically (Lejeusne et al., 2010).

Aims of thesis

The aim of this thesis is to study the possible ecosystem stimulating effects of Saharan dust deposition in the Mediterranean Sea surface waters.
We focus our research to describe the patterns of deposition both geographically and seasonally and to relate such deposition to chlorophyll, as a proxy for ecosystem production, dynamics in the ocean.
The specific objectives of the thesis were:
1. to define the patterns of modeled dust deposition in the Mediterranean both geographically and seasonally.
2. to define the differences in the patterns of dust transport and deposition.
3. to quantify the degree of relationship between modeled dust deposition and surface chlorophyll
4. to quantify the effect of case studies of large deposition events on chlorophyll dynamics in the Mediterranean.

The studies conducted for this thesis are reported in three chapters (2-4), each of which addresses some of the above-defined specific objectives. They are structured as scientific papers because some of them are already published. Finally, this thesis ends with a general conclusions chapter.

CHAPTER 2: Mediterranean basin-wide correlations between Saharan dust deposition and ocean chlorophyll concentration. Biogeosci. Disc, 2012, 9, 8611-8639.
R. Gallisai, F. Peters, S. Basart, and J. M. Baldasano

Weekly satellite chlorophyll concentration data and dust deposition modeled data from 2000 to 2008 were investigated at the Mediterranean basin scale to describe the spatial and temporal patterns of dust deposition and to find relationships between the dynamics of both variables. Positive annual cross correlation values were found, especially in the Eastern Mediterranean. Seasonally detrended data showed high responsiveness in the Western and in the Central Mediterranean.

CHAPTER 3: Saharan dust deposition may affect phytoplankton growth in the Mediterranean Sea at ecological time scales. PLoS ONE, 2014.
Gallisai, R., Peters, F., Volpe, G., Basart, S., and Baldasano, J. M.

Improved satellite chlorophyll data using an optimized regional algorithm and a series of data filters were applied to relate chlorophyll data to modeled Saharan dust deposition data. Dust deposition can significantly explain between 1 and 10% of seasonally detrended chlorophyll variability. The main positive effects occur during spring in the Eastern and Central Mediterranean. A few negative correlations are found in coincidence of regions under anthropogenic aerosol influence in the northwest Mediterranean and especially in the Aegean Sea.

CHAPTER 4: Large Saharan dust storms: implications for chlorophyll dynamics in the Mediterranean Sea. Submitted to Global Biogeochemical Cycles
Rachele Gallisai, Gianluca Volpe, and Francesc Peters

We defined large and very large dust deposition events occurred between 2000 and 2007 based on their large dust deposition load and their surface extension. A total of 153 large and 31 very large deposition events were identified during this time period. The short-term chlorophyll response to atmospheric dust stimulation was investigated for the very large dust deposition events. Trends showed larger effects in winter and autumn compared to spring and, in seasonally-detrended chlorophyll data, a decrease with longitude to the East. The role of heterotrophic bacteria, competing for limiting nutrients, emerges out from the observed patterns.

Gallisai, R., Peters, F., Basart, S., and Baldasano, J. M., (2012).
Mediterranean basin-wide correlations between Saharan dust deposition and ocean chlorophyll concentration. Biogeosci. Disc., 9, 28, doi:10.5194/bgd-9-8611-2012.

Abstract

The fertilizing potential of atmospheric deposition on ocean production in the Mediterranean is a matter of debate. In this study, eight years (from 2000 to 2007) of weekly chlorophyll concentration data derived from SeaWiFS satellite observations and dust deposition data provided by the BSC-DREAM8b model are investigated in a basin-wide scale in the Mediterranean Sea to describe the geographical distribution and dynamics of both variables and to find potential relationships between them. In all analyses the largest positive cross correlation values are found with a time lag of 0 8-d periods. The coupling between annual cycles of chlorophyll and dust deposition may on average explain an 11.5% in chlorophyll variation in a large part of the Mediterranean. The Eastern Mediterranean shows the largest annual correlations, while the responsiveness to large events is small. The contrary is true for the Western and Northwestern Mediterranean where, if anything, only large events may add to the chlorophyll variability. The Central Mediterranean shows the highest responsiveness of chlorophyll to mineral dust deposition with annual contributions from seasonal variability as well as stimulations owing to large events. These results highlight the importance of dust deposition from African and Middle East origin in the potential stimulation of phytoplankton production in the nutrient depleted surface layers of the Mediterranean Sea.

Introduction

The atmosphere and the ocean are dynamic compartments within the Earth system that are constantly interacting with each other. In addition to exchanges of thermal and mechanical energy, ocean and atmosphere exchange a multitude of chemical compounds such as gases and particulate matter that have effects on global biogeochemical cycles and are highly variable in space and time (Bonnet et al., 2005)

Atmospheric dust deposition supplies several macronutrients, such as phosphorus (Bergametti et al., 1992; Migon and Sandroni, 1999; Ridame and Guieu, 2002; Markaki et al., 2003; Guieu et al., 2010; Pulido-Villena et al., 2010), nitrogen (Loÿe-Pilot et al., 1990; Kouvarakis et al., 2001; Herut et al., 1999; Markaki et al., 2010; Sandroni et al., 2007; Bonnet et al., 2005), iron (Bonnet and Guieu, 2006; Theodosi et al., 2010) and silicate (Moreno et al., 2006) to the surface waters of the Mediterranean Sea.

The Mediterranean Basin is considered one of the most oligotrophic marine ecosystems on Earth (Béthoux et al., 1998). During the stratification period, the phytoplankton community is strongly limited by nutrients. In general, it seems that the strongest limiting nutrient is phosphorus, followed by nitrogen (Estrada, 1996; Thingstad et al., 2005).

On the other hand, the Mediterranean atmosphere is subject to a continuous presence of Saharan mineral dust particles (Barnaba and Gobbi, 2004; Antoine and Nobileau, 2006; Banzon et al., 2009). The deposition on surface waters of these particles and soluble compounds, rich in nutrients, may influence biological production. The contribution through atmospheric deposition of nitrogen and phosphorus in the Mediterranean Sea has been shown to be significant (Guerzoni et al., 1999; Migon and Sandroni, 1999; Guieu et al., 2002a; Guieu et al., 2002b; Markaki et al., 2003; Morales-Baquero et al., 2006a), at least during certain events. Therefore, atmospheric deposition is a major potential source through which “new” nutrients, essential for primary production, are deposited on Mediterranean surface waters (Ternon et al., 2011).

Despite the potential contribution of atmospheric nutrients to marine production, evidence of direct effects is hard to find and results are unclear. Previous studies on the effect of Saharan dust inputs on phytoplankton have been restricted to a few specific areas in the Mediterranean basin, having provided contrasting estimates. Different amendment experiments have shown fertilization effects of Saharan dust on both heterotrophic and autotrophic communities (Klein et al., 1997; Ridame and Guieu, 2002; Bonnet et al., 2005; Eker-Develi et al., 2006; Pulido-Villena et al., 2008; Lekunberri et al., 2010; Romero et al., 2011; Ternon et al., 2011). On the other hand, Volpe et al. (2009) found interferences between satellite-derived aerosol optical thickness data and chlorophyll data masking possible functional relationships between both variables.

Aerosol optical thickness (AOT) is a radiometer-based datum that can be used as a proxy of atmospheric turbidity. It is an estimation of the particle load in the air column. Saharan dust transport over the Mediterranean shows a seasonal pattern (Moulin et al., 1998; Barnaba and Gobbi, 2004). The peak of transport over the Eastern Mediterranean occurs in spring, in the Central Mediterranean during the spring-summer period, and in the Western Mediterranean during the summer. While in Antoine and Nobileau (2006) the largest AOT values over the Mediterranean Sea were found in the Eastern basin during spring, then this moves to a lower extent in the Western during summer and in Central in autumn. In any case, atmospheric dust load does not necessarily mean deposition over a certain area. In the Mediterranean Basin, transport of desert dust aerosols occurs at different heights (Mona et al., 2006; Papayannis et al., 2008; Sicard et al., 2011). The winter and spring aerosol plumes that cross the Mediterranean in general travel at low altitude. During the rest of the year the aerosol load tends to travel in higher atmospheric layers reaching 6000m in height (Pappalardo et al., 2003; Bartoli et al., 2005; Papayannis et al., 2008). Thus, in order to relate possible effects of aerosols on plankton dynamics, not only atmospheric load is relevant but also whether aerosols are depositing over the sea.

The aim of this study is to find trends between Saharan dust deposition and surface ocean chlorophyll distribution and dynamics in the Mediterranean Sea using correlational analyses with data from eight years (from 2000 and 2007). Based on the analyses, we identify areas were potential fertilization of primary production from deposition seems related to seasonal dynamics and other areas were potential fertilization seems more event driven.

Materials and methods

Chlorophyll

Chlorophyll concentration data were derived from SeaWiFS (Sea-viewing Wide Field of-view Sensor) satellite radiometer measurements. An 8-d averaged, 9 km resolution 5 product (reprocessing 5) was downloaded from the Giovanni online data system (http://disc.sci.gsfc.nasa.gov/giovanni). We extracted data for 179 different 1º x 1º areas in the Mediterranean Sea (Fig. 1), in order to have a wide coverage of the entire basin, from 2000 to 2007.

Saharan dust deposition

Saharan dust deposition was obtained from the BSC-DREAM8b model (Pérez et al., 2006a; Pérez et al., 2006b). The model simulates or predicts the 3-dimensional field of the dust concentration in the troposphere and takes into account all major processes of dust life cycle, such as dust production, horizontal and vertical diffusion and advection as well as wet and dry deposition. The main

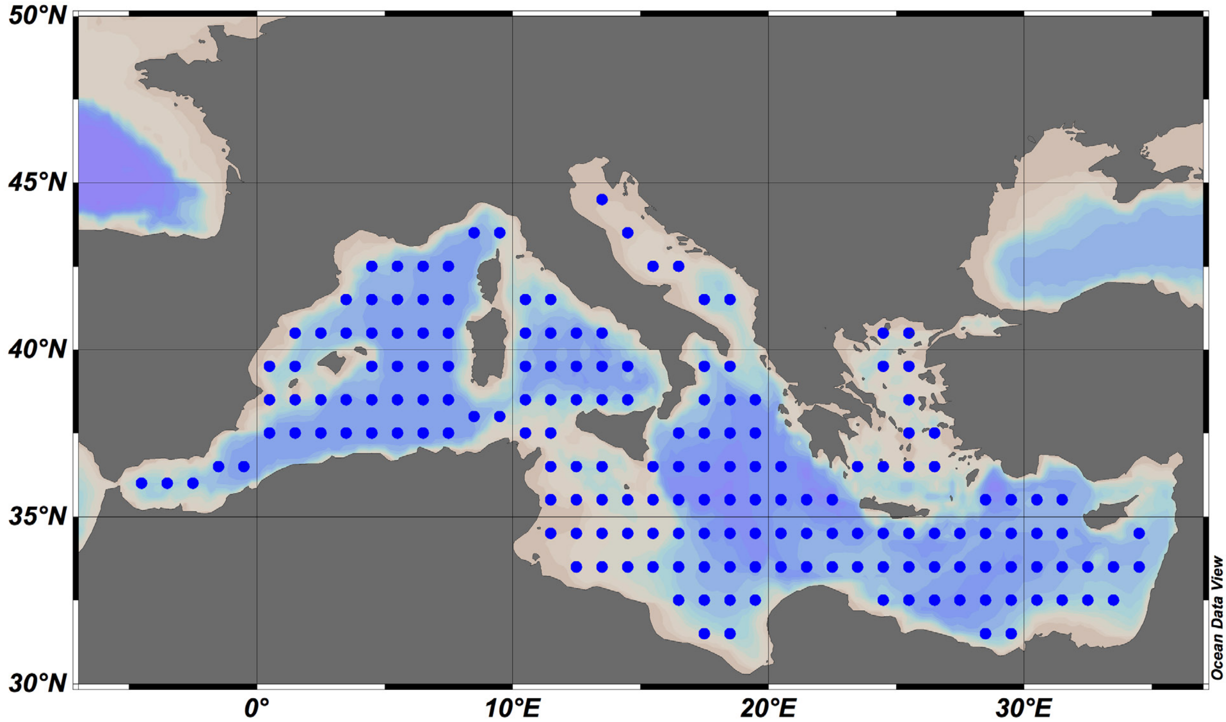

Figure 1. Map that shows the 179 areas, 1° x1° in size, used for the chlorophyll and Saharan dust deposition data analyses.

features of BSC-DREAM8b, described in detail in (Pérez et al., 2006b), are a source function based on the arid and semi-arid categories of the 1 km USGS land use data set, 8 size bins within the 0.1–10 µm radius range according to Tegen and Lacis (1996) are used to describe the size distribution of dust, a source distribution derived from D'Almeida (1987), and the inclusion of dust radioactive feedbacks (Pérez et al., 2006b).

In the last years, the model has been used for dust forecasting and as dust research tools in North Africa and the Mediterranean (Amiridis et al., 2009; Alonso-Perez et al., 2011; Pay et al., 2012). Several case studies have outlined the good skills of BSCDREAM8b (Pérez et al., 2006a; Papanastasiou et al., 2010) concerning both the horizontal and vertical extent of the dust plume in the Mediterranean Basin. Furthermore, daily evaluation of BSC-DREAM8b with near-real time observations is conducted in BSC-CNS. Currently, the daily operational model evaluation includes satellites (MODIS and MSG) and AERONET sun photometers. BSC-DREAM8b has also been validated and tested over longer time periods in the European region (Jiménez-Guerrero et al., 2008; Pay et al., 2010; Basart et al., 2012) and against measurements at source regions for the SAMUM (Haustein et al., 2009) and BODEX campaigns (Todd et al., 2008).

For the present study, a dust simulation of the BSC-DREAM8b model is used for the period between 1 January 2000 and 31 December 2007 over the Mediterranean basin. The resulting daily-accumulated dust deposition fields have been bilineally interpolated to the chlorophyll grid. A 1º x 1º grid between 31º N and 44º N and 5º W and 35º E was used to extract data for the same 179 areas as for chlorophyll.

A low cut-off threshold is applied to the numerical deposition output from BSC-DREAM8b since the dataset showed numerically correct but physically unrealistic low value spikes. Highest deposition rates have been reported in the Eastern Mediterranean (Mamane et al., 1982; Pye, 1992; Kubilay et al., 2000; Singer et al., 2003). Furthermore, minimum deposition values are observed in the northern part of the western Mediterranean. Morales-Baquero et al. (2006b) report a minimum of $3x10^{-5}$ kgm^{-2} d^{-1} in Sierra Nevada (southern Spain) over a 3 yr period. Avila et al. (1997) monitored deposition in the Montseny mountains (Northeastern Spain) during 11 yr and the minimum dust deposition was $1.01x10^{-6}$ kgm^{-2} d^{-1}. De Angelis and Gaudichet (1991) reported a minimum value of $0.6x10^{-6}$ kgm^{-2} d^{-1} over 30 yr in the Alps region. In order to be on the safe side, we chose a value of 10^{-8} kgm^{-2} d^{-1} as the low cut-off threshold for realistic measurable deposition. Finally, daily deposition was log averaged to 8-d periods to match the chlorophyll dataset.

Analyses

Pearson's correlation coefficient (r) and the associated p-level are calculated between dust deposition and chlorophyll concentration for each of the 179 1° x 1º areas. These analyses are done with and without seasonally detrending one or both variables. Previously, we cross-correlated chlorophyll and deposition with lags from 0 up to 4 8-d periods to determine that the highest correlations were found with a lag of 0, that is within a period of 8-d.

Results

The surface chlorophyll distribution shows both an overall west to east decreasing trend and a south to north increasing trend (Fig. 2a). These trends together with a few local spots of higher chlorophyll concentration (e.g. Alboran Sea, the Tunisian coast, Dardanelle strait and the Northern Adriatic) characterize different biogeographical regions in the Mediterranean (D'Ortenzio and Ribera D'Alcalà, 2009). Chlorophyll concentration shows mean concentrations between 0.09mg m^{-3} and 0.13mg m^{-3} in the southern part of the Eastern basin and between 0.23 and 0.36mg m^{-3} in the Northwest Mediterranean. The coefficient of variation (CV) of chlorophyll also shows a major decreasing trend with longitude except for some coastal areas (Fig. 2b). In the Central and Eastern Mediterranean, the chlorophyll CV ranges between 14% to the 20% excepting two outlier regions (near the Tunisian coast and in the Dardanelle strait) with CVs higher than 90%. In the Western Mediterranean, the CV of chlorophyll ranges from 20% to 66% with the highest CV in areas with the largest mean concentration.

Dust deposition distribution shows a decreasing trend from south to north (Fig. 3a). The highest

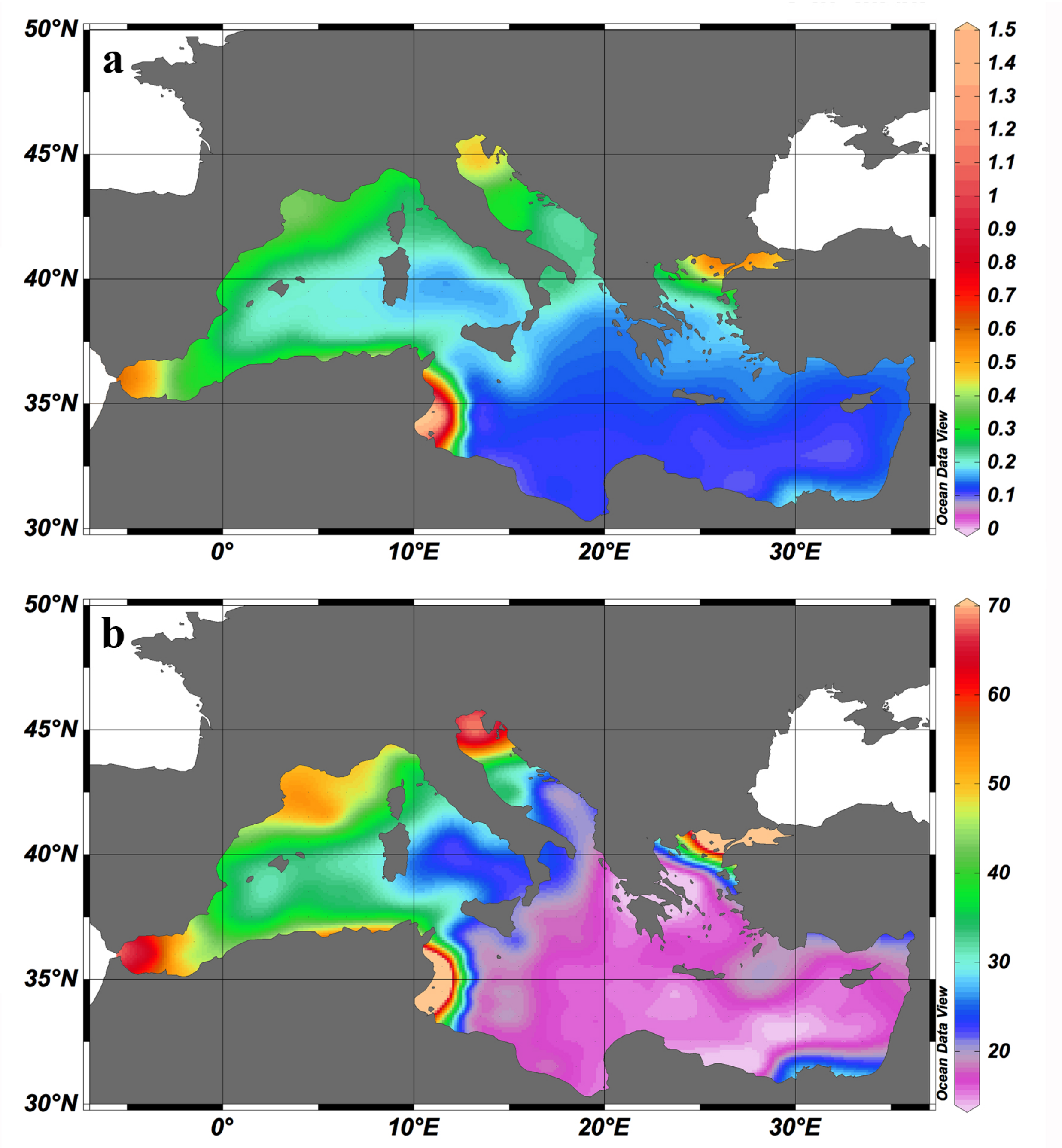

Figure 2. Observed mean surface chlorophyll concentration from SeaWIFS for the Mediterranean Sea **(a)** and the corresponding coefficient of variation **(b)** for the years 2000–2007.

mean concentration values are present in the southern part of the Mediterranean Sea, with mean values ranging between 5°x10^{-7} kgm^{-2} d^{-1} and 1°x 10^{-7} kgm^{-2} d^{-1}. On the other hand, the lowest values are located in the northern part of the Mediterranean Sea (North Spain, South France and North Italy), where the range is from 2°x10^{-8} kgm^{-2} d^{-1} to 3°x10^{-8} kgm^{-2} d^{-1} in mean concentrations.

In contrast to mean dust deposition values a different scenario is represent for the CV of dust deposition. The CV decreased from north to south (Fig. 3b). The smallest CV in dust deposition occurs in the southern part of the Mediterranean basin where it oscillates between 7% and 20 %.

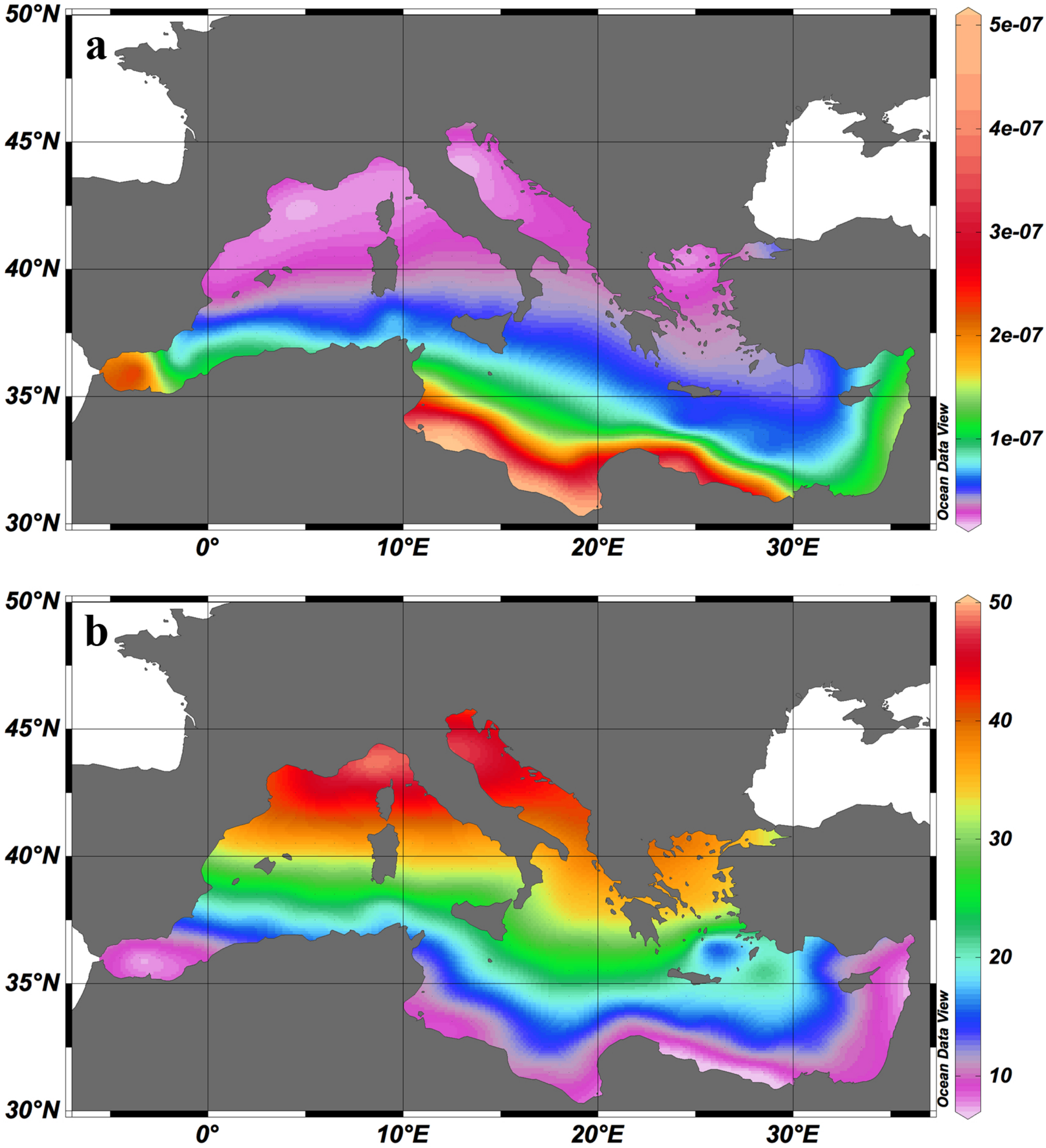

Figure 3. Modeled dust deposition mean value from BSC-DREAM8b for the Mediterranean Sea **(a)** and the corresponding coefficient of variation **(b)** for the years 2000–2007.

Conversely, in the northern part, we found the highest CV in dust deposition, with values close to 50 %.

The seasonal trend of deposition shows the largest values in late autumn-winter for the Central and Eastern Mediterranean (Fig. 4), albeit with a lower overall annual variability than in the Northern-Northwestern Mediterranean. The spatial distribution shows larger deposition close to the African and Middle East sources and a decrease of average monthly deposition to the Northern Mediterranean.

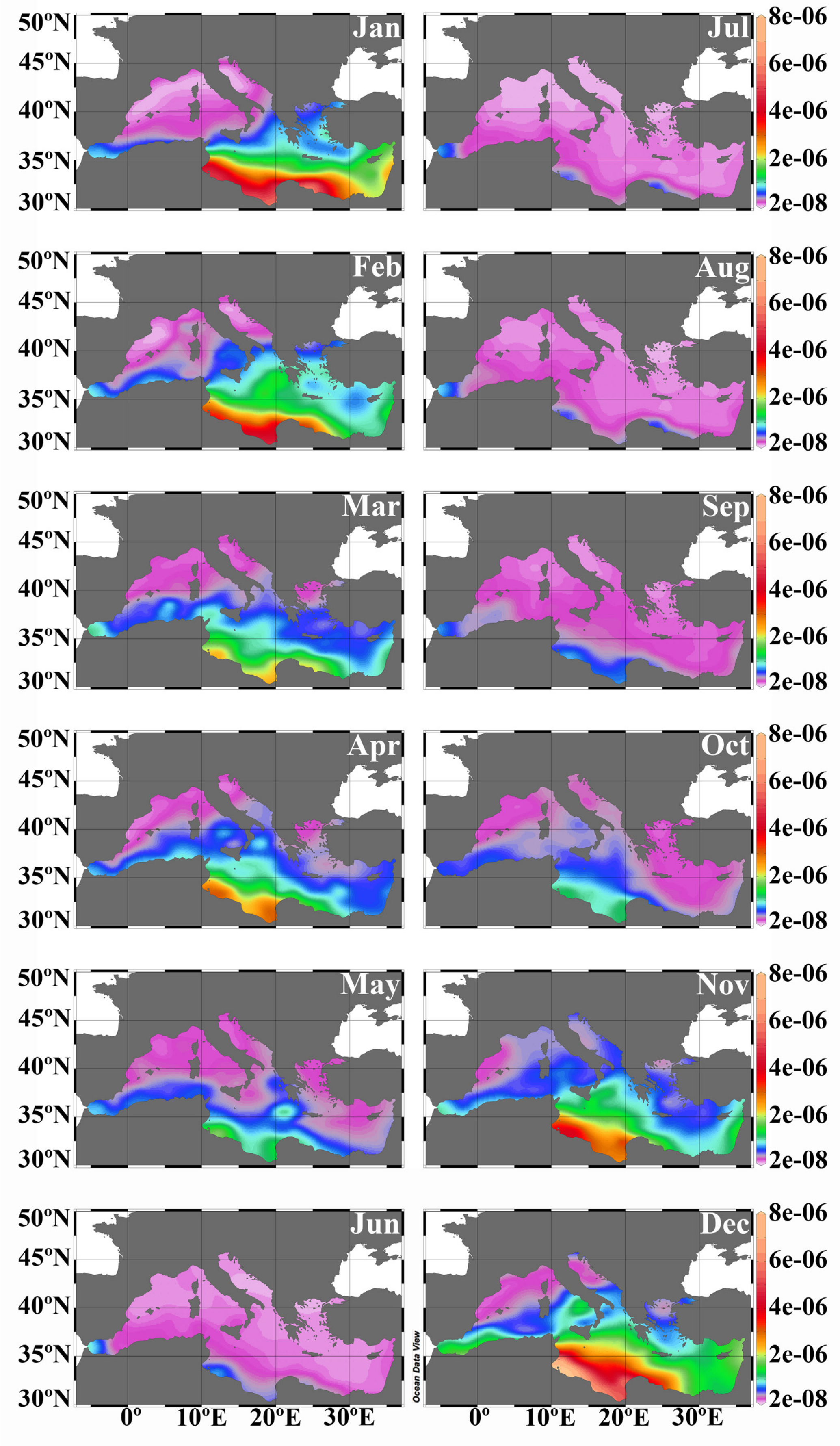

Figure 4. Monthly climatic modeled deposition (2000–2007) in the Mediterranean Sea from BSCDREAM8b.

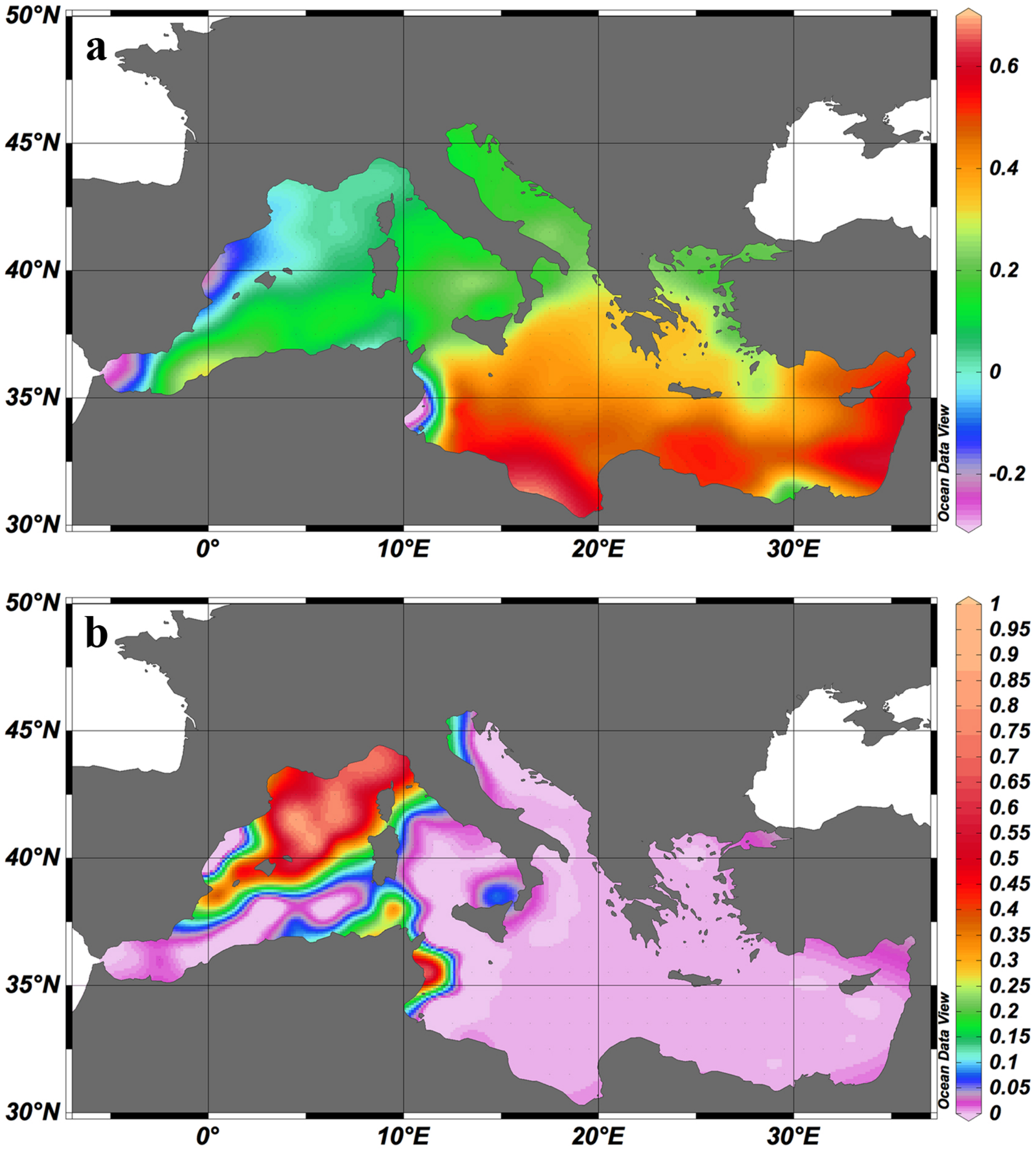

Figure 5. Chlorophyll and dust deposition correlation at time lag 0 **(a)** and the corresponding p-level statistical significance **(b)**.

The correlation between Sahara dust deposition and chlorophyll concentration is shown in Fig. 5a. In the East sub-basin values are between 0.3 and 0.65, with the exception of the west side of the Nile delta where the correlation coefficient was of 0.18 and the Dardanelle area where it was lower than 0.2. Negative correlations occur in the southeastern coast of Tunisia, in the Alboran Sea, in the northeastern coast of Spain and in the Gulf of Lions. Furthermore, the Central Mediterranean Sea shows values between 0 and 0.2. The significance of the correlations is shown in Fig. 5b. In most of the Mediterranean basin, the p-levels were lower than 0.05, with the exception of the Tunisian coast, the Sicily strait, the Northwest Mediterranean and the Northern Adriatic Sea, basically

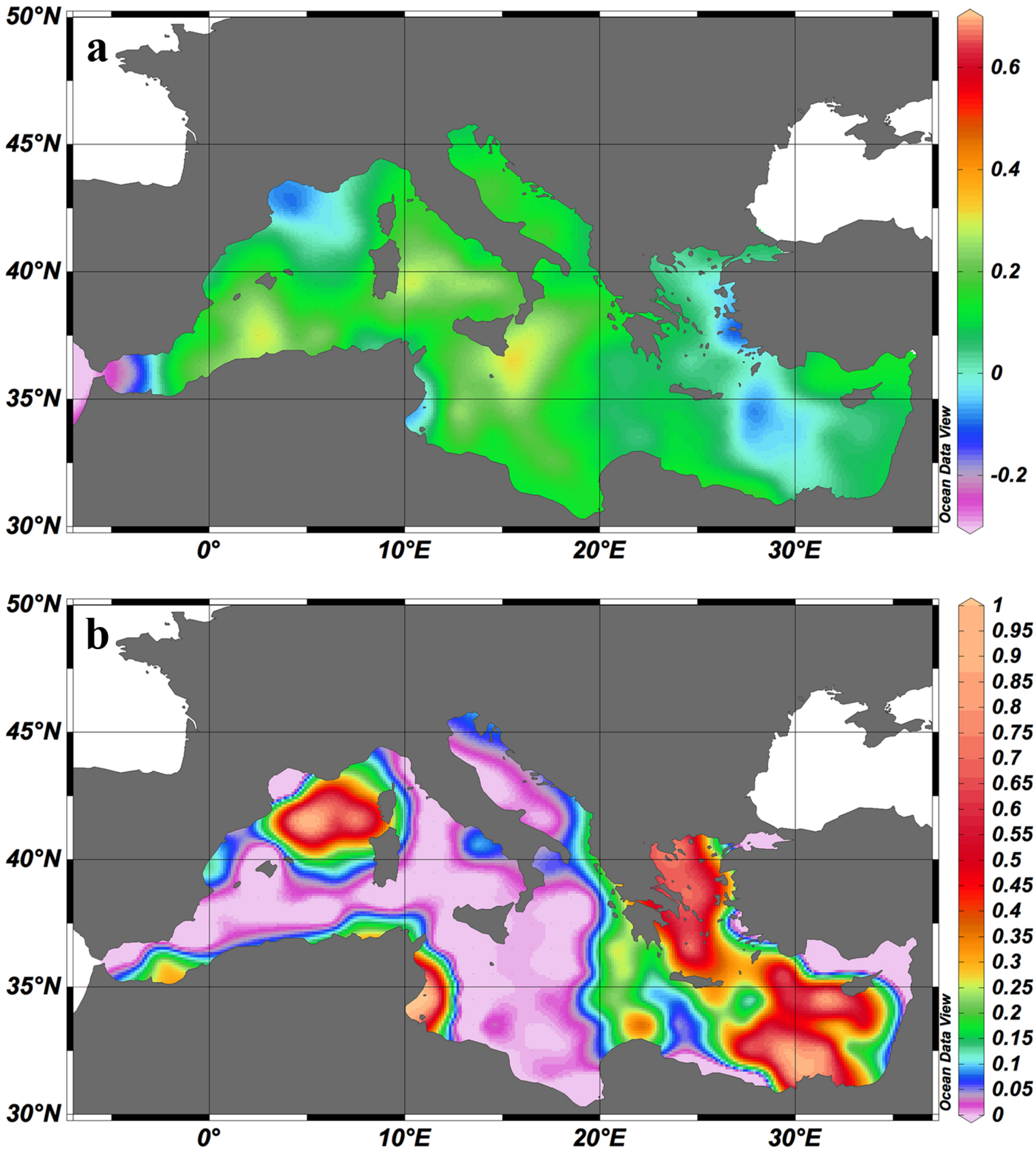

Figure 6. Same as Fig. 5 but for the correlation between seasonally detrended chlorophyll and deposition.

areas that were showing low correlations, either positive or negative.

The correlations were non significant, with p-levels close to 1, between the Ligurian coast and the open water in front of the Gulf of Lions and up to the Menorca.

When deposition is correlated with the seasonally detrended chlorophyll (Fig. 6), correlation coefficients show a general drop mostly to values between 0.1 and 0.2. Three areas show relatively high correlation coefficients (southeast of Sardinia, south of the Balearic islands and south of Sicily). Negative correlation values appear in the Gulf of Lions, in the Alboran Sea, in the southeast

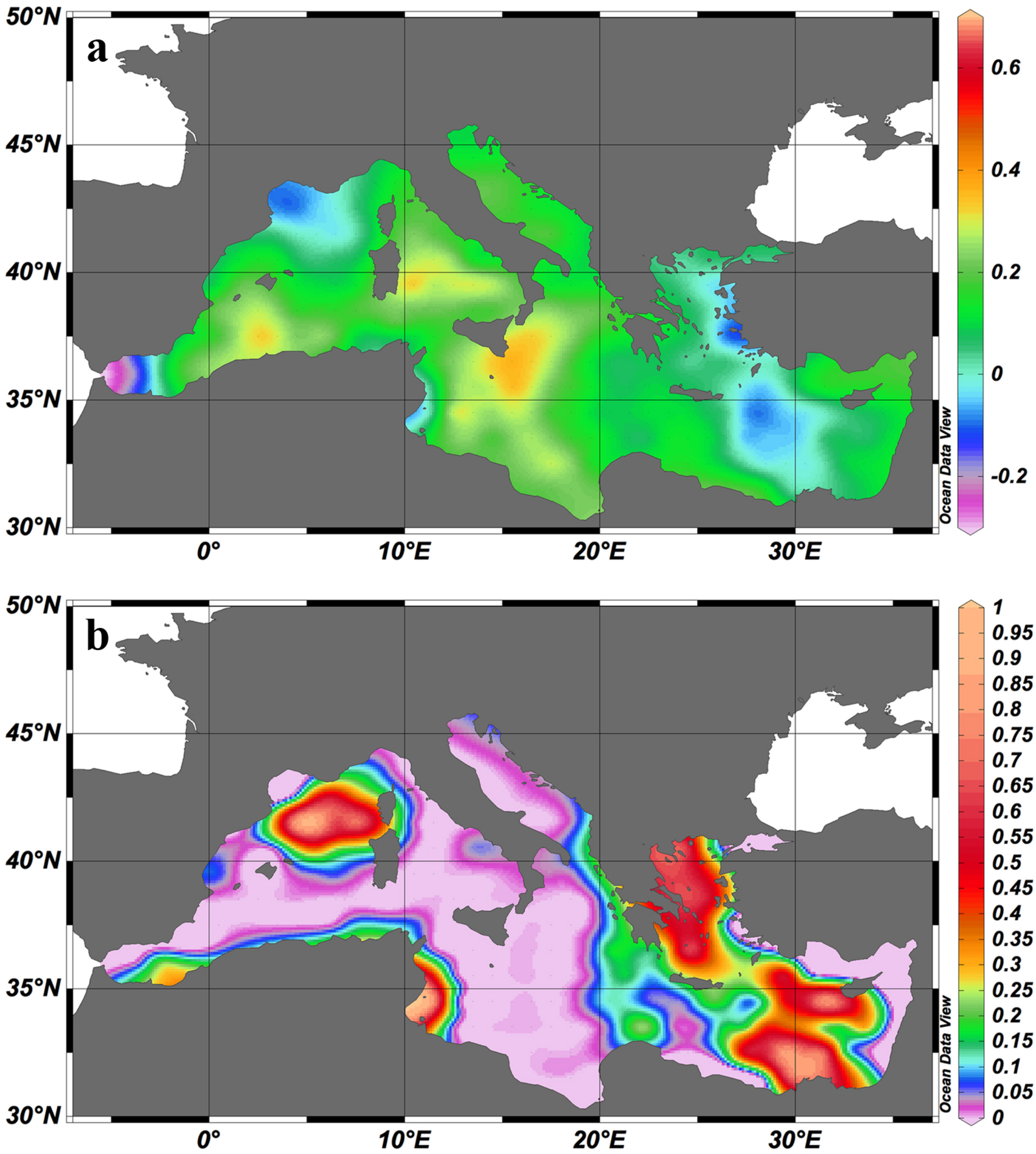

Figure 7. Same as Fig. 5 but for the correlation between seasonally detrended chlorophyll and seasonally detrended deposition.

coast of Tunisia and in the eastern part of Mediterranean Sea. Even if correlation coefficients are low, all the areas with an *r* larger than 0.1 are statistically significant. On the other hand, areas showing a negative *r* are not statistically significant.

Finally, when correlations between deposition and chlorophyll are done after seasonally detrending both data sets (Fig. 7) the results are very similar to the case when only chlorophyll is seasonally detrended. Most areas showed correlation values between 0.1 and 0.2. The highest correlation coefficients were present in the Ionian Sea, in the south Tyrrhenian and in the northern part of Algeria with values between 0.3 and 0.4. The lowest values appeared in the Gulf of Lions, in the

Alboran Sea, in the southeastern coast of Tunisia and in the Levantine Sea, all with a negative correlation coefficient. Again, negative correlation coefficients were not statistically significant. There 5 were 113 areas with p-levels smaller or equal to 0.05, so only 66 areas showed non-significant correlations.

Discussion

Previous studies (Bosc et al., 2004; D'Ortenzio and Ribera D'Alcalà, 2009) had already evidenced that the Mediterranean is not uniform in surface chlorophyll concentrations and temporal dynamics, and our data is very similar to previous more extensive descriptions. On the contrary, aerosol deposition over the sea is much more difficult or currently impossible to map over time with measurements. There are some point measurements obtained during cruises and some more or less permanent stations that measure mostly aerosols in air and not so much deposition, scattered around the Mediterranean coast and some islands (Querol et al., 2009a; Markaki et al., 2010). We have resorted to use synthetic deposition data from an operational aerosol transport model. The BSC-DREAM8b model tracks mineral aerosols from North Africa and Middle East. Other sources, such as of anthropogenic origin and sea spray (Querol et al., 2009b), also contribute to aerosols but have not been considered in this study.

The spatial distribution and temporal dynamics of our deposition data shows considerable and more constant deposition rate close to the mineral origin source. A decreasing gradient is present from south to north. In addition, in the North Mediterranean the temporal variability is larger indicating a shift from a more constant annual dynamics to an exposure to large events. Aerosol deposition is favored in the winter and spring, when the prevailing meteorological conditions make dust plumes travel close to the ocean surface (Alpert et al., 2004). For the remainder of the year, dust plumes in general travel high in the atmosphere over the Mediterranean with reduced chances of deposition over the sea.

Model deposition data seems robust. Papayannis et al. (2008) found a good agreement between lidar observations and BSC-DREAM8b model calculations. Pay et al. (2012) and Basart et al. (2012) found a good agreement between measured PM10 levels at surface stations and BSC-DREAM8b model calculations. The Saharan dust deposition patterns (seasonal maps) shown in this paper are in a good agreement with Basart et al. (2012). Markaki et al. (2010) observed that the maximum in DIN deposition in the Mediterranean basin was in winter, and about 65% of DIP deposition occurred in winter as well, in agreement with our modeled deposition data. Dust deposition is subject to large-scale weather features (such as particularly dry years, latitudinal position of the inter-tropical convergence zone, etc.), which can lead to large interannual fluctuations. High deposition rates are also highly dependent on rains, that is, wet deposition (Kubilay et al., 2000; Ridame and Guieu, 2002). Guerzoni et al. (1997) estimated that about 3/4 of Saharan dust is deposited with precipitation. Thus, the annual cycles of deposition show more variability than other variables, but it is clear that these deposition patterns do not completely match AOT data (Moulin et al., 1998; Barnaba and Gobbi, 2004; Antoine and Nobileau, 2006). The fact that we do not consider the deposition of aerosols other than of North African and Middle East mineral origin may also increase the discrepancy between AOT patterns and modeled deposition data. Surface ocean chlorophyll and dust deposition are highly correlated in the Eastern Mediterranean where it seems that the annual cycles of both variables have a higher match. When seasonally detrended data is used, the correlation is decreased and is mostly non-significant. Thus the Eastern Mediterranean would benefit from a background supply of nutrients from deposition that may fuel the system to some extend while large dust deposition events have a minor role fueling primary production peaks in the area. In the Northwestern Mediterranean deposition tends to be much lower and more events driven, and most likely the higher background water nutrients and chlorophyll levels preclude an effect from deposited material most of the times. A recent study (Izquierdo et al., 2012) shows that atmospheric-derived P normally contributes less than 1% of primary production in the NW Mediterranean but occasional events may contribute up to a 30% of annual new production.

The Central Mediterranean, taken here loosely between 10º E and 20º E, stands out as an area where both the relatively high background deposition can add to the fueling of the system over the annual cycle and where single large events may also alter significantly the dynamics of chlo-

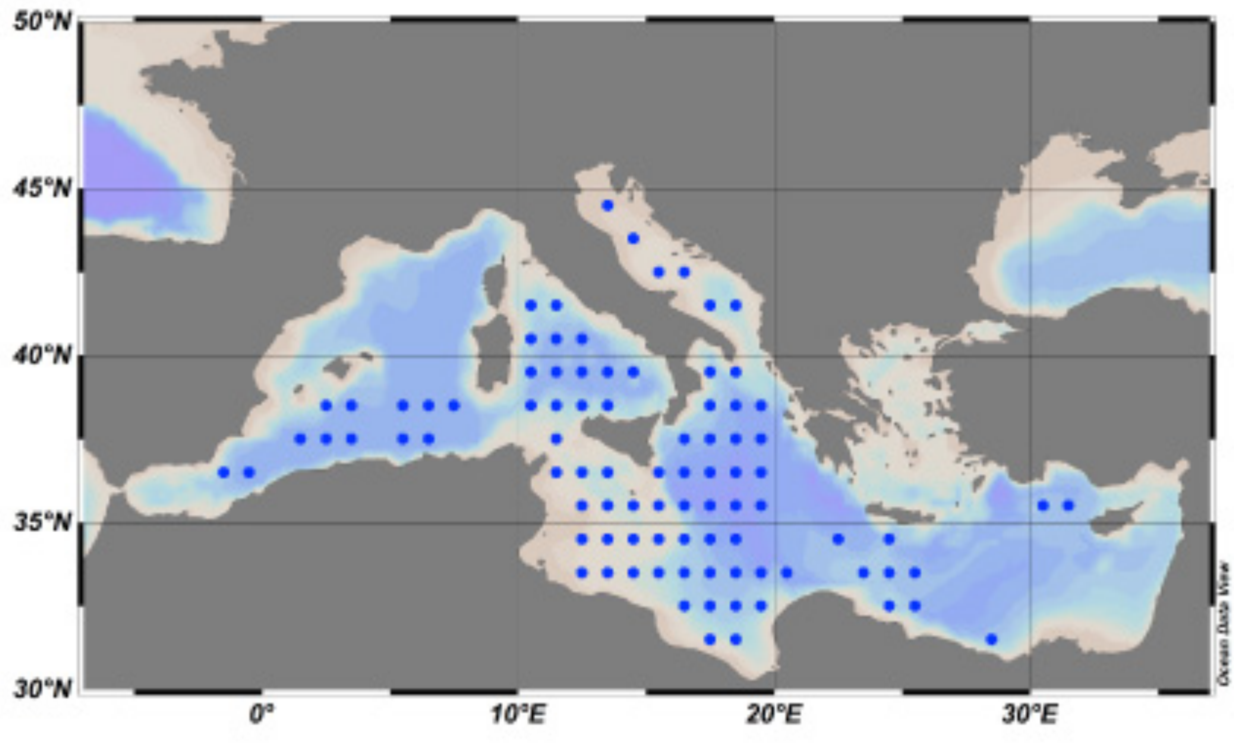

Figure 8. Map that shows the 95 areas with positive and statistically significant cross correlations, in both chlorophyll vs dust deposition and seasonally detrended chlorophyll vs seasonally detrended deposition.

rophyll. There are 95 areas, coinciding mostly with the Central Mediterranean, where correlations between deposition and chlorophyll are significant when using seasonally detrended as well as and non-detrended data (Fig. 8). In these areas the coupling between annual cycles of both variables was responsible of 11.5% of the annual chlorophyll variability. Moreover, if we detrended the seasonal patterns of both variables, dust deposition events were responsible for 4.6% of the variability in chlorophyll. We could distinguish three different zones: one between the Balearic islands and the north African coast, one corresponding to the Tyrrhenian Sea and one corresponding to the Central Mediterranean proper, between Southern Italy, Sicily and the Libyan coast. South of the Balearic Islands and the Tyrrhenian Sea lie the areas most susceptible of event-driven dust deposition, explaining 6% of the seasonally-detrended variability in chlorophyll. The Central Mediterranean proper is where the annual matching between deposition and chlorophyll explains on average the largest proportion in chlorophyll variability, 19% (Table 1).

As mentioned earlier, cross-correlations between dust deposition and chlorophyll show its maximum at lag 0, that is, within the first 8-d, as was also found by Volpe et al. (2009) for AOT and chlorophyll. Because of the potential signal interference of AOT with chlorophyll reported by

	Chl vs dust	**Chl vs dust (detrended)**
All 95 areas	11.5%	4.6%
South Baleares	1.8%	6.0%
Tyrrhenian Sea	3.0%	6.0%
Central Mediterranean	18.7%	5.2%

Table 1. Results of dust deposition impact on chlorophyll variability

these authors we shy away from a higher temporal resolution that may give spurious correlations. In the areas where our correlations are significant, the absolute correlation values do not exceed 0.65 at the most. This means that the relationship would explain a maximum of 42% of the temporal variation in chlorophyll, and in most cases around 2 %. Of course, nutrients being fueled from rich bottom waters, either through a large annual overturning or through more steady diffusion, is the main driver of phytoplankton in the upper layers. The load of nutrients added through atmospheric deposition at a certain time tends to be small compared to the enrichment from bottom waters, and thus significant correlations, even is small, are highly relevant. In addition it has to be taken into account that small nutrient additions in oligotrophic waters tend to favor first competing bacteria (Pulido-Villena et al., 2008; Lekunberri et al., 2010; Marañón et al., 2010; Romero et al., 2011) and that phytoplankton should then respond to the recycled nutrients. Thus, even in experiments, while responses in activity occur, it is not always straightforward to see responses at the biomass level. Finally, we have only considered deposition of North African and Middle East, mineral sources while we expect that at least in some cases, in some areas or for some events the contribution of other sources, for instance of anthropogenic origin, may also explain part of the seasonally-detrended chlorophyll series.

Conclusions

An eight years (2000–2007) analysis of SeaWIFS and BSC-DREAM8b model data, shows that mineral dust deposition from North African and Middle East desert dust sources correlates with chlorophyll in fairly large areas of the Mediterranean. This is especially true for the Eastern Mediterranean where aerosol deposition dynamics matches chlorophyll annual dynamics and the atmospheric input may be an intrinsic part of ecosystem dynamics. However, the Eastern Mediterranean shows little responsiveness to large dust outbreaks, perhaps because they travel over this area to distances further away. For most of the Western Mediterranean a coupling between the dynamics of both variables is weak or inexistent. This seems to be due to the presence of some relatively rich hot spot areas such as the Gulf of Lions and the Alboran Sea, where other factors are mostly in control of nutrient and chlorophyll dynamics. Only in the South Tyrrhenian and in the northern part of Algeria, a positive connection between the dust outbreaks and chlorophyll is present. Overall, the area most responsive to dust deposition is the Central Mediterranean, with both an annual match in the timings and a response to dust outbreaks. It is possible that previous studies between chlorophyll and aerosols in air showed less obvious correlations because not all aerosols in the atmosphere are depositing and many aerosol plumes travel at great heights during most of the year with little chance of deposition.

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References

Alonso-Perez, S., Cuevas, E., Perez, C., Querol, X., Baldasano, J. M., Draxler, R., and De Bustos, J. J., (2011). Trend changes of African airmass intrusions in the marine boundary layer over the subtropical Eastern North Atlantic region in winter. Tellus (B Chem. Phys. Meteorol.), 63, 255-265, doi:10.1111/j.1600-0889.2010.00524.x.

Alpert, P., Kishcha, P., Shtivelman, A., Krichak, S. O., and Joseph, J. H., (2004). Vertical distribution of Saharan dust based on 2.5-year model predictions. Atmospheric Research, 70, 109-130, doi:10.1016/j.atmosres.2003.11.001.

Amiridis, V., Kafatos, M., Perez, C., Kazadzis, S., Gerasopoulos, E., Mamouri, R. E., Papayannis, A., Kokkalis, P., Giannakaki, E., Basart, S., Daglis, I., and Zerefos, C., (2009). The potential of the synergistic use of passive and active remote sensing measurements for the validation of a regional dust model. Ann. Geophys., 27, 3155-3164, doi:10.5194/angeo-27-3155-2009.

Antoine, D., and Nobileau, D., (2006). Recent increase of Saharan dust transport over the Mediterranean Sea, as revealed from ocean color satellite (SeaWiFS) observations. J. Geophys. Res. (Atmos.), 111, doi:10.1029/2005JD006795.

Avila, A., QueraltMitjans, I., and Alarcon, M., (1997). Mineralogical composition of African dust delivered by red rains over northeastern Spain. J. Geophys. Res. (Atmos.), 102, 21977-21996, doi:10.1029/97jd00485.

Banzon, V. F., Gordon, H. R., Kuchinke, C. P., Antoine, D., Voss, K. J., and Evans, R. H., (2009). Validation of a SeaWiFS dust-correction methodology in the Mediterranean Sea: Identification of an algorithm-switching criterion. Remote Sens. Environ., 113, 2689-2700, doi:10.1016/j.rse.2009.08.007.

Barnaba, F., and Gobbi, G. P., (2004). Aerosol seasonal variability over the Mediterranean region and relative impact of maritime, continental and Saharan dust particles over the basin from MODIS data in the year 2001. Atmos. Chem. Phys., 4, 2367-2391, doi:10.5194/acp-4-2367-2004.

Bartoli, G., Migon, C., and Losno, R., (2005). Atmospheric input of dissolved inorganic phosphorus and silicon to the coastal northwestern Mediterranean Sea: Fluxes, variability and possible impact on phytoplankton dynamics. Deep-Sea Res. Part I., 52, 2005-2016, doi:10.1016/j.dsr.2005.06.006.

Basart, S., Pay, M. T., Jorba, O., Pérez, C., Jiménez-Guerrero, P., Schulz, M., and Baldasano, J. M., (2012). Aerosols in the CALIOPE air quality modelling system: Evaluation and analysis of PM levels, optical depths and chemical composition over Europe. Atmos. Chem. Phys., 12, 3363-3392, doi:10.5194/acp-12-3363-2012.

Bergametti, G., Remoudaki, E., Losno, R., Steiner, E., Chatenet, B., and Buatmenard, P., (1992). Source, transport and deposition of atmospheric phosphorus over the northwestern Mediterranean. J. Atmos. Chem., 14, 501-513, doi:10.1007/bf00115254.

Béthoux, J. P., Morin, P., Chaumery, C., Connan, O., Gentili, B., and Ruiz-Pino, D., (1998). Nutrients in the Mediterranean Sea, mass balance and statistical analysis of concentrations with respect to environmental change. Mar. Chem., 63, 155-169, doi:10.1016/S0304-4203(98)00059-0.

Bonnet, S., Guieu, C., Chiaverini, J., Ras, J., and Stock, A., (2005). Effect of atmospheric nutrients on the autotrophic communities in a low nutrient, low chlorophyll system. Limnol. Oceanogr., 50, 1810-1819, doi:10.4319/lo.2005.50.6.1810.

Bonnet, S., and Guieu, C., (2006). Atmospheric forcing on the annual iron cycle in the western Mediterranean Sea: A 1-year survey. J. Geophys. Res. (Oceans), 111, C09010, doi:10.1029/2005jc003213.

Bosc, E., Bricaud, A., and Antoine, D., (2004). Seasonal and interannual variability in algal biomass and primary production in the Mediterranean Sea, as derived from 4 years of SeaWiFS observations. Global. Biogeochem. Cy., 18, GB1005, doi:10.1029/2003GB002034.

D'Almeida, G. A.: Desert aerosol characteristics and effects on climate, in: Paleoclimatology and Paleometeorology : Modern and Past Patterns of Global Atmospheric Transport, edited by: Leinen, M., and Sarnthein, M., Kluwer Academic Publ, Dordrecht, 311-338, 1987.

D'Ortenzio, F., and Ribera D'Alcalà, M., (2009). On the trophic regimes of the Mediterranean Sea: A satellite analysis. Biogeosciences, 6, 139-148, doi:10.5194/bg-6-139-2009.

De Angelis, M., and Gaudichet, A., (1991). Saharan dust deposition over Mont-Blanc (French Alps) during the last 30 years. Tellus (B Chem. Phys. Meteorol.), 43, 61-75, doi:10.1034/j.1600-0889.1991.00005.x.

Eker-Develi, E., Kideys, A. E., and Tugrul, S., (2006). Role of Saharan dust on phytoplankton dynamics in the northeastern Mediterranean. Mar. Ecol. Prog. Ser., 314, 61-75, doi:10.3354/meps314061.

Estrada, M., (1996). Primary production in the northwestern Mediterranean. Scientia Marina, 60, 55-64.

Guerzoni, S., Molinaroli, E., and Chester, R., (1997). Saharan dust inputs to the western Mediterranean Sea: Depositional patterns, geochemistry and sedimentological implications. Deep-Sea Res. Part II., 44, 631-654, doi:10.1016/S0967-0645(96)00096-3.

Guerzoni, S., Chester, R., Dulac, F., Herut, B., Loÿe-Pilot, M. D., Measures, C., Migon, C., Molinaroli, E., Moulin, C., Rossini, P., Saydam, C., Soudine, A., and Ziveri, P., (1999). The role of atmospheric deposition in the biogeochemistry of the Mediterranean Sea. Prog. Oceanogr., 44, 147-190, doi:10.1016/S0079-6611(99)00024-5.

Guieu, C., Bozec, Y., Blain, S., Ridame, C., Sarthou, G., and Leblond, N., (2002a). Impact of high Saharan dust inputs on dissolved iron concentrations in the Mediterranean Sea. Geophys. Res. Lett., 29, 1911, doi:10.1029/2001GL014454.

Guieu, C., Loÿe-Pilot, M. D., Ridame, C., and Thomas, C., (2002b). Chemical characterization of the Saharan dust end-member: Some biogeochemical implications for the western Mediterranean Sea. J. Geophys. Res. (Atmos.), 107, doi:10.1029/2001JD000582.

Guieu, C., Loÿe-Pilot, M. D., Benyahya, L., and Dufour, A., (2010). Spatial variability of atmospheric fluxes of metals (Al, Fe, Cd, Zn and Pb) and phosphorus over the whole Mediterranean from a one-year monitoring experiment: Biogeochemical implications. Mar. Chem., 120, 164-178, doi:10.1016/j.marchem.2009.02.004.

Haustein, K., Pérez, C., Baldasano, J. M., Müller, D., Tesche, M., Schladitz, A., Esselborn, M., Weinzierl, B., Kandler, K., and Von Hoyningen-Huene, W., (2009). Regional dust model performance during SAMUM 2006. Geophys. Res. Lett., 36, L03812, doi:10.1029/2008GL036463.

Herut, B., Krom, M. D., Pan, G., and Mortimer, R., (1999). Atmospheric input of nitrogen and phosphorus to the Southeast Mediterranean: Sources, fluxes, and possible impact. Limnol. Oceanogr., 44, 1683-1692, doi:10.4319/lo.1999.44.7.1683.

Izquierdo, R., Benítez-Nelson, C. R., Masqué, P., Castillo, S., Alastuey, A., and Àvila, A., (2012). Atmospheric phosphorus deposition in a near-coastal rural site in the NE Iberian Peninsula and its role in marine productivity. Atmos. Environ., 49, 361-370, doi:10.1016/j.atmosenv.2011.11.007.

Jiménez-Guerrero, P., Pérez, C., Jorba, O., and Baldasano, J. M., (2008). Contribution of Saharan dust in an integrated air quality system and its on-line assessment. Geophys. Res. Lett., 35, L03814, doi:10.1029/2007GL031580.

Klein, C., Dolan, J. R., and Rassoulzadegan, F., (1997). Experimental examination of the effects of rainwater on microbial communities in the surface layer of the NW Mediterranean Sea. Mar. Ecol. Prog. Ser., 158, 41-50, doi:10.3354/meps158041.

Kouvarakis, G., Mihalopoulos, N., Tselepides, A., and Stavrakakis, S., (2001). On the importance of atmospheric inputs of inorganic nitrogen species on the productivity of the Eastern Mediterranean sea. Global. Biogeochem. Cy., 15, 805-817, doi:10.1029/2001GB001399.

Kubilay, N., Nickovic, S., Moulin, C., and Dulac, F., (2000). An illustration of the transport and deposition of mineral dust onto the eastern Mediterranean. Atmos. Environ., 34, 1293-1303, doi:10.1016/S1352-2310(99)00179-X.

Lekunberri, I., Lefort, T., Romero, E., Vázquez-Domínguez, E., Romera-Castillo, C., Marrasé, C., Peters, F., Weinbauer, M., and Gasol, J. M., (2010). Effects of a dust deposition event on coastal marine microbial abundance and activity, bacterial community structure and ecosystem function. J. Plankton Res., 32, 381-396, doi:10.1093/plankt/fbp137.

Loÿe-Pilot, M. D., Martin, J. M., and Morelli, J., (1990). Atmospheric input of inorganic nitrogen to the Western Mediterranean. Biogeochemistry, 9, 117-134, doi:10.1007/BF00692168.

Mamane, Y., Ganor, E., and Donagi, A. E., (1982). Aerosol composition of urban and desert origin in the eastern Mediterranean. II. Deposition of large particles. Water, Air, and Soil Pollution, 18, 475-484, doi:10.1007/BF02426055.

Marañón, E., Fernández, A., Mouriño-Carballido, B., Martínez-García, S., Teira, E., Cermeño, P., Chouciño, P., Huete-Ortega, M., Fernández, E., Calvo-Díaz, A., Morán, X. A. G., Bode, A., Moreno-Ostos, E., Varela, M. M., Patey, M. D., and Achterberg, E. P., (2010). Degree of oligotrophy controls the response of microbial plankton to Saharan dust. Limnol. Oceanogr., 55, 2339-2352, doi:10.4319/lo.2010.55.6.2339.

Markaki, Z., Oikonomou, K., Kocak, M., Kouvarakis, G., Chaniotaki, A., Kubilay, N., and Mihalopoulos, N., (2003). Atmospheric deposition of inorganic phosphorus in the Levantine Basin, eastern Mediterranean: Spatial and temporal variability and its role in seawater productivity. Limnol. Oceanogr., 48, 1557-1568, doi:10.4319/lo.2003.48.4.1557.

Markaki, Z., Loÿe-Pilot, M. D., Violaki, K., Benyahya, L., and Mihalopoulos, N., (2010). Variability of atmospheric deposition of dissolved nitrogen and phosphorus in the Mediterranean and possible link to the anomalous seawater N/P ratio. Mar. Chem., 120, 187-194, doi:10.1016/j.marchem.2008.10.005.

Migon, C., and Sandroni, V., (1999). Phosphorus in rainwater: Partitioning inputs and impact on the surface coastal ocean. Limnol. Oceanogr., 44, 1160-1165, doi:10.4319/lo.1999.44.4.1160.

Mona, L., Amodeo, A., Pandolfi, M., and Pappalardo, G., (2006). Saharan dust intrusions in the Mediterranean area: Three years of Raman lidar measurements. J. Geophys. Res. (Atmos.), 111, D16203, doi:10.1029/2005jd006569.

Morales-Baquero, R., Pulido-Villena, E., and Reche, I., (2006a). Atmospheric Inputs of Phosphorus and Nitrogen to the Southwest Mediterranean Region: Biogeochemical Responses of High Mountain Lakes. Limnol. Oceanogr., 51, 830-837, doi:10.4319/lo.2006.51.2.0830.

Morales-Baquero, R., Pulido-Villena, E., Romera, O., Ortega-Retuerta, E., Conde-Porcuna, J. M., Pérez-Martínez, C., and Reche, I., (2006b). Significance of atmospheric deposition to freshwater ecosystems in the southern Iberian Peninsula. Limnetica, 25, 171-180.

Moreno, T., Querol, X., Castillo, S., Alastuey, A., Cuevas, E., Herrmann, L., Mounkaila, M., Elvira, J., and Gibbons, W., (2006). Geochemical variations in aeolian mineral particles from the Sahara-Sahel Dust Corridor. Chemosphere, 65, 261-270, doi:10.1016/j.chemosphere.2006.02.052.

Moulin, C., Lambert, C. E., Dayan, U., Masson, V., Ramonet, M., Bousquet, P., Legrand, M., Balkanski, Y. J., Guelle, W., Marticorena, B., Bergametti, G., and Dulac, F., (1998). Satellite climatology of African dust transport in the Mediterranean atmosphere. J. Geophys. Res. (Atmos.), 103, 13137-13144, doi:10.1029/98JD00171.

Papanastasiou, D. K., Poupkou, A., Katragkou, E., Amiridis, V., Melas, D., Mihalopoulos, N., Basart, S., Pérez, C., and Baldasano, J. M., (2010). An Assessment of the Efficiency of Dust Regional Modelling to Predict Saharan Dust Transport Episodes. Adv. Meteo., 2010, 154368 doi:10.1155/2010/154368.

Papayannis, A., Amiridis, V., Mona, L., Tsaknakis, G., Balis, D., Bösenberg, J., Chaikovski, A., De Tomasi, F., Grigorov, I., Mattis, I., Mitev, V., Müller, D., Nickovic, S., Pérez, C., Pietruczuk, A., Pisani, G., Ravetta, F., Rizi, V., Sicard, M., Trickl, T., Wiegner, M., Gerding, M., Mamouri, R. E., D'Amico, G., and Pappalardo, G., (2008). Systematic lidar observations of Saharan dust over Europe in the frame of EARLINET (2000-2002). J. Geophys. Res. (Atmos.), 113, D10204, doi:10.1029/2007JD009028.

Pappalardo, G., Amodeo, A., Amoruso, S., Mona, L., Pandolfi, M., and Cuomo, V., (2003). One year of tropospheric lidar measurements of aerosol extinction and backscatter. Annals of Geophysics, 46, 401-414, doi:10.4401/ag-3412.

Pay, M. T., Piot, M., Jorba, O., Gassó, S., Gonçalves, M., Basart, S., Dabdub, D., Jiménez-Guerrero, P., and Baldasano, J. M., (2010). A full year evaluation of the CALIOPE-EU air quality modeling system over Europe for 2004. Atmos. Environ., 44, 3322-3342, doi:10.1016/j.atmosenv.2010.05.040.

Pay, M. T., Jiménez-Guerrero, P., Jorba, O., Basart, S., Querol, X., Pandolfi, M., and Baldasano, J. M., (2012). Spatio-temporal variability of concentrations and speciation of particulate matter across Spain in the CALIOPE modeling system. Atmos. Environ., 46, 376-396, doi:10.1016/j.atmosenv.2011.09.049.

Pérez, C., Nickovic, S., Baldasano, J. M., Sicard, M., Rocadenbosch, F., and Cachorro, V. E., (2006a). A long Saharan dust event over the western Mediterranean: Lidar, Sun photometer observations, and regional dust modeling. J. Geophys. Res. (Atmos.), 111, D15214, doi:10.1029/2005JD006579.

Pérez, C., Nickovic, S., Pejanovic, G., Baldasano, J. M., and Özsoy, E., (2006b). Interactive dust-radiation modeling: A step to improve weather forecasts. J. Geophys. Res. (Atmos.), 111, D16206, doi:10.1029/2005JD006717.

Pulido-Villena, E., Wagener, T., and Guieu, C., (2008). Bacterial response to dust pulses in the western Mediterranean: Implications for carbon cycling in the oligotropic ocean. Global. Biogeochem. Cy., 22, GB1020, doi:10.1029/2007GB003091.

Pulido-Villena, E., Rerolle, V., and Guieu, C., (2010). Transient fertilizing effect of dust in P-deficient LNLC surface ocean. Geophys. Res. Lett., 37, L01603, doi:10.1029/2009gl041415.

Pye, K., (1992). Aeolian dust transport and deposition over Crete and adjacent parts of the Mediterranean Sea. Earth Surface Processes & Landforms, 17, 271-288, doi:10.1002/esp.3290170306.

Querol, X., Alastuey, A., Pey, J., Cusack, M., Perez, N., Mihalopoulos, N., Theodosi, C., Gerasopoulos, E., Kubilay, N., and Kocak, M., (2009a). Variability in regional background aerosols within the Mediterranean. Atmos. Chem. Phys., 9, 4575-4591, doi:10.5194/acp-9-4575-2009.

Querol, X., Pey, J., Pandolfi, M., Alastuey, A., Cusack, M., Pérez, N., Moreno, T., Viana, M., Mihalopoulos, N., Kallos, G., and Kleanthous, S., (2009b). African dust contributions to mean ambient PM10 mass-levels across the Mediterranean Basin. Atmos. Environ., 43, 4266-4277, doi:10.1016/j.atmosenv.2009.06.013.

Ridame, C., and Guieu, C., (2002). Saharan input of phosphate to the oligotrophic water of the open western Mediterranean sea. Limnol. Oceanogr., 47, 856-869, doi:10.4319/lo.2002.47.3.0856.

Romero, E., Peters, F., Marrasé, C., Guadayol, T., Gasol, J. M., and Weinbauer, M. G., (2011). Coastal Mediterranean plankton stimulation dynamics through a dust storm event: An experimental simulation. Estuar. Coast. Shelf Sci., 93, 27-39, doi:10.1016/j.ecss.2011.03.019.

Sandroni, V., Raimbault, P., Migon, C., Garcia, N., and Gouze, E., (2007). Dry atmospheric deposition and diazotrophy as sources of new nitrogen to northwestern Mediterranean oligotrophic surface waters. Deep-Sea Res. Part I., 54, 1859-1870, doi:10.1016/j.dsr.2007.08.004.

Sicard, M., Rocadenbosch, F., Reba, M. N. M., ComerÓn, A., Tomás, S., García-Vízcaino, D., Batet, O., Barrios, R., Kumar, D., and Baldasano, J. M., (2011). Seasonal variability of aerosol optical properties observed by means of a Raman lidar at an EARLINET site over Northeastern Spain. Atmos. Chem. Phys., 11, 175-190, doi:10.5194/acp-11-175-2011.

Singer, A., Ganor, E., Dultz, S., and Fischer, W., (2003). Dust deposition over the Dead Sea. J. Arid Environ., 53, 41-59, doi:10.1006/jare.2002.1023.

Tegen, I., and Lacis, A. A., (1996). Modeling of particle size distribution and its influence on the radiative properties of mineral dust aerosol. J. Geophys. Res. (Atmos.), 101, 19237-19244, doi:10.1029/95JD03610.

Ternon, E., Guieu, C., Ridame, C., L'Helguen, S., and Catala, P., (2011). Longitudinal variability of the biogeochemical role of Mediterranean aerosols in the Mediterranean Sea. Biogeosciences, 8, 1067-1080, doi:10.5194/bg-8-1067-2011.

Theodosi, C., Markaki, Z., and Mihalopoulos, N., (2010). Iron speciation, solubility and temporal variability in wet and dry deposition in the Eastern Mediterranean. Mar. Chem., 120, 100-107, doi:10.1016/j.marchem.2008.05.004.

Thingstad, T. F., Krom, M. D., Mantoura, R. F. C., Flaten, C. A. F., Groom, S., Herut, B., Kress, N., Law, C. S., Pasternak, A., Pitta, P., Psarra, S., Rassoulzadegan, F., Tanaka, T., Tselepides, A., Wassmann, P., Woodward, E. M. S., Riser, C. W., Zodiatis, C., and Zohary, T., (2005). Nature of phosphorus limitation in the ultraoligotrophic eastern Mediterranean. Science, 309, 1068-1071, doi:10.1126/science.1112632.

Todd, M. C., Bou Karam, D., Cavazos, C., Bouet, C., Heinold, B., Baldasano, J. M., Cautenet, G., Koren, I., Perez, C., Solmon, F., Tegen, I., Tulet, P., Washington, R., and Zakey, A., (2008). Quantifying uncertainty in estimates of mineral dust flux: An intercomparison of model performance over the BodÈlÈ Depression, northern Chad. Journal of Geophysical Research, 113, D24107, doi:10.1029/2008jd010476.

Volpe, G., Banzon, V. F., Evans, R. H., Santoleri, R., Mariano, A. J., and Sciarra, R., (2009). Satellite observations of the impact of dust in a low-nutrient, low-chlorophyll region: Fertilization or artifact? Global. Biogeochem. Cy., 23, GB3007, doi:10.1029/2008gb003216.

Gallisai, R., Peters, F., Volpe, G., Basart, S., and Baldasano, J. M., (2014).
Saharan dust deposition may affect phytoplankton growth in the Mediterranean Sea at ecological time scales. PLoS ONE, 9, 10, doi: 10.1371/journal.pone.0110762.

Abstract

The surface waters of the Mediterranean Sea are extremely poor in the nutrients necessary for plankton growth. At the same time, the Mediterranean Sea borders with the largest and most active desert areas in the world and the atmosphere over the basin is subject to frequent injections of mineral dust particles. We describe statistical correlations between dust deposition over the Mediterranean Sea and surface chlorophyll concentrations at ecological time scales. Aerosol deposition of Saharan origin may explain 1 to 10% (average 5%) of seasonally detrended chlorophyll variability in the low nutrient – low chlorophyll Mediterranean. Most of the statistically significant correlations are positive with main effects in spring over the Eastern and Central Mediterranean, conforming to a view of dust events fueling needed nutrients to the planktonic community. Some areas show negative effects of dust deposition on chlorophyll, coinciding with regions under a large influence of aerosols from European origin. The influence of dust deposition on chlorophyll dynamics may become larger in future scenarios of increased aridity and shallowing of the mixed layer.

Introduction

Aerosols have major impacts on weather and climate regulations (Booth et al., 2012; Creamean et al., 2013) and even on crop production (Liu et al., 2013). Atmospheric desert dust may travel large distances from its source and has been proposed to have ocean production regulation effects over geological times scales (Jaccard et al., 2013). The Mediterranean Sea (hereafter Med) atmosphere is subject to the continuous injection of Saharan and Middle East mineral dust particles (Pey et al., 2013). The deposition of these mineral particles supply numerous macro and micro- nutrients to the ocean surface (Bonnet et al., 2005; Bergametti et al., 1992; Ridame and Guieu, 2002; Markaki et al., 2003; Guieu et al., 2010b; Pulido-Villena et al., 2010; Herut et al., 1999; Jordi et al., 2012; Goudie and Middleton, 2001) and some authors consider it as the major source of "new" nutrients (Ternon et al., 2011) for system production.

Calculations show that the atmospheric input of nutrients in the Med is of the same magnitude as riverine inputs (Guieu et al., 1991; Ludwig et al., 2010; Markaki et al., 2010), thus playing a significant role in the regulation of the nutrient balance of the basin at decadal or longer time scales (Bethoux et al., 2002; Herut et al., 2005). The contribution of atmospheric deposition can be especially important and efficient in oligotrophic environments such as the Med, which has a marked stratification period and a pronounced nutrient limitation (Estrada, 1996). The deposition of some of these soluble compounds on surface waters may influence biological production, at least during certain events (Markaki et al., 2003; Guerzoni et al., 1999; Morales-Baquero et al., 2006). Dust deposition spreads over vast areas and dilutes into the water column often preventing the potential effects on system production to be unequivocally detected at ecological time scales. Experiments and observations in low nutrient – low chlorophyll areas have so far shown mixed results (Romero et al., 2011; Lekunberri et al., 2010; Volpe et al., 2009). Reasons may include a tremendous variability in dust nutrient bioavailability content (Carbo et al., 2005; Herut et al., 2002; Baker et al., 2006) and a relatively small increase of the background nutrient concentration when vertical mixing is active and represents the major source of nutrients (Estrada, 1996) as well as a rapid transfer of increased primary production to other trophic levels and a variety of plankton community structures and physiological states.

Given the episodic nature of dust events, an additional complication may reside in the human capacity of detecting the dust event deposition with sufficient space-time resolution in order to build a statistically significant dust event database. Previous attempts used satellite-derived aerosol optical thickness (AOT) as a proxy of dust in the atmosphere to infer the deposition events (Volpe et al., 2009; Cropp et al., 2005; Gabric et al., 2002). Dust generally travels from several hundreds to thousands of meters high in the atmosphere, making this approach not quantitatively adequate for discerning between transport and deposition. Deposition is measured *in situ* at a few terrestrial (mainland and islands) sites, which are extremely valuable for ground truth validation but are dependent on local conditions, making generalizations hard to draw especially towards the open ocean. Here, we employ a state-of-art atmospheric transport and deposition model, the BSC-DREAM8b model (Gallisai et al., 2012), which has been validated (Pérez et al., 2006a; Basart et al., 2012a; Basart et al., 2012b) and gives the power of having aerosol deposition data over the whole Med basin with daily temporal resolution. A previous study showed the potential positive effects of dust deposition on SeaWiFS-derived chlorophyll (Chl) in the Med (Gallisai et al., 2012). However, in the Med, the used NASA OC4v4 algorithm falls far short to retrieve Chl with accuracy smaller than 100%, casting doubts on the relationships found. Here we extend this approach by relating deposition to SeaWiFS Chl using the Med-specific algorithm MedOC4 (Volpe et al., 2007). When we think of dust deposition, we tend to think about very large events, those that are obvious in true color images or that we recognize because we find our cars covered with red dust, but the truth is that, to some extent, there is Saharan dust in the atmosphere over the Mediterranean almost continuously and deposition does not occur only during large events but also when atmospheric aerosol concentrations are not so high. Thus, rather than focusing on single events or experiments, we take a correlational approach using an 8-year data time series in order to find relationships between Chl dynamics and dust deposition over the Mediterranean Sea.

Material and methods

Chlorophyll data.

SeaWiFS HRPT Level-1A data (2000-2007) were collected at the Istituto di Scienze dell'Atmosfera e del Clima of Rome, Italy, and processed up to Level-3 using the MedOC4 regional algorithm (http://www.myocean.eu/web/69-myocean-interactive-catalogue.php) (Volpe et al., 2007). This algorithm takes into account the peculiar blue-green ratio of Med waters. Level-3 Chl data, with a native 1 km resolution, were $\log_{10}$-transformed averaged, over a period of eight days, and regridded over the 1° resolution grid of the basin (179 cells, see Table S1). A previous study showed that it is recommended not to use weekly averages when computing correlation analysis between Chl and dust events (Volpe et al., 2009). To account for the possible contamination by atmospheric dust mimicking chlorophyll, here, before averaging over the period of eight days, the quality of the entire Chl dataset was carefully checked by i) applying all the SeaDAS Level-2 processing masks and flags (http://oceancolor.gsfc.nasa.gov/VALIDATION/flags.html), ii) removing all isolated pixels, iii) removing all pixels exceeding 3 standard deviations within a moving box of 3x3 pixels, and iv) by applying a median filter over all remaining good pixels. This procedure increases the confidence level on data quality, with the only shortcoming of reducing the number of observations with respect to the NASA standard processing. The time series of daily observations was temporally binned into periods of 8 days. This results into 45 bins up to the 360^{th} day of the year. The last bin was computed with the remaining 5 days, and in the case of leap years, with the remaining 6 days. The climatic mean is then calculated across years for each of the natural 8-d time periods.

Dust deposition.

For the present study, a dust deposition simulation from the BSC-DREAM8b (http://www.bsc.es/earth-sciences/mineral-dust/catalogo-datos-dust) model (Pérez et al., 2006a; Pérez et al., 2006b) was used for the period between 1 January 2000 and 31 December 2007, over the Med basin. BSC-DREAM8b tracks mineral dust particles from their sources in the Sahara and Middle East regions. Output, after being $\log_{10}$-transformed, was provided for the same space and time resolution as for chlorophyll. A low cut-off threshold (10^{-8} Kg m^{-2} d^{-1}) is applied to the numerical deposition output from BSC-DREAM8b since the dataset showed numerically correct but physically unrealistic low value spikes (Gallisai et al., 2012). The model main features were described in detail in Pérez et al. (2006b) and Basart et al. (Basart et al., 2012b). It has been used for dust forecasting and as a dust research tool in North Africa and the Med (Gallisai et al., 2012; Amiridis et al., 2009; Alonso-Perez et al., 2011; Pay et al., 2012) Several studies have checked its performance (Pérez et al., 2006a; Papanastasiou et al., 2010), concerning both the horizontal and vertical extent of the dust plumes in the Med Basin. The model daily evaluation with near-real time observations is conducted at the Barcelona Supercomputing Center, and includes satellite data (MODIS and MSG) and AERONET sun photometers. BSC-DREAM8b has also been validated and tested over longer time periods in the European region (Basart et al., 2012a; Jiménez-Guerrero et al., 2008; Pay et al., 2010) and against measurements at source regions (Haustein et al., 2009).

Aerosol Optical Thickness.

AOT at 865 nm data were derived from SeaWiFS radiometer measurements and they were downloaded from the Giovanni database (http://gdata1.sci.gsfc.nasa.gov/daac-bin/G3/gui.cgi?instance_id=ocean_8day). We acquired 8-d averaged, 9 km resolution product from 2000 to 2007. Similarly to Chl and deposition data, AOT data were $\log_{10}$-transformed and regridded over the 1° resolution grid of the basin, with the same temporal binning. This was done for the same 179 1º X 1º cells as for chlorophyll. It should be noted that AOT contains information of total aerosol particles in the atmosphere, not only of particles from Saharan origin. However, over much of the Mediterranean Sea most particles are indeed of Saharan origin (Barnaba and Gobbi, 2004).

Statistical analyses.

Pearson's correlation coefficient (*r*) was calculated between chlorophyll concentration, modeled dust deposition and AOT time series for each grid cell. Significance was considered at $p<0.05$ using Student's t-test. In addition, the degrees of freedom used for significance testing were adjusted to take into account the possible presence of autocorrelation in the time series. The number of effective independent observations, N*, were calculated as described in Pyper el al. (Pyper and Peterman, 1998). Correlations were computed both for the entire series and for each season. The same analyses were performed after seasonally detrending the data by subtracting the climatic mean at each time series data point. The r^2 of the correlation in a cell is the variance explained by the correlation in that cell. The minimum, maximum and average variance-explained values (expressed in %variability) were calculated for the population of cells with a p<0.05.

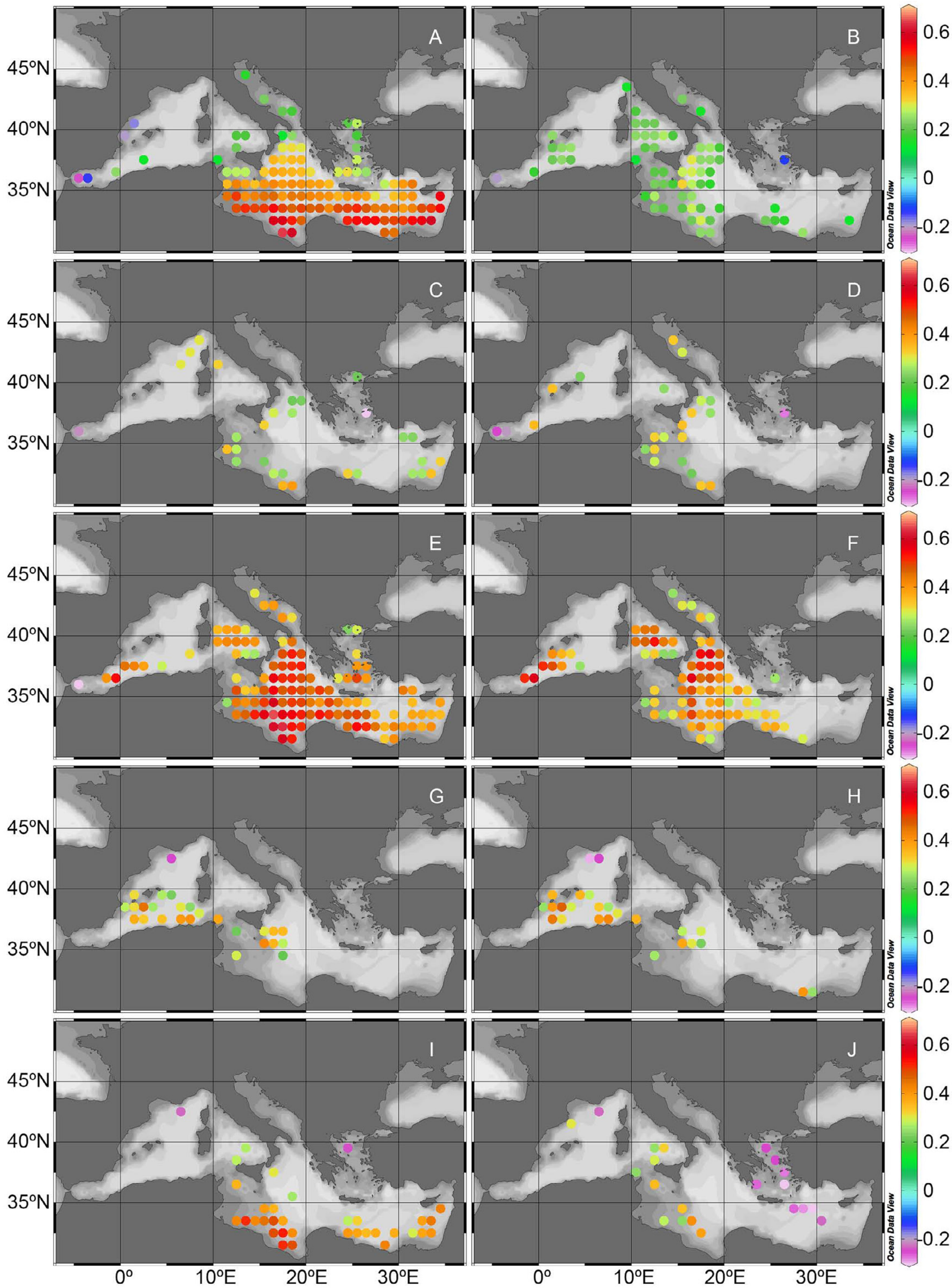

Figure 1. Correlation between chlorophyll concentration and dust deposition Statistically significant (p<0.05) correlation coefficient (r) between chlorophyll concentration and dust deposition (left panels) and between the seasonally detrended chlorophyll concentration and the seasonally detrended dust deposition (right panels) for the whole time series and for different seasons. Panels: a, b) annual; c, d) winter (January to March); e, f) spring (April to June); g, h) summer (July to September) and i, j) autumn (October to December).

Results and Discussion

We have found statistically significant positive correlations between surface chlorophyll and mineral dust deposition in large areas of the Med, covering 64% of the analyzed surface and located mainly in the Central and Eastern basins (Fig. 1) and with a clear south to north gradient in correlation intensity from 0.63 to 0.12. Significant negative correlations (r from -0.15 to -0.25) are observed in only 4 cells located in the Alboran Sea and in the eastern coast of Spain. Positive correlations can be found during all seasons, although it is in spring when we see the largest effects with correlations ranging from 0.22 to 0.65 mainly in the Central, Eastern and Southwestern Med.

The Western and Central Med also show regions with positive correlations in summer, while in autumn there are some areas affected in the Central and Eastern Med. Most of the Med phytoplankton variability (>80%) is well explained by the variability of the mixed layer depth (Volpe et al., 2012), and especially the winter-spring mixing bringing nutrient-rich deep waters to the surface. Thus, at least part of the explained variability between our deposition and chlorophyll time series must be due to the partial matching of the annual cycles of both variables.

The relationship between the seasonally detrended data of chlorophyll and dust deposition, that represents more of a response of short-term chlorophyll peaks to dust outbreaks, is somewhat weaker in intensity and in area covered. Largest positive correlations are found in the Central Med (from 0.13 to 0.32) (Fig. 1).

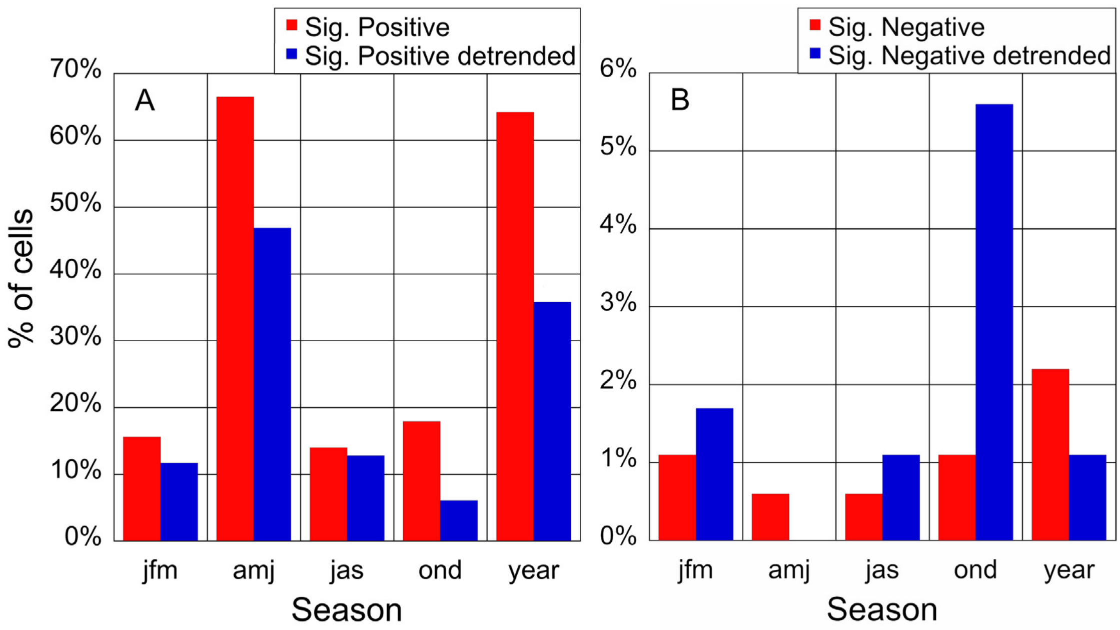

Figure 2. Percentage of cells showing significant correlations between chlorophyll and deposition. Left panel: positive correlations. Right panel: negative correlations. Red bars represent non seasonally detrended data and blue bars seasonally detrended data.

Again, it is in spring where the largest impacted area is found, mainly in the Central Med and extending into the Eastern Med and Southwestern Med with r ranging from 0.24 to 0.58. The Western Med shows the largest area affected in summer. This is not surprising given that the seasonal dust event frequency peaks during spring in the Central-Eastern Med, and during summer over the Central-Western basin (Volpe et al., 2009). Seasonally detrended data tend to slightly increase the number of cells showing significant negative correlations and decrease the number of significantly positive correlated cells (Fig. 2). It is in autumn when we see the largest number of negatively correlated cells (6% of analyzed surface) and located mainly in the Aegean Sea and extending southeasterly of Crete.

For the seasonally detrended data, we checked that the correlation values were not caused by chance. We generated synthetic seasonally detrended chlorophyll time series with the observed mean and standard deviation for each cell. Correlations were computed with dust deposition model outputs, and the process repeated 100 times (Fig. 3). The observed significant correlations were compared to the distribution of the synthetic correlations for each cell, and in all cases they were statistically different with an $\alpha<0.001$ and a power (1-β) undistinguishable from 1. This confirmed the non-spurious nature of the relationships between dust deposition and non-seasonal chlorophyll time series.

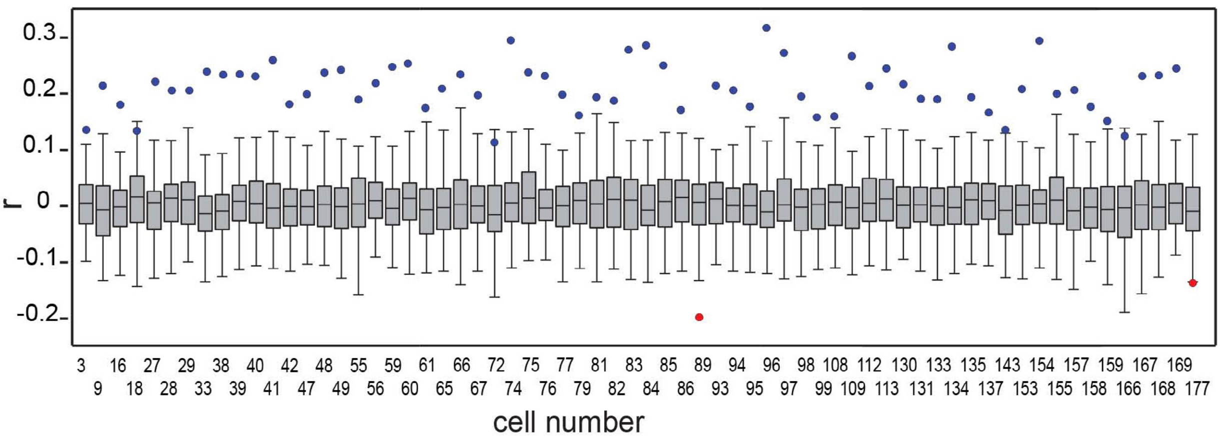

Figure 3. Analysis of the chance of significant correlations being spurious. Comparison between the box plots of the distribution of correlation coefficients between synthetic seasonally detrended chlorophyll time series and seasonally detrended dust deposition model outputs (N=100) and the actual observed correlation between the seasonally detrended chlorophyll and the seasonally detrended dust deposition model outputs (dots). Data is shown only for those cells showing significant ($p<0.05$) observed correlations. Dots in blue represent significantly positive correlations and red significantly negative correlations. Box plots show the median, the grey box englobing all data between the 25 and 75 percentiles, and the range between the smallest and largest values that are not outliers. Starting from the detrended data of the cells that show statistically significant correlations between detrended chlorophyll and deposition (Fig.1b), synthetic Chl time series with the same mean and standard deviation (normal distribution) as the original detrended chlorophyll time series, were computed for each cell. The correlation between these synthetic Chl time series and the modelled dust deposition were computed. For each cell, this process was repeated 100 times, and the probability distribution functions (PDFs) of the correlations were then obtained and presented as box plots.

Bulk Saharan dust deposition over the Med is not straightforwardly related to dust travelling in the atmosphere (Fig. S1). Meteorological conditions and wind patterns at different times of the year often have large amounts of dust (AOT) travelling at altitude with little deposition (Papayannis et al., 2008; Mona et al., 2006; Gobbi et al., 2013). AOT and dust deposition show positive correlation in the Western Med (Fig. S2.) especially in spring and summer with correlated areas shifting depending on the season.

The Eastern sub-basin presents the highest correlations in spring (from 0.23 to 0.51) and the Central Med (Tyrrhenian Sea, Sicily channel and Dardanelle strait) in autumn. Once the data are seasonally detrended, deposition events are more related to AOT events, both when the whole series is considered and when the data are analyzed for the different times of the year (Fig. S2). With respect to non-detrended data, seasonally detrended data show main increases in correlation and correlated area in the Central Med for most of the year, as well as in the Eastern Med in autumn. Some overall hotspots appear in the Alboran and in the Tyrrhenian Sea and around Crete, where the correlations ranged from 0.36 to 0.48.

The annual cycles of chlorophyll and AOT do not match (Fig. S1). The maximum chlorophyll concentrations occur in winter and minima coincide with the summer months. On the contrary, the highest AOT is found in summer and the minimum in autumn. Overall, AOT and chlorophyll (Fig S3) show no significant correlations in the Med, except for some areas near the African coasts, where the correlation is negative (from -0.28 to -0.36). While no correlations are evident between AOT and chlorophyll there are significant correlations between seasonally detrended AOT and chlorophyll data (Fig. S3). A plume of higher correlation, with r-values between 0.33 and 0.39, appear in the northern part of Cyrenaica with an extension up to the south of Italy. The best match between both series was found in summer (Fig. S3). Volpe et al. (Volpe et al., 2007) ground truthed the chlorophyll satellite estimates with in situ measurements and concluded that the atmospheric correction was appropriate. In addition, we compared the data from the chlorophyll measurements at the DYFAMED station (1998-2007) with SeaWiFS estimates corrected with a regional algorithm giving a slope of ~1 (logDYF = 0.0129 + 1.0497·logSW; Adjusted R^2=0.68; N=91; $p<0.001$). Moreover DYFAMED chlorophyll was unrelated to AOT, providing further evidence of the independence between satellite measurements of chlorophyll and AOT. Aerosols travelling over a certain area are not necessarily depositing. When a deposition event is occurring, it should coincide with high aerosol content in the air (AOT), thus if we find relationships between dust deposition events and non-seasonal chlorophyll peaks it is also logical to expect that chlorophyll is related to AOT, while the non-detrended AOT data show little or no relationship.

As mentioned before, the largest positive correlations between dust deposition and chlorophyll occur around the Central and Eastern Med. Calculations (Ridame and Guieu, 2002; Herut et al., 2005; Eker-Develi et al., 2006) and experiments (Pulido-Villena et al., 2010; Herut et al., 2005; Romero et al., 2011) tell us that aerosol deposition effects on primary production should be small

	Non-detrended data				Seasonally detrended data			
Season	**n**	**Min**	**Max**	**Average**	**n**	**Min**	**Max**	**Average**
Annual	115	1,4	40,2	16,4	64	1,3	10,1	4,7
Winter	28	4,3	15,1	7,8	21	4,4	12,6	8,5
Spring	119	5	41,6	19,1	84	5,7	33,3	15
Summer	25	4,2	19,6	10,2	23	4,7	21,3	11,1
Autumn	32	6,5	26,9	15,6	11	5,4	16,5	9,6

Table 1. Percentage of observed chlorophyll variability explained by modeled dust deposition. Number of cells (n) with significantly ($p<0.05$) positive correlations. Minimum (min), maximum (max) and average percentage of chlorophyll variability explained in the significantly positive cells.

in most situations and thus we do not expect African dust deposition in general to explain a large portion of chlorophyll variability. Accordingly, positive significant correlations between mineral dust deposition of Saharan origin and chlorophyll do explain only 1 to 10% (average 5%) of chlorophyll variability for seasonally detrended and a 1 to 40% (average 16%) for non-detrended data although it may be higher for certain seasons (Table 1). It should be noted that the explained variability does not provide direct information of the magnitude of chlorophyll impacted.

Winter shows overall the lowest significantly positive correlations, while spring presents the highest. This is to be expected since the entrance of new nutrients should be mostly due to seasonal winter overturning and mixing of nutrient-rich deep waters with upper ocean surface waters, through a number of physical processes that increase vertical diffusion at certain moments. But even at times when nutrient concentrations are expected to be relatively high in the water, low concentrations and strong imbalances between N and P are often observed (Diaz et al., 2001; Rahav et al., 2013), opening windows of opportunity for the nutrients from atmospheric deposition to have an impact in the sustainment of phytoplankton production. We can only speculate on the positive cause-effect relationship between aerosol deposition and chlorophyll in the Med at certain times. Terrestrial inputs through major rivers occur mainly in the Western Med (Struglia et al., 2004), and atmospheric inputs may dominate nutrient supply at certain times (Guieu et al., 2010b; Durrieu de Madron et al., 2011). Phosphorus (P) limitation alleviation has often been invoked (Ridame and Guieu, 2002; Izquierdo et al., 2012) as the surface waters of the Med are among the most P-limited in the world (Marty et al., 2002). Although aerosols show a disproportionally large ratio of nitrogen to phosphorus (Markaki et al., 2010), potentially only exacerbating P-limitation, they do carry an amount of P that could be used by phytoplankton and bacteria, especially in spring and summer when the concentrations of this element in surface waters of the open Med are at their lowest. Guieu et al. (Guieu et al., 2010b) calculated that, if P is considered the limiting element for phytoplankton growth, atmospheric deposition could account for chlorophyll increases of ca. 0.2 $\mu g\ L^{-1}$ in the upper mixed layer for a single large deposition event or for the average total deposition during the summer-stratified period. The Central and Eastern Med do not show the typical spring phytoplankton bloom and have been defined as no blooming areas (D'Ortenzio and Ribera D'Alcalà, 2009). The ultra-oligotrophic conditions (Pujo-Pay et al., 2011) found in these areas should make them most responsive to external nutrient supplies. As is the case for high nutrient – low chlorophyll areas, micronutrients such as iron from aerosols have also been proposed to stimulate Med phytoplankton production under certain situations (Bonnet and Guieu, 2006), albeit addition experiments have not shown a direct increase in dissolved iron (Fe) (Wagener et al., 2010). Fe in the mixed layer of the Mediterranean is found at concentrations from 0.13 to 2.7 nM (Sarthou and Jeandel, 2001; Guieu et al., 2002). It seems though that Fe is, relative to the needs of plankton, in excess with respect to P in the Mediterranean (Guieu et al., 2010a). Nevertheless, in a system where all elements are relatively scarce, responses to the combination of elements arriving through aerosol deposition, may be very complex, with elements becoming successively limiting in a chained reaction. Ridame et al. (2011) found stimulation of nitrogen (N) fixation in dust pulse experiments, in general related to a primary alleviation of P-limitation. In their Central Med experiment though, they found high N-fixation stimulation unrelated to P- or Fe-limitation, further showing the complexity of the processes involved and the potential spatial and temporal variability. An initial stimulation of heterotrophic bacteria (Lekunberri et al., 2010; Pulido-Villena et al., 2008) should not be discarded since these organisms have a potential advantage at low nutrient concentrations owing to their high surface to volume ratio. Secondarily, released nutrients from recycling could then stimulate phytoplankton processes. Contrary to the Eastern Med showing the lowest nutrient concentrations in the Mediterranean (Pujo-Pay et al., 2011), the Central Med was found somewhat more responsive to dust deposition in the present study. Pey et al. (Pey et al., 2013) mention the Central Med as a transitional area, receiving a higher frequency of dust outbreaks than similar latitudes in the Western and Eastern Med. Additionally, the dust source areas are not homogeneous. The Libyan Desert is the main source of dust for the Central Med while the Eastern Med receives dust from Libya and from the Middle East (Gaetani and Pasqui, 2012). Thus, positive correlations between dust deposition and surface chlorophyll seem to arise from the combination of areas of low nutrient concentrations with the right nature, timing and frequency of dust outbreaks.

Negative relationships between dust deposition and chlorophyll have been related to metal (mainly Cu but also Al) inhibition of phytoplankton growth (Jordi et al., 2012; Paytan et al., 2009). The toxicity of Cu in reducing phytoplankton growth rate has been shown in laboratory experiments (see (Paytan et al., 2009) and references therein). A recent correlation study between chlorophyll and metals from onshore-measured aerosols in the Northwest Med shows negative relationships in the area under northerly wind (Tramontane) conditions (Jordi et al., 2012). These winds favor the transport of anthropogenic aerosols from Europe to the Med. Although most Cu pulses are anthropogenically derived, pulses originating in Africa showed effects on chlorophyll undistinguishable from those originating locally (Jordi et al., 2012). A reduction in chlorophyll growth of up to 20% can be seen along the French and Spanish coasts. In addition, Jordi et al. (Jordi et al., 2012) argue that since Cu toxicity seems to be taxon specific, the summer phytoplankton community with a predominance of nanoflagellates over the less sensitive diatoms, is more vulnerable to atmospheric deposition. This is an area where we also see some negative correlations between the modeled deposition and chlorophyll. We only track Saharan mineral dust, while some of the high loads of metals may be more related to local anthropogenic sources. Most of the large deposition events in the Northwest Med come in the form of wet deposition (Avila and Peñuelas, 1999). In our model, the deposition field only originates from Saharan and middle East dust transport and does not account for local anthropogenic aerosol sources, but rain washes out the entire atmospheric column aerosol loading, no matter the origin. Results from our correlation analysis agree with previous more detailed local studies (Jordi et al., 2012). We also see a negative relationship between deposition and chlorophyll, both seasonally detrended, mainly in autumn in the Aegean region (Fig. 1). This area is affected by long-range transport of air pollutants from Eastern Europe (Lelieveld et al., 2002) but it is also heavily impacted by anthropogenic emissions generated in Athens and Istanbul (Kanakidou et al., 2011; Querol et al., 2009). A high-density population together with a massive number of vehicles, many of them still using non-catalytic or old technology diesel engines, contributed to exceed the EU annual aerosol limit. The amount of Cu in these aerosols is high with an annual mean concentration between 0.013 and 0.22 $\mu g\ m^{-3}$ ((Theodosi et al., 2011) and references therein). An estimated dry deposition flux of Cu over the sea ranges then between 22 and 380 μg Cu $m^{-2}\ d^{-1}$ surpassing the threshold limit for Cu to inhibit phytoplankton growth rate according to (Jordi et al., 2012) and (Paytan et al., 2009).

Conclusion

Desert dust storm events seem to be increasing in frequency and intensity (Goudie and Middleton, 2001; Avila and Peñuelas, 1999; Ganor et al., 2010; Goudie, 2009; Mahowald et al., 2010) in the last decades, due to human activities and climate forcing. This means that the presence of aerosols over the Med is likely to increase with future aridity. Thus, it is important to understand basin level patterns in the response of Med biogeochemistry to aerosol deposition. Only a few studies (Volpe et al., 2009; Cropp et al., 2005) have tried to analyze the potential links between aerosols in the air column and chlorophyll for the entire Med basin, with non definitive results. In this study, we use a modeled actual aerosol deposition product and show both positive and negative significant correlations with chlorophyll dynamics in certain areas and times of the year. Mineral dust from North Africa and the Middle East correlates to chlorophyll in large areas of the Med Sea. This is especially true for the Central and Eastern Med sub-basins, where Saharan dust deposition dynamics matches that of chlorophyll, particularly during spring. Here the atmospheric input may be an intrinsic part of the annual ecosystem dynamics. In terms of large dust outbreaks, chlorophyll best relates to aerosol deposition in the Central Med, extending both into the Eastern and Southwestern Med (Fig. S3). Some areas of the Western Med and Aegean Sea show negative correlations between chlorophyll and deposition, in accordance with some recent findings of toxicity brought by metals in aerosols. As expected, dust deposition does not explain an overall large amount of chlorophyll variability since the main ecosystem production driver in the Med is the vertical mixing of nutrients from deep waters. Variability related to carbon to chlorophyll ratios, the consumption of biomass with a varying degree of coupling and the variable settling of primary production, are all additional sources of surface chlorophyll variability that we could not account for in our correlations and thus add to the noise. No matter how small significant correlations are, they are not distributed randomly in space and coincide with independent estimates that follow the same trend. Thus, albeit the mechanisms that affect chlorophyll through aerosol deposition cannot be pinpointed and may be indirect, non-unique, and dependent on local spatio-temporal conditions, our study shows a clear potential for effects at ecological scales. These effects should become more important in a future scenario with increased aerosols over the Med and a shallower mixed layer depth, owing to increased temperatures, over which aerosols may leach out nutrients.

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References

Alonso-Perez, S., Cuevas, E., Perez, C., Querol, X., Baldasano, J. M., Draxler, R., and De Bustos, J. J., (2011). Trend changes of African airmass intrusions in the marine boundary layer over the subtropical Eastern North Atlantic region in winter. Tellus (B Chem. Phys. Meteorol.), 63, 255-265, doi:10.1111/j.1600-0889.2010.00524.x.

Amiridis, V., Kafatos, M., Perez, C., Kazadzis, S., Gerasopoulos, E., Mamouri, R. E., Papayannis, A., Kokkalis, P., Giannakaki, E., Basart, S., Daglis, I., and Zerefos, C., (2009). The potential of the synergistic use of passive and active remote sensing measurements for the validation of a regional dust model. Ann. Geophys., 27, 3155-3164, doi:10.5194/angeo-27-3155-2009.

Avila, A., and Peñuelas, J., (1999). Increasing frequency of Saharan rains over northeastern Spain and its ecological consequences. Sci. Total Environ., 228, 153-156, doi:10.1016/s0048-9697(99)00041-8.

Baker, A. R., French, M., and Linge, K. L., (2006). Trends in aerosol nutrient solubility along a west–east transect of the Saharan dust plume. Geophys. Res. Lett., 33, L07805, doi:10.1029/2005gl024764.

Barnaba, F., and Gobbi, G. P., (2004). Aerosol seasonal variability over the Mediterranean region and relative impact of maritime, continental and Saharan dust particles over the basin from MODIS data in the year 2001. Atmos. Chem. Phys., 4, 2367-2391, doi:10.5194/acp-4-2367-2004.

Basart, S., Pay, M. T., Jorba, O., Pérez, C., Jiménez-Guerrero, P., Schulz, M., and Baldasano, J. M., (2012a). Aerosols in the CALIOPE air quality modelling system: Evaluation and analysis of PM levels, optical depths and chemical composition over Europe. Atmos. Chem. Phys., 12, 3363-3392, doi:10.5194/acp-12-3363-2012.

Basart, S., Perez, C., Nickovic, S., Cuevas, E., and Baldasano, J. M., (2012b). Development and evaluation of the BSC-DREAM8b dust regional model over Northern Africa, the Mediterranean and the Middle East. Tellus (B Chem. Phys. Meteorol.), 64, 18539, doi:10.3402/tellusb.v64i0.18539.

Bergametti, G., Remoudaki, E., Losno, R., Steiner, E., Chatenet, B., and Buatmenard, P., (1992). Source, transport and deposition of atmospheric phosphorus over the northwestern Mediterranean. J. Atmos. Chem., 14, 501-513, doi:10.1007/bf00115254.

Bethoux, J. P., Morin, P., and Ruiz-Pino, D. P., (2002). Temporal trends in nutrient ratios: chemical evidence of Mediterranean ecosystem changes driven by human activity. Deep-Sea Res. Part II., 49, 2007-2016, doi:10.1016/s0967-0645(02)00024-3.

Bonnet, S., Guieu, C., Chiaverini, J., Ras, J., and Stock, A., (2005). Effect of atmospheric nutrients on the autotrophic communities in a low nutrient, low chlorophyll system. Limnol. Oceanogr., 50, 1810-1819, doi:10.4319/lo.2005.50.6.1810.

Bonnet, S., and Guieu, C., (2006). Atmospheric forcing on the annual iron cycle in the western Mediterranean Sea: A 1-year survey. J. Geophys. Res. (Oceans), 111, C09010, doi:10.1029/2005jc003213.

Booth, B. B. B., Dunstone, N. J., Halloran, P. R., Andrews, T., and Bellouin, N., (2012). Aerosols implicated as a prime driver of twentieth-century North Atlantic climate variability. Nature, 484, 228–232, doi:10.1038/nature10946.

Carbo, P., Krom, M. D., Homoky, W. B., Benning, L. G., and Herut, B., (2005). Impact of atmospheric deposition on N and P geochemistry in the southeastern Levantine basin. Deep-Sea Res. Part II., 52, 3041-3053, doi:10.1016/j.dsr2.2005.08.014

Creamean, J. M., Suski, K. J., Rosenfeld, D., Cazorla, A., DeMott, P. J., Sullivan, R. C., White, A. B., Ralph, F. M., Minnis, P., Comstock, J. M., Tomlinson, J. M., and Prather, K. A., (2013). Dust and Biological Aerosols from the Sahara and Asia Influence Precipitation in the Western U.S. Science, 339, 1572-1578, doi:10.1126/science.1227279.

Cropp, R. A., Gabric, A. J., McTainsh, G. H., Braddock, R. D., and Tindale, N., (2005). Coupling between ocean biota and atmospheric aerosols: Dust, dimethylsulphide, or artifact? Global. Biogeochem. Cy., 19, GB4002 doi:10.1029/2004gb002436.

D'Ortenzio, F., and Ribera D'Alcalà, M., (2009). On the trophic regimes of the Mediterranean Sea: A satellite analysis. Biogeosciences, 6, 139-148, doi:10.5194/bg-6-139-2009.

Diaz, F., Raimbault, P., Boudjellal, B., Garcia, N., and Moutin, T., (2001). Early spring phosphorus limitation of primary productivity in a NW Mediterranean coastal zone (Gulf of Lions). Mar. Ecol. Prog. Ser., 211, 51-62, doi:10.3354/meps211051.

Durrieu de Madron, X., Guieu, C., Sempéré, R., Conan, P., Cossa, D., D'Ortenzio, F., Estournel, C., Gazeau, F., Rabouille, C., Stemmann, L., Bonnet, S., Diaz, F., Koubbi, P., Radakovitch, O., Babin, M., Baklouti, M., Bancon-Montigny, C., Belviso, S., Bensoussan, N., Bonsang, B., Bouloubassi, I., Brunet, C., Cadiou, J. F., Carlotti, F., Chami, M., Charmasson, S., Charrière, B., Dachs, J., Doxaran, D., Dutay, J. C., Elbaz-Poulichet, F., Eléaume, M., Eyrolles, F., Fernandez, C., Fowler, S., Francour, P., Gaertner, J. C., Galzin, R., Gasparini, S., Ghiglione, J. F., Gonzalez, J. L., Goyet, C., Guidi, L., Guizien, K., Heimbürger, L. E., Jacquet, S. H. M., Jeffrey, W. H., Joux, F., Le Hir, P., Leblanc, K., Lefèvre, D., Lejeusne, C., Lemé, R., Loÿe-Pilot, M. D., Mallet, M., Méjanelle, L., Mélin, F., Mellon, C., Mérigot, B., Merle,

P. L., Migon, C., Miller, W. L., Mortier, L., Mostajir, B., Mousseau, L., Moutin, T., Para, J., Pérez, T., Petrenko, A., Poggiale, J. C., Prieur, L., Pujo-Pay, M., Pulido, V., Raimbault, P., Rees, A. P., Ridame, C., Rontani, J. F., Ruiz Pino, D., Sicre, M. A., Taillandier, V., Tamburini, C., Tanaka, T., Taupier-Letage, I., Tedetti, M., Testor, P., Thébault, H., Thouvenin, B., Touratier, F., Tronczynski, J., Ulses, C., Van Wambeke, F., Vantrepotte, V., Vaz, S., and Verney, R., (2011). Marine ecosystems' responses to climatic and anthropogenic forcings in the Mediterranean. Prog. Oceanogr., 91, 97-166, doi:10.1016/j.pocean.2011.02.003.

Eker-Develi, E., Kideys, A. E., and Tugrul, S., (2006). Role of Saharan dust on phytoplankton dynamics in the northeastern Mediterranean. Mar. Ecol. Prog. Ser., 314, 61-75, doi:10.3354/meps314061.

Estrada, M., (1996). Primary production in the northwestern Mediterranean. Scientia Marina, 60, 55-64.

Gabric, A. J., Cropp, R., Ayers, G. P., McTainsh, G., and Braddock, R., (2002). Coupling between cycles of phytoplankton biomass and aerosol optical depth as derived from SeaWiFS time series in the Subantarctic Southern Ocean. Geophys. Res. Lett., 29, 1112, doi:10.1029/2001gl013545.

Gaetani, M., and Pasqui, M., (2012). Synoptic patterns associated with extreme dust events in the Mediterranean Basin. Reg. Environ. Change., 1-14, doi:10.1007/s10113-012-0386-2.

Gallisai, R., Peters, F., Basart, S., and Baldasano, J. M., (2012). Mediterranean basin-wide correlations between Saharan dust deposition and ocean chlorophyll concentration. Biogeosci. Disc., 9, 28, doi:10.5194/bgd-9-8611-2012.

Ganor, E., Osetinsky, I., Stupp, A., and Alpert, P., (2010). Increasing trend of African dust, over 49 years, in the eastern Mediterranean. J. Geophys. Res. (Atmos.), 115, D07201, doi:10.1029/2009jd012500.

Gobbi, G. P., Angelini, F., Barnaba, F., Costabile, F., Baldasano, J. M., Basart, S., Sozzi, R., and Bolignano, A., (2013). Changes in particulate matter physical properties during Saharan advections over Rome (Italy): a four-year study, 2001–2004. Atmos. Chem. Phys., 13, 7395-7404, doi:10.5194/acp-13-7395-2013.

Goudie, A. S., and Middleton, N. J., (2001). Saharan dust storms: nature and consequences. Earth-Sci. Rev., 56, 179-204, doi:10.1016/s0012-8252(01)00067-8.

Goudie, A. S., (2009). Dust storms: Recent developments. J. Environ. Manage., 90, 89-94, doi:10.1016/j.jenvman.2008.07.007.

Guerzoni, S., Chester, R., Dulac, F., Herut, B., Loÿe-Pilot, M. D., Measures, C., Migon, C., Molinaroli, E., Moulin, C., Rossini, P., Saydam, C., Soudine, A., and Ziveri, P., (1999). The role of atmospheric deposition in the biogeochemistry of the Mediterranean Sea. Prog. Oceanogr., 44, 147-190, doi:10.1016/S0079-6611(99)00024-5.

Guieu, C., Martin, J. M., Thomas, A. J., and Elbazpoulichet, F., (1991). Atmospheric versus river inputs of metals to the Gulf of Lions: Total concentrations, partitioning and fluxes. Mar. Pollut. Bull., 22, 176-183, doi:10.1016/0025-326x(91)90467-7.

Guieu, C., Bozec, Y., Blain, S., Ridame, C., Sarthou, G., and Leblond, N., (2002). Impact of high Saharan dust inputs on dissolved iron concentrations in the Mediterranean Sea. Geophys. Res. Lett., 29, 1911, doi:10.1029/2001GL014454.

Guieu, C., Dulac, F., Desboeufs, K., Wagener, T., Pulido-Villena, E., Grisoni, J. M., Louis, F., Ridame, C., Blain, S., Brunet, C., Nguyen, E. B., Tran, S., Labiadh, M., and Dominici, J. M., (2010a). Large clean mesocosms and simulated dust deposition: a new methodology to investigate responses of marine oligotrophic ecosystems to atmospheric inputs. Biogeosciences, 7, 2765-2784, doi:10.5194/bg-7-2765-2010.

Guieu, C., Loÿe-Pilot, M. D., Benyahya, L., and Dufour, A., (2010b). Spatial variability of atmospheric fluxes of metals (Al, Fe, Cd, Zn and Pb) and phosphorus over the whole Mediterranean from a one-year monitoring experiment: Biogeochemical implications. Mar. Chem., 120, 164-178, doi:10.1016/j.marchem.2009.02.004.

Haustein, K., Pérez, C., Baldasano, J. M., Müller, D., Tesche, M., Schladitz, A., Esselborn, M., Weinzierl, B., Kandler, K., and Von Hoyningen-Huene, W., (2009). Regional dust model performance during SAMUM 2006. Geophys. Res. Lett., 36, L03812, doi:10.1029/2008GL036463.

Herut, B., Krom, M. D., Pan, G., and Mortimer, R., (1999). Atmospheric input of nitrogen and phosphorus to the Southeast Mediterranean: Sources, fluxes, and possible impact. Limnol. Oceanogr., 44, 1683-1692, doi:10.4319/lo.1999.44.7.1683.

Herut, B., Collier, R., and Krom, M. D., (2002). The role of dust in supplying nitrogen and phosphorus to the Southeast Mediterranean. Limnol. Oceanogr., 47, 870-878, doi:10.4319/lo.2002.47.3.0870.

Herut, B., Zohary, T., Krom, M. D., Mantoura, R. F. C., Pitta, P., Psarra, S., Rassoulzadegan, F., Tanaka, T., and Thingstad, T. F., (2005). Response of East Mediterranean surface water to Saharan dust: On-board microcosm experiment and field observations. Deep-Sea Res. Part II., 52, 3024-3040, doi:10.1016/j.dsr.2005.09.003.

Izquierdo, R., Benítez-Nelson, C. R., Masqué, P., Castillo, S., Alastuey, A., and Àvila, A., (2012). Atmospheric phosphorus deposition in a near-coastal rural site in the NE Iberian Peninsula and its role in marine productivity. Atmos. Environ., 49, 361-370, doi:10.1016/j.atmosenv.2011.11.007.

Jaccard, S. L., Hayes, C. T., Martinez-Garcia, A., Hodell, D. A., Anderson, R. F., Sigman, D. M., and Haug, G. H., (2013). Two Modes of Change in Southern Ocean Productivity Over the Past Million Years. Science, 339, 1419-1423, doi:10.1126/science.1227545.

Jiménez-Guerrero, P., Pérez, C., Jorba, O., and Baldasano, J. M., (2008). Contribution of Saharan dust in an integrated air quality system and its on-line assessment. Geophys. Res. Lett., 35, L03814, doi:10.1029/2007GL031580.

Jordi, A., Basterretxea, G., Tovar-Sanchez, A., Alastuey, A., and Querol, X., (2012). Copper aerosols inhibit phytoplankton growth in the Mediterranean Sea. Proc. Natl. Acad. Sci. USA., 109, 21246-21249, doi:10.1073/pnas.1207567110.

Kanakidou, M., Mihalopoulos, N., Kindap, T., Im, U., Vrekoussis, M., Gerasopoulos, E., Dermitzaki, E., Unal, A., Kocak, M., Markakis, K., Melas, D., Kouvarakis, G., Youssef, A. F., Richter, A., Hatzianastassiou, N., Hilboll, A., Ebojie, F., Wittrock, F., von Savigny, C., Burrows, J. P., Ladstaetter-Weissenmayer, A., and Moubasher, H., (2011). Megacities as hot spots of air pollution in the East Mediterranean. Atmos. Environ., 45, 1223-1235, doi:10.1016/j.atmosenv.2010.11.048.

Lekunberri, I., Lefort, T., Romero, E., Vázquez-Domínguez, E., Romera-Castillo, C., Marrasé, C., Peters, F., Weinbauer, M., and Gasol, J. M., (2010). Effects of a dust deposition event on coastal marine microbial abundance and activity, bacterial community structure and ecosystem function. J. Plankton Res., 32, 381-396, doi:10.1093/plankt/fbp137.

Lelieveld, J., Berresheim, H., Borrmann, S., Crutzen, P. J., Dentener, F. J., Fischer, H., Feichter, J., Flatau, P. J., Heland, J., Holzinger, R., Korrmann, R., Lawrence, M. G., Levin, Z., Markowicz, K. M., Mihalopoulos, N., Minikin, A., Ramanathan, V., de Reus, M., Roelofs, G. J., Scheeren, H. A., Sciare, J., Schlager, H., Schultz, M., Siegmund, P., Steil, B., Stephanou, E. G., Stier, P., Traub, M., Warneke, C., Williams, J., and Ziereis, H., (2002). Global air pollution crossroads over the Mediterranean. Science, 298, 794-799, doi:10.1126/science.1075457.

Liu, X., Zhang, Y., Han, W., Tang, A., Shen, J., Cui, Z., Vitousek, P., Erisman, J. W., Goulding, K., Christie, P., Fangmeier, A., and Zhang, F., (2013). Enhanced nitrogen deposition over China. Nature, 494, 459-462, doi:10.1038/nature11917.

Ludwig, W., Bouwman, A. F., Dumont, E., and Lespinas, F., (2010). Water and nutrient fluxes from major Mediterranean and Black Sea rivers: Past and future trends and their implications for the basin-scale budgets. Global. Biogeochem. Cy., 24, GB0A13 doi:10.1029/2009gb003594.

Mahowald, N. M., Kloster, S., Engelstaedter, S., Moore, J. K., Mukhopadhyay, S., McConnell, J. R., Albani, S., Doney, S. C., Bhattacharya, A., Curran, M. A. J., Flanner, M. G., Hoffman, F. M., Lawrence, D. M., Lindsay, K., Mayewski, P. A., Neff, J., Rothenberg, D., Thomas, E., Thornton, P. E., and Zender, C. S., (2010). Observed 20th century desert dust variability: impact on climate and biogeochemistry. Atmos. Chem. Phys., 10, 10875-10893, doi:10.5194/acp-10-10875-2010.

Markaki, Z., Oikonomou, K., Kocak, M., Kouvarakis, G., Chaniotaki, A., Kubilay, N., and Mihalopoulos, N., (2003). Atmospheric deposition of inorganic phosphorus in the Levantine Basin, eastern Mediterranean: Spatial and temporal variability and its role in seawater productivity. Limnol. Oceanogr., 48, 1557-1568, doi:10.4319/lo.2003.48.4.1557.

Markaki, Z., Loÿe-Pilot, M. D., Violaki, K., Benyahya, L., and Mihalopoulos, N., (2010). Variability of atmospheric deposition of dissolved nitrogen and phosphorus in the Mediterranean and possible link to the anomalous seawater N/P ratio. Mar. Chem., 120, 187-194, doi:10.1016/j.marchem.2008.10.005.

Marty, J. C., Chiaverini, J., Pizay, M. D., and Avril, B., (2002). Seasonal and interannual dynamics of nutrients and phytoplankton pigments in the western Mediterranean Sea at the DYFAMED time-series station (1991-1999). Deep-Sea Res. Part II., 49, 1965-1985, doi:10.1016/s0967-0645(02)00022-x.

Mona, L., Amodeo, A., Pandolfi, M., and Pappalardo, G., (2006). Saharan dust intrusions in the Mediterranean area: Three years of Raman lidar measurements. J. Geophys. Res. (Atmos.), 111, D16203, doi:10.1029/2005jd006569.

Morales-Baquero, R., Pulido-Villena, E., and Reche, I., (2006). Atmospheric Inputs of Phosphorus and Nitrogen to the Southwest Mediterranean Region: Biogeochemical Responses of High Mountain Lakes. Limnol. Oceanogr., 51, 830-837, doi:10.4319/lo.2006.51.2.0830.

Papanastasiou, D. K., Poupkou, A., Katragkou, E., Amiridis, V., Melas, D., Mihalopoulos, N., Basart, S., Pérez, C., and Baldasano, J. M., (2010). An Assessment of the Efficiency of Dust Regional Modelling to Predict Saharan Dust Transport Episodes. Adv. Meteo., 2010, 154368 doi:10.1155/2010/154368.

Papayannis, A., Amiridis, V., Mona, L., Tsaknakis, G., Balis, D., Bösenberg, J., Chaikovski, A., De Tomasi, F., Grigorov, I., Mattis, I., Mitev, V., Müller, D., Nickovic, S., Pérez, C., Pietruczuk, A., Pisani, G., Ravetta, F., Rizi, V., Sicard, M., Trickl, T., Wiegner, M., Gerding, M., Mamouri, R. E., D'Amico, G., and Pappalardo, G., (2008). Systematic lidar observations of Saharan dust over Europe in the frame of EARLINET (2000-2002). J. Geophys. Res. (Atmos.), 113, D10204, doi:10.1029/2007JD009028.

Pay, M. T., Piot, M., Jorba, O., Gassó, S., Gonçalves, M., Basart, S., Dabdub, D., Jiménez-Guerrero, P., and Baldasano, J. M., (2010). A full year evaluation of the CALIOPE-EU air quality modeling system over Europe for 2004. Atmos. Environ., 44, 3322-3342, doi:10.1016/j.atmosenv.2010.05.040.

Pay, M. T., Jiménez-Guerrero, P., Jorba, O., Basart, S., Querol, X., Pandolfi, M., and Baldasano, J. M., (2012). Spatio-temporal variability of concentrations and speciation of particulate matter across Spain in the CALIOPE modeling system. Atmos. Environ., 46, 376-396, doi:10.1016/j.atmosenv.2011.09.049.

Paytan, A., Mackey, K. R. M., Chen, Y., Lima, I. D., Doney, S. C., Mahowald, N., Labiosa, R., and Postf, A. F., (2009). Toxicity of atmospheric aerosols on marine phytoplankton. Proc. Natl. Acad. Sci. USA., 106, 4601-4605, doi:10.1073/pnas.0811486106.

Pérez, C., Nickovic, S., Baldasano, J. M., Sicard, M., Rocadenbosch, F., and Cachorro, V. E., (2006a). A long Saharan dust event over the western Mediterranean: Lidar, Sun photometer observations, and regional dust modeling. J. Geophys. Res. (Atmos.), 111, D15214, doi:10.1029/2005JD006579.

Pérez, C., Nickovic, S., Pejanovic, G., Baldasano, J. M., and Özsoy, E., (2006b). Interactive dust-radiation modeling: A step to improve weather forecasts. J. Geophys. Res. (Atmos.), 111, D16206, doi:10.1029/2005JD006717.

Pey, J., Querol, X., Alastuey, A., Forastiere, F., and Stafoggia, 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, doi:10.5194/acp-13-1395-2013.

Pujo-Pay, M., Conan, P., Oriol, L., Cornet-Barthaux, V., Falco, C., Ghiglione, J. F., Goyet, C., Moutin, T., and Prieur, L., (2011). Integrated survey of elemental stoichiometry (C, N, P) from the western to eastern Mediterranean Sea. Biogeosciences, 8, 883-899, doi:10.5194/bg-8-883-2011.

Pulido-Villena, E., Wagener, T., and Guieu, C., (2008). Bacterial response to dust pulses in the western Mediterranean: Implications for carbon cycling in the oligotropic ocean. Global. Biogeochem. Cy., 22, GB1020, doi:10.1029/2007GB003091.

Pulido-Villena, E., Rerolle, V., and Guieu, C., (2010). Transient fertilizing effect of dust in P-deficient LNLC surface ocean. Geophys. Res. Lett., 37, L01603, doi:10.1029/2009gl041415.

Pyper, B. J., and Peterman, R. M., (1998). Comparison of methods to account for autocorrelation in correlation analyses of fish data. Can. J. Fish. Aquat. Sci., 55, 2127-2140, doi: 10.1139/cjfas-55-9-2127.

Querol, X., Alastuey, A., Pey, J., Cusack, M., Perez, N., Mihalopoulos, N., Theodosi, C., Gerasopoulos, E., Kubilay, N., and Kocak, M., (2009). Variability in regional background aerosols within the Mediterranean. Atmos. Chem. Phys., 9, 4575-4591, doi:10.5194/acp-9-4575-2009.

Rahav, E., Herut, B., Levi, A., Mulholland, M. R., and Berman-Frank, I., (2013). Springtime contribution of dinitrogen fixation to primary production across the Mediterranean Sea. Ocean Sci., 9, 489-498, doi:10.5194/os-9-489-2013.

Ridame, C., and Guieu, C., (2002). Saharan input of phosphate to the oligotrophic water of the open western Mediterranean sea. Limnol. Oceanogr., 47, 856-869, doi:10.4319/lo.2002.47.3.0856.

Ridame, C., Le Moal, M., Guieu, C., Ternon, E., Biegala, I. C., L'Helguen, S., and Pujo-Pay, M., (2011). Nutrient control of N-2 fixation in the oligotrophic Mediterranean Sea and the impact of Saharan dust events. Biogeosciences, 8, 2773-2783, doi:10.5194/bg-8-2773-2011.

Romero, E., Peters, F., Marrasé, C., Guadayol, T., Gasol, J. M., and Weinbauer, M. G., (2011). Coastal Mediterranean plankton stimulation dynamics through a dust storm event: An experimental simulation. Estuar. Coast. Shelf Sci., 93, 27-39, doi:10.1016/j.ecss.2011.03.019.

Sarthou, G., and Jeandel, C., (2001). Seasonal variations of iron concentrations in the Ligurian Sea and iron budget in the Western Mediterranean Sea. Mar. Chem., 74, 115-129, doi:10.1016/s0304-4203(00)00119-5.

Struglia, M. V., Mariotti, A., and Filograsso, A., (2004). River discharge into the Mediterranean Sea: Climatology and aspects of the observed variability. J. Clim., 17, 4740-4751, doi:10.1175/jcli-3225.1.

Ternon, E., Guieu, C., Ridame, C., L'Helguen, S., and Catala, P., (2011). Longitudinal variability of the biogeochemical role of Mediterranean aerosols in the Mediterranean Sea. Biogeosciences, 8, 1067-1080, doi:10.5194/bg-8-1067-2011.

Theodosi, C., Grivas, G., Zarmpas, P., Chaloulakou, A., and Mihalopoulos, N., (2011). Mass and chemical composition of size-segregated aerosols (PM1, PM2.5, PM10) over Athens, Greece: local versus regional sources. Atmos. Chem. Phys., 11, 11895-11911, doi:10.5194/acp-11-11895-2011.

Volpe, G., Santoleri, R., Vellucci, V., d'Alcala, M. R., Marullo, S., and D'Ortenzio, F., (2007). The colour of the Mediterranean Sea: Global versus regional bio-optical algorithms evaluation and implication for satellite chlorophyll estimates. Remote Sens. Environ., 107, 625-638, doi:10.1016/j.rse.2006.10.017.

Volpe, G., Banzon, V. F., Evans, R. H., Santoleri, R., Mariano, A. J., and Sciarra, R., (2009). Satellite observations of the impact of dust in a low-nutrient, low-chlorophyll region: Fertilization or artifact? Global. Biogeochem. Cy., 23, GB3007, doi:10.1029/2008gb003216.

Volpe, G., Nardelli, B. B., Cipollini, P., Santoleri, R., and Robinson, I. S., (2012). Seasonal to interannual phytoplankton response to physical processes in the Mediterranean Sea from satellite observations. Remote Sens. Environ., 117, 223-235, doi:10.1016/j.rse.2011.09.020.

Wagener, T., Guieu, C., and Leblond, N., (2010). Effects of dust deposition on iron cycle in the surface Mediterranean Sea: results from a mesocosm seeding experiment. Biogeosciences, 7, 3769-3781, doi:10.5194/bg-7-3769-2010.

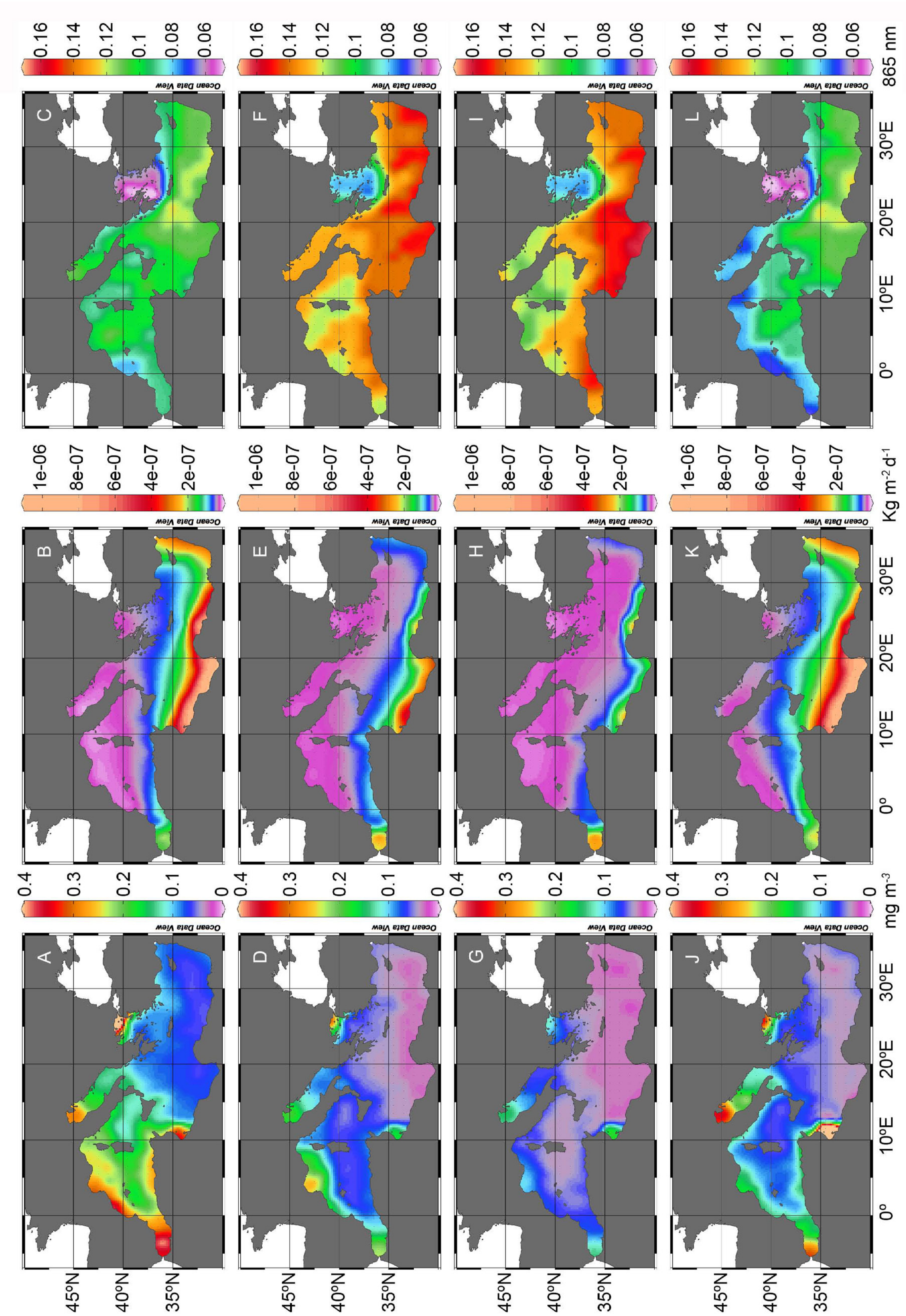

Figure S1. Seasonal average values of chlorophyll concentration, dust deposition and aerosol optical thickness. Average chlorophyll concentration (left panels). Average dust deposition (central panels) and average aerosol optical thickness (right panels) for different seasons. Winter (a, b, c), spring (d, e, f), summer (g, h, i) and autumn (j, k, l).

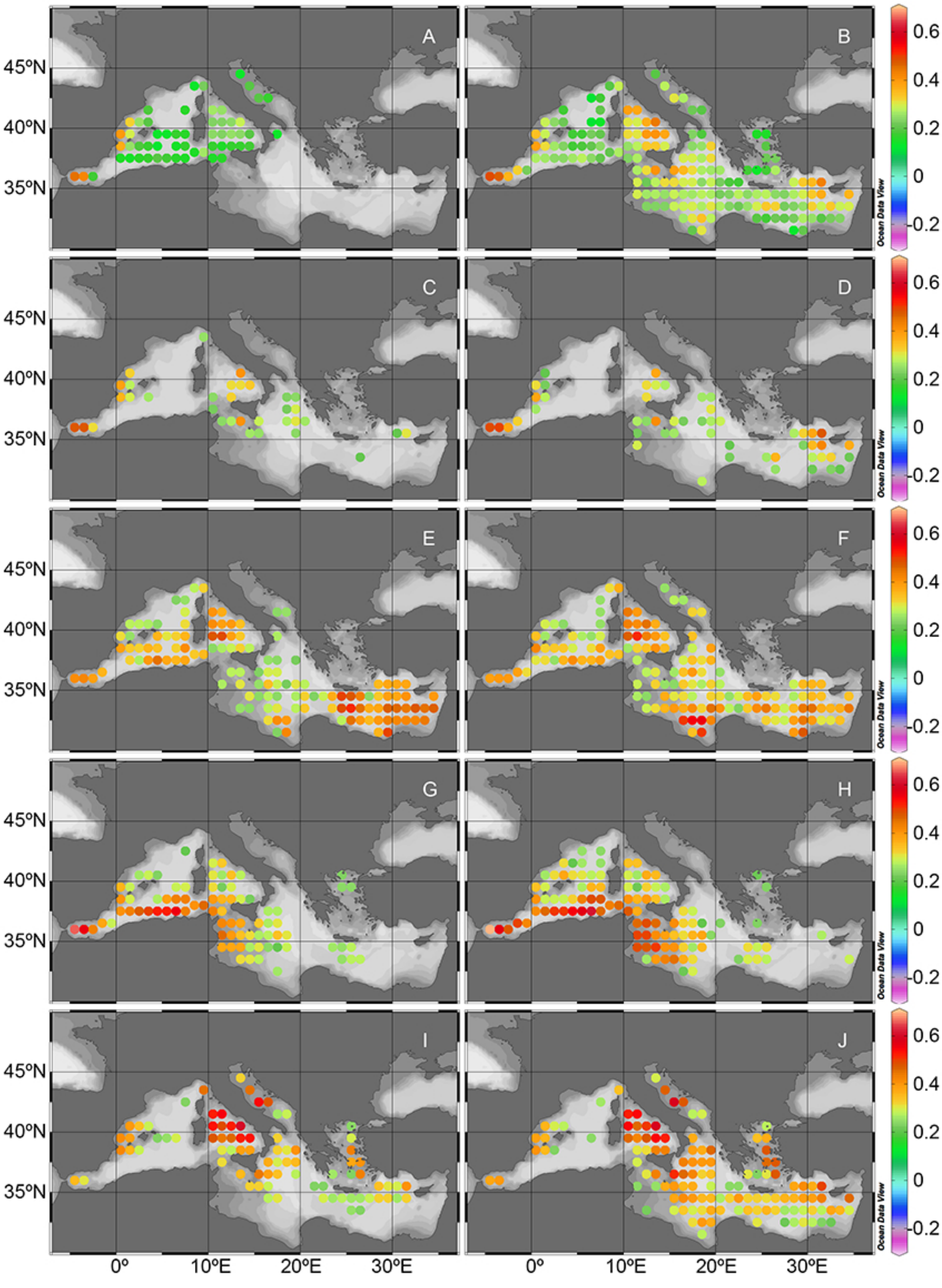

Figure S2. Correlation between dust deposition and aerosol optical thickness. Statistically significant (p<0.05) correlation coefficient (r) between dust deposition and aerosol optical thickness (left panels) and between seasonally detrended dust deposition and seasonally detrended aerosol optical thickness (right panels) for the whole time series and for different seasons. Panels: a, b) annual; c, d) winter (January to March); e, f) spring (April to June); g, h) summer (July to September) and i, j) autumn (October to December).

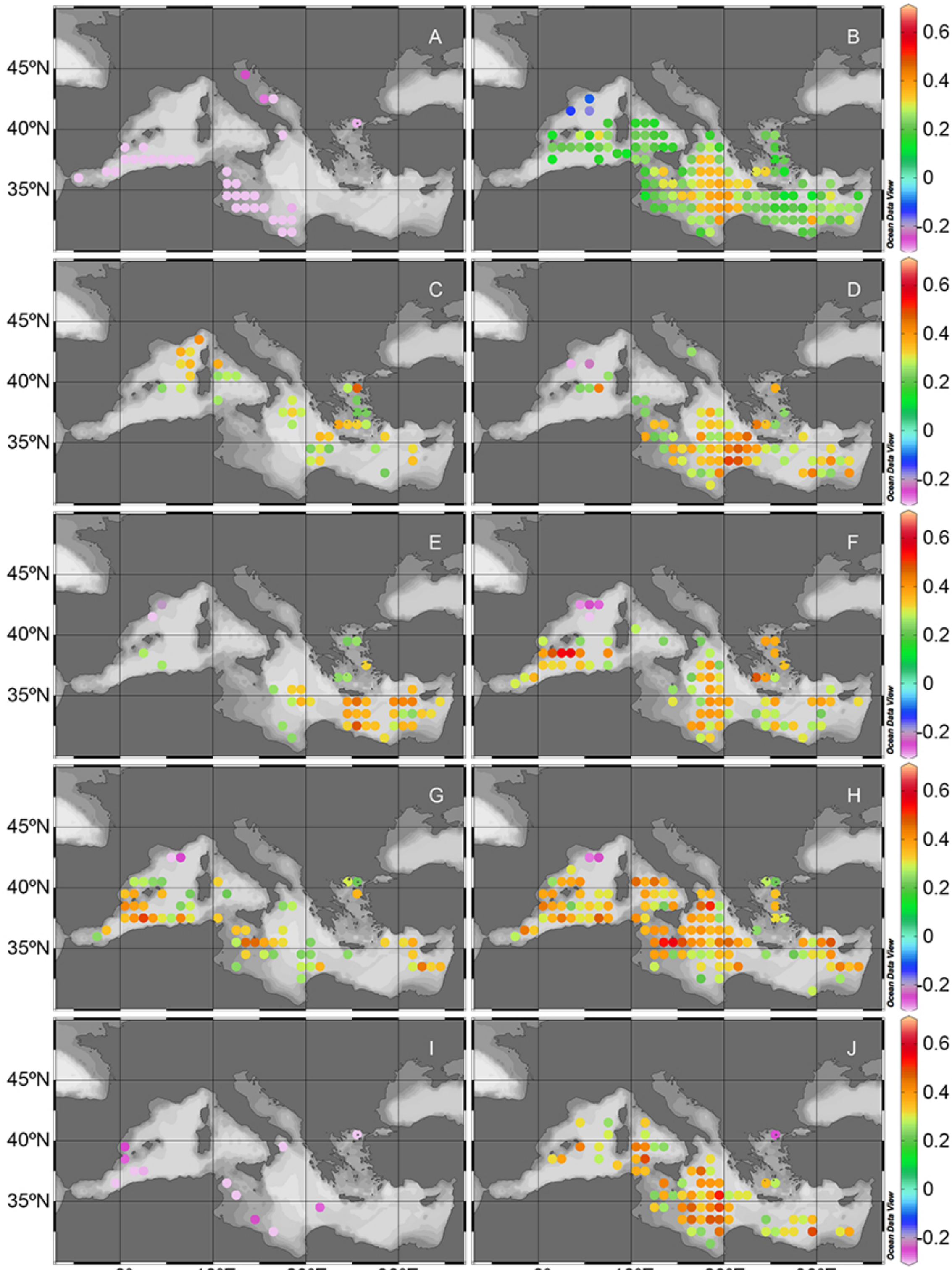

Figure S3. Correlation between chlorophyll concentration and aerosol optical thickness. Same as Fig. S2 but for chlorophyll concentration versus aerosol optical thickness.

Cell nº	Latitude	Longitude	Cell nº	Latitude	Longitude	Cell nº	Latitude	Longitude
1	44,5	13,5	61	38,5	17,5	**121**	34,5	15,5
2	43,5	8,5	62	38,5	18,5	**122**	34,5	16,5
3	43,5	9,5	63	38,5	19,5	**123**	34,5	17,5
4	43,5	14,5	64	38,5	25,5	**124**	34,5	18,5
5	42,5	4,5	65	38	8,5	**125**	34,5	19,5
6	42,5	5,5	66	38	9,5	**126**	34,5	20,5
7	42,5	6,5	67	37,5	0,5	**127**	34,5	21,5
8	42,5	7,5	68	37,5	1,5	**128**	34,5	22,5
9	42,5	15,5	69	37,5	2,5	**129**	34,5	23,5
10	42,5	16,5	70	37,5	3,5	**130**	34,5	24,5
11	41,5	3,5	71	37,5	4,5	**131**	34,5	25,5
12	41,5	4,5	72	37,5	5,5	**132**	34,5	26,5
13	41,5	5,5	73	37,5	6,5	**133**	34,5	27,5
14	41,5	6,5	74	37,5	7,5	**134**	34,5	28,5
15	41,5	7,5	75	37,5	10,5	**135**	34,5	29,5
16	41,5	10,5	76	37,5	11,5	**136**	34,5	30,5
17	41,5	11,5	77	37,5	16,5	**137**	34,5	31,5
18	41,5	17,5	78	37,5	17,5	**138**	34,5	34,5
19	41,5	18,5	79	37,5	18,5	**139**	33,5	12,5
20	40,5	1,5	80	37,5	19,5	**140**	33,5	13,5
21	40,5	2,5	81	37,5	25,5	**141**	33,5	14,5
22	40,5	3,5	82	37,5	26,5	**142**	33,5	15,5
23	40,5	4,5	83	36,5	-1,5	**143**	33,5	16,5
24	40,5	5,5	84	36,5	-0,5	**144**	33,5	17,5
25	40,5	6,5	85	36,5	11,5	**145**	33,5	18,5
26	40,5	7,5	86	36,5	12,5	**146**	33,5	19,5
27	40,5	10,5	87	36,5	13,5	**147**	33,5	20,5
28	40,5	11,5	88	36,5	15,5	**148**	33,5	21,5
29	40,5	12,5	89	36,5	16,5	**149**	33,5	22,5
30	40,5	13,5	90	36,5	17,5	**150**	33,5	23,5
31	40,5	24,5	91	36,5	18,5	**151**	33,5	24,5
32	40,5	25,5	92	36,5	19,5	**152**	33,5	25,5
33	39,5	0,5	93	36,5	20,5	**153**	33,5	26,5
34	39,5	1,5	94	36,5	23,5	**154**	33,5	27,5
35	39,5	4,5	95	36,5	24,5	**155**	33,5	28,5
36	39,5	5,5	96	36,5	25,5	**156**	33,5	29,5
37	39,5	6,5	97	36,5	26,5	**157**	33,5	30,5
38	39,5	7,5	98	36	-4,5	**158**	33,5	31,5
39	39,5	10,5	99	36	-3,5	**159**	33,5	32,5
40	39,5	11,5	100	36	-2,5	**160**	33,5	33,5
41	39,5	12,5	101	35,5	11,5	**161**	33,5	34,5
42	39,5	13,5	102	35,5	12,5	**162**	32,5	16,5
43	39,5	14,5	103	35,5	13,5	**163**	32,5	17,5

44	39,5	17,5	104	35,5	14,5	**164**	32,5	18,5
45	39,5	18,5	105	35,5	15,5	**165**	32,5	19,5
46	39,5	24,5	106	35,5	16,5	**166**	32,5	24,5
47	39,5	25,5	107	35,5	17,5	**167**	32,5	25,5
48	38,5	0,5	108	35,5	18,5	**168**	32,5	26,5
49	38,5	1,5	109	35,5	19,5	**169**	32,5	27,5
50	38,5	2,5	110	35,5	20,5	**170**	32,5	28,5
51	38,5	3,5	111	35,5	21,5	**171**	32,5	29,5
52	38,5	4,5	112	35,5	22,5	**172**	32,5	30,5
53	38,5	5,5	113	35,5	28,5	**173**	32,5	31,5
54	38,5	6,5	114	35,5	29,5	**174**	32,5	32,5
55	38,5	7,5	115	35,5	30,5	**175**	32,5	33,5
56	38,5	10,5	116	35,5	31,5	**176**	31,5	17,5
57	38,5	11,5	117	34,5	11,5	**177**	31,5	18,5
58	38,5	12,5	118	34,5	12,5	**178**	31,5	28,5
59	38,5	13,5	119	34,5	13,5	**179**	31,5	29,5
60	38,5	14,5	120	34,5	14,5			

Table S1.

Geographical coordinates for the 1°×1° grid cells analyzed in this study. Coordinates refer to the central point of the cell. Latitudes are all North. Positive longitudes are East and negative longitudes West.

Gallisai, R., Volpe, G., Peters, F., (2016).
Large Saharan dust storms:implications for chlorophyll dynamics in the Mediterranean Sea. Globalbiogeochemicalcycles. Submitted

Abstract

We investigate the large (LDE) and very large (VLDE) Saharan dust deposition events occurred between 2000 and 2007 and their short-term impact on the marine phytoplankton dynamics in the Mediterranean Sea. A total of 153 LDE were identified. Events were more frequent during winter in the Eastern Mediterranean followed by autumn, when they affected both the Western and the Central Mediterranean. The LDE average frequency is of 19 events per year, unevenly distributed over the years. Most of the 31 VLDE occurred during winter and autumn in the Central Mediterranean. The chlorophyll response to dust addition was investigated during the VLDE events as a proxy for phytoplankton response to short-term atmospheric nutrient stimulation. Chlorophyll increases of up to 345% were recorded in the Western Mediterranean, up to 146% and 121% in the Central and Eastern Mediterranean respectively. Overall, chlorophyll response behavior is quite heterogeneous reasonably due to both the uniqueness of each VLDE and the ecological differences among the Mediterranean areas in terms of the phytoplankton community structure and the interaction between bacteria and phytoplankton for new resources. An eastward decreasing trend in chlorophyll response is observed that conforms to the relative importance of bacteria with respect to phytoplankton and other biogeochemical trends. The increase of mineral aerosols with increased aridity in the region together with the decrease of the mixed layer depth of the oceans should increase the importance of aerosols fueling marine production.

Introduction

Deserts cover one fifth of Earth's land surface, equal to an area of 49 million Km^2. It is estimated that the global desert dust production is about 1536 $Tg \cdot yr^{-1}$ [Ginoux et al., 2012]. The Sahara desert is the world's largest desert region, extending from the Atlantic Ocean to the Red Sea. It covers an area of about 9 million km^2 with an annual dust production of 840 $Tg \cdot yr^{-1}$, accounting for 55% of the global dust emissions [Ginoux et al., 2012]. In the Northern part, the Sahara desert limits with the Mediterranean Sea, which is a quasi-enclosed basin characterized for containing one of the most oligotrophic waters in the world [UNEP, 1989; UNEP/FAO/WHO, 1996]. The Mediterranean seawaters are thus part of the ca. 60% of the global ocean showing low-nutrient low-chlorophyll characteristics [Longhurst, 1995]. The seasonal dynamics of surface chlorophyll in the Mediterranean show an increase in winter-spring following winter mixing with deeper nutrient-richer waters. In the Western Mediterranean bloom dynamics similar to the North Atlantic occur while the Eastern Mediterranean shows much more inconspicuous increases and has even been defined as non-blooming *[D'Ortenzio and Ribera D'Alcalà, 2009; Volpe et al., 2012]*. Atmospheric sources of nutrients [Paerl, 1997; Ternon et al., 2011] have attracted the attention of the scientific community during the last two decades in order to explain, at least partially, the nutrient starvation relief in the surface ocean. *Nutrient* is here used in a broad sense without discerning specific macro- or micro-elements. The atmospheric inputs into the Mediterranean area, derived mainly from the Sahara and Middle East [Pey et al., 2013], could be the principal nutrient sources for the surface depleted waters of the Mediterranean Sea, especially during the stratification period [Bonnet et al., 2005; Ternon et al., 2011] when nutrient concentrations are at their lowest.

The amount of nutrients that reach the Mediterranean Sea through aerosol deposition has been demonstrated to have important consequences for the biogeochemical budget of the basin over geological time scales [Ludwig et al., 2010]. On the contrary, its effect on phytoplankton dynamics at ecological time scales is not so obvious. Several studies have been performed testing the effects of Saharan dust deposition on phytoplankton community, either in the Western [Bonnet et al., 2005; Laghdass et al., 2011; Ridame et al., 2014; Romero et al., 2011], the Eastern [Eker-Develi et al., 2006; Herut et al., 2005] or the entire Mediterranean Sea..[Gallisai et al., 2012; Gallisai et al., 2014; Ternon et al., 2011; Volpe et al., 2009] *In situ* microcosm and mesocosm experiments (Bonnet et al. [2005]; Laghdass et al. [2011] and Ternon et al. [2011]) found that phytoplankton cells were generally stimulated by the dust. Romero et al. [2011] reported that the response was quicker for bacteria, subsequently affecting the response of algal cells. Ridame et al. [2014] found no response to a simulated dry deposition event and a clear stimulation to a simulated wet deposition event. Eker-Develi et al. [2006] monitored a couple of coastal stations and found mixed results regarding the response of phytoplankton to dust deposition episodes. Using satellite data, Volpe et al. [2009] did not observe changes in chlorophyll concentration while Gallisai et al. [2014] found small but significant correlations between dust deposition and chlorophyll in some areas and times of the year. Thus, the response of phytoplankton to atmospheric nutrient addition does not show clear patterns. This might be due to different factors such as the biophysical state of the phytoplankton community at the time of dust deposition, or the chemical composition and state of the atmospheric dust, or both.

Saharan dust deposition over the Mediterranean occurs 20 to 37% of the days with a decreasing gradient to the north [Pey et al., 2013]. It is important to discriminate between the effect of this almost continuous but generally slight deposition [Gallisai et al., 2014] and the effect of short-term large dust deposition events that have the potential of a larger impact.

The goal of the present study is to analyze the geographical and seasonal variability of large Saharan dust deposition events (LDE), as well as the changes in chlorophyll concentration to synoptically very large dust deposition events (VLDE) occurred during an 8 year-period (2000 to 2007) in the Mediterranean Sea. The Eastern Mediterranean, with its ultraoligotrophic conditions, receives overall more dust than the Western Mediterranean and we would expect a larger response of chlorophyll to dust deposition events. However, our data and approach does not allow discerning the chemical macro- or micro-nutrients in the dust that eventually trigger these changes.

Materials and methods:

Chlorophyll data. Daily satellite Level-1A chlorophyll concentration data (mg m^{-3}) were derived from SeaWIFS. Data were acquired from 2000 to 2007 and then reprocessed to Level-3 using the MedOC4 regional algorithm [Volpe et al., 2007], which takes into account the peculiar Mediterranean Sea color. In addition, we used the same filtering procedure presented in *Gallisai et al.* [2014], which considers the possible disturbance by dust mimicking chlorophyll concentration values [*Moulin et al.*, 2001]. The 2000-2007 temporal range is the one during which SeaWiFS data were constantly available without any significant gap. Since the scope of the work is to evaluate the impact of large and very large deposition events on the phytoplankton dynamics, to reduce the low scale variability, data for the entire Mediterranean basin, were regridded at 1º spatial resolution, for a total of 179 grid-cells.

Dust deposition. For the present study, daily Saharan dust deposition output data (kg m^{-2} d^{-1}) were obtained from the BSC-DREAM8b model simulation [*Pérez et al.*, 2006a; *Pérez et al.*, 2006b]. Data were gathered for the same time period and grid resolution as for chlorophyll concentration. The model simulated the dust concentration field in the troposphere, taking into account all principal dust life cycle processes. Improvements to previous versions of the model include a dust source function based on the 1-km USGS land use dataset, a particle size distribution in eight categories (0.1 to 1 µm), a source size distribution derived from *D'Almeida* [1987], and dust radiative feedbacks [*Pérez et al.*, 2006a]. Further information about the model features can be found in *Nickovic et al.* [2001], *Pérez et al.* [2006a] and *Basart et al.* [2012]. BSC-DREAM8b has been used both as a forecasting and research tool in North Africa and the Mediterranean [*Alonso-Perez et al.*, 2011; *Amiridis et al.*, 2009; *Gallisai et al.*, 2014; *Pay et al.*, 2012; *Pey et al.*, 2013]. Several studies have checked the goodness about the horizontal and vertical extension of dust plume [*Papanastasiou et al.*, 2010; *Pérez et al.*, 2006b]. Daily near-real time satellite and sun-photometer observational data are used in the operational model evaluation.

Dust deposition events. The identification of dust deposition events was made as follows. For each grid-cell, we calculated the mean and standard deviation (SD) for the whole time series after a $\log_{10}$ transformation. Then, we identified the days showing values larger that 5·SD. A large deposition event (LDE) was defined as a day that showed at least 5 grid-cells with values above 5·SD. Similarly, a very large deposition event (VLDE) was defined as a day that showed at least 20 grid-cells with values larger than 5·SD. This means that all VLDE were also contained in LDE. LDE were used for temporal and geographical analyses of deposition events.

LDE were largely coherent in space, showing mostly adjoining grid-cells. All VLDE presented spatially coherent areas and coincided with true-colour satellite images of aerosols over the Mediterranean (Fig. S1). In addition, we checked that air masses close to the surface (between 100 and 500 m) were originating in Africa using back-trajectories obtained with the HYSPLIT model (Fig. S1).

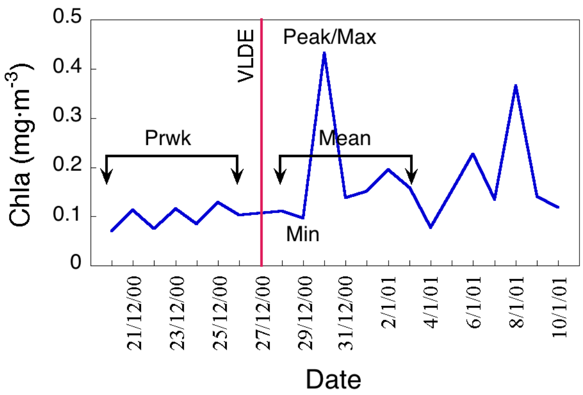

Figure 1. Example of chlorophyll concentration diagram used to analyze the chlorophyll variation following a very large dust deposition event. The vertical line marks the very large dust deposition event (VLDE).

Chlorophyll trends. For each VLDE, we analyzed the chlorophyll time series. First, for each grid-cell of the VLDE we selected the chlorophyll time series for 22 days (7 days prior to the day of the VLDE and 14 days after the day of the VLDE). Then, for each of the 22 days of the VLDE we filtered the data to remove the values of the cells that were above or below 3·SD in chlorophyll, using $\log_{10}$ transformed data. Next, we computed the area averaged chlorophyll for each day and obtained the 22 days chlorophyll time series for the event. From the time series we obtained the day and value of the chlorophyll peak (Peak) after the VLDE, and we also computed the average of the chlorophyll for the week previous (Prwk) to the VLDE, and the average (Mean) and maximum value (Max) of chlorophyll for the week after the VLDE (Fig. 1). The Peak was defined as the first maximum following a minimum right after one dust deposition event.

Finally, we computed the percent change in chlorophyll for the peak, the maximum value and the average value after the VLDE with respect to the average chlorophyll before the VLDE as follows:

$$\Delta\text{Peak} = \frac{\text{Peak} - \text{Prwk}}{\text{Prwk}} \cdot 100$$

$$\Delta\text{Max} = \frac{\text{Max} - \text{Prwk}}{\text{Prwk}} \cdot 100;\ \Delta\text{Mean} = \frac{\text{Mean} - \text{Prwk}}{\text{Prwk}} \cdot 100$$

We also did the whole process using a seasonally-detrended chlorophyll, that is, using the residuals after subtracting the daily climatic mean (obtained from the 8-year time series). In this case, the actual chlorophyll was used in the denominator to calculate the increments. We call this data set *detrended*. We used all VLDE and the matched pair statistical test of the software package JMP (Version 10, SAS Institute Inc., Cary, NC, USA) to see if there were statistical differences between the values of Peak, Prwk, Max and Mean for both the chlorophyll and detrended-chlorophyll data sets. We also performed stepwise model selection and used generalized linear models to see if chlorophyll responses across VLDE could be related to other parameters such as geographic location, season or the intensity of dust deposition. In all cases, the statistical significance level was set at p-values < 0.05.

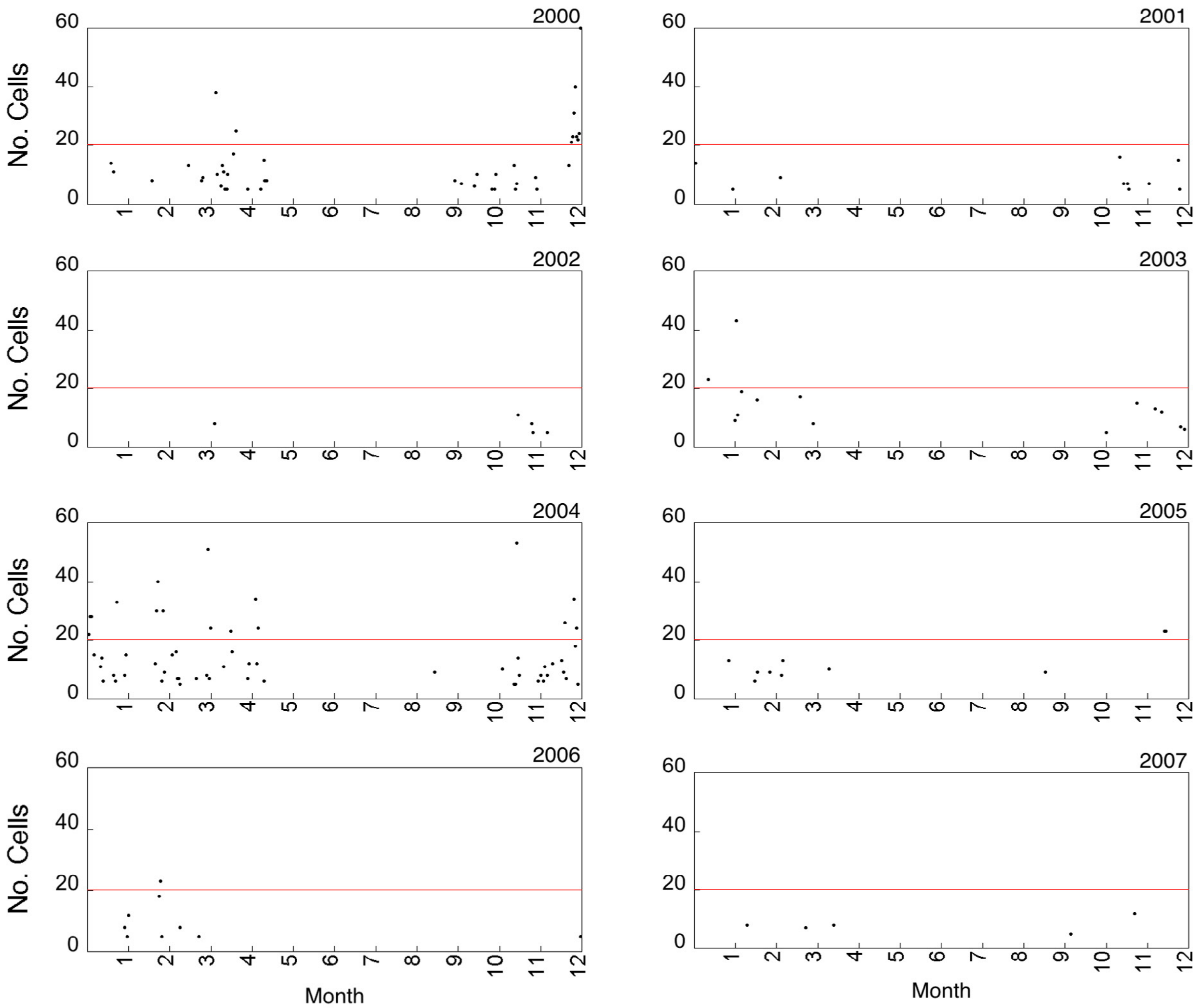

Figure 2. Dust deposition events (LDE) and number of grid-cells affected by them, between 2000 and 2007 in the Mediterranean Sea. The red line indicates the cutoff for VLDE.

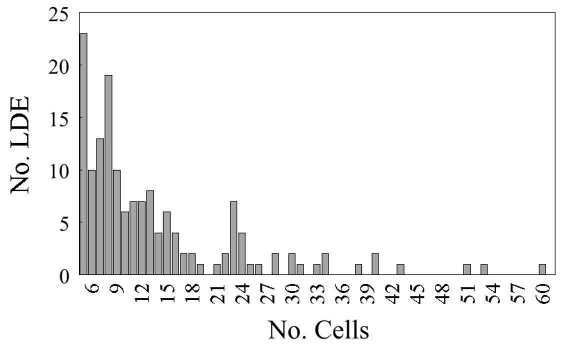

Figure 3. Relationship between grid-cells affected by dust deposition and number of LDE.

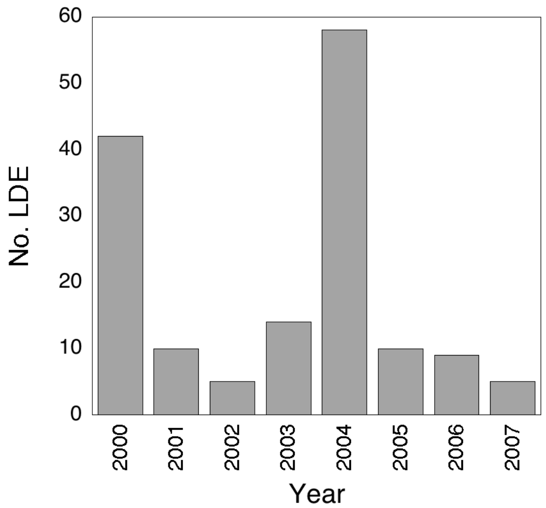

Figure 4. Number of LDE between 2000 and 2007.

Results

A total number of 153 LDE occurred during the period between 2000 and 2007 (Fig. 2). The LDE involved between 5 (set artificially as the lower limit) and 60 grid-cells over the Mediterranean Sea. As one would expect, the frequency of LDE generally decreases with the number of affected grid-cells (Fig. 3). This is mostly the case with the largest frequency for the 5 grid-cell lower limit, although there appear to be some exceptions centered around 8 and 13 and 23 grid-cells with a higher frequency than expected. 50% of the LDE involved less than 10 cells. Traditionally, only the largest events have been looked at such as those covering wide ocean regions but we show that there is a large number of deposition events affecting smaller but still relevant areas.

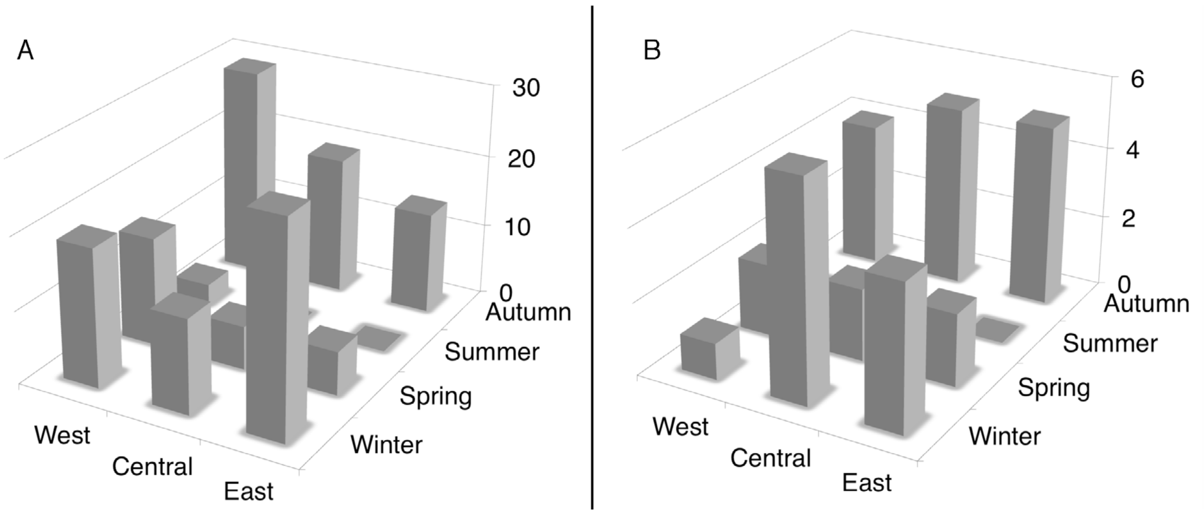

Figure 5. Distribution of LDE (A plot) and VLDE (B plot) by geographical area and season.

Many of the LDE occurred in years 2000 and 2004, especially in autumn and winter (Fig. 4).

In particular, 62 LDE took place in autumn, 61 in winter, 27 in spring and only 3 in summer (Fig. 5a). In general, the occurrence of LDE showed some variability. Depending on the year and season they affected different parts of the Mediterranean Sea.

In 2000, the Western Mediterranean was the most affected region and the majority of events occurred in autumn. In contrast, during 2004 the Central Mediterranean was the most affected region and mainly in winter. The only 3 summer LDEs occurred in the Western Mediterranean. In general, the Western Mediterranean was the area with most LDE, followed by the Central and then the Eastern Mediterranean (Fig. 5a). While the VLDE (Fig. 5b) followed the same seasonal pattern as LDE with most events occurring in autumn (14) and winter (12) and no events in summer, the geographical distribution was different with most events in the Central and Eastern Mediterranean (13 and 11 respectively) and substantially less in the Western (7). For each of the 31 VLDE chlorophyll dynamics (Fig. 6), the magnitude of dust deposition, and the geographical and seasonal distributions were analyzed. For most of the events (27 of 31), Peak, Max and Mean were larger than Prwk (Table 1).

The results of the matched pair t-tests (Table 2), show significant chlorophyll peaks after VLDE with respect to the week previous to the events, even though differences are smaller than 0.1 mg m^{-3}. This is taken as an indication of phytoplankton stimulation after the VLDE.

The higher values of chlorophyll after the VLDE are not due to a single peak day since the dynamics of chlorophyll tend to be of increasing chlorophyll beyondthe first peak as the maximum value of chlorophyll after VLDE is significantly larger the mean Peak. The Mean of chlorophyll after the VLDE tends to be larger than Prwk although it is only significantly different when using the non-parametric sign-rank test. This occurs because of the event of 26/02/2004 with a negative Peak that influences the parametric statistics. In fact if this event is discarded out as an outlier, all matched pairs show significance at a $p < 0.001$ when using parametric tests. In any case, the difference between Mean and Prwk is quite small, indicating a fast turnover of the produced chlorophyll after the VLDE. The analyses are almost identical when seasonally-detrended chlorophyll was used so that rising trends over time could not be attributed to underlying seasonal dynamics.

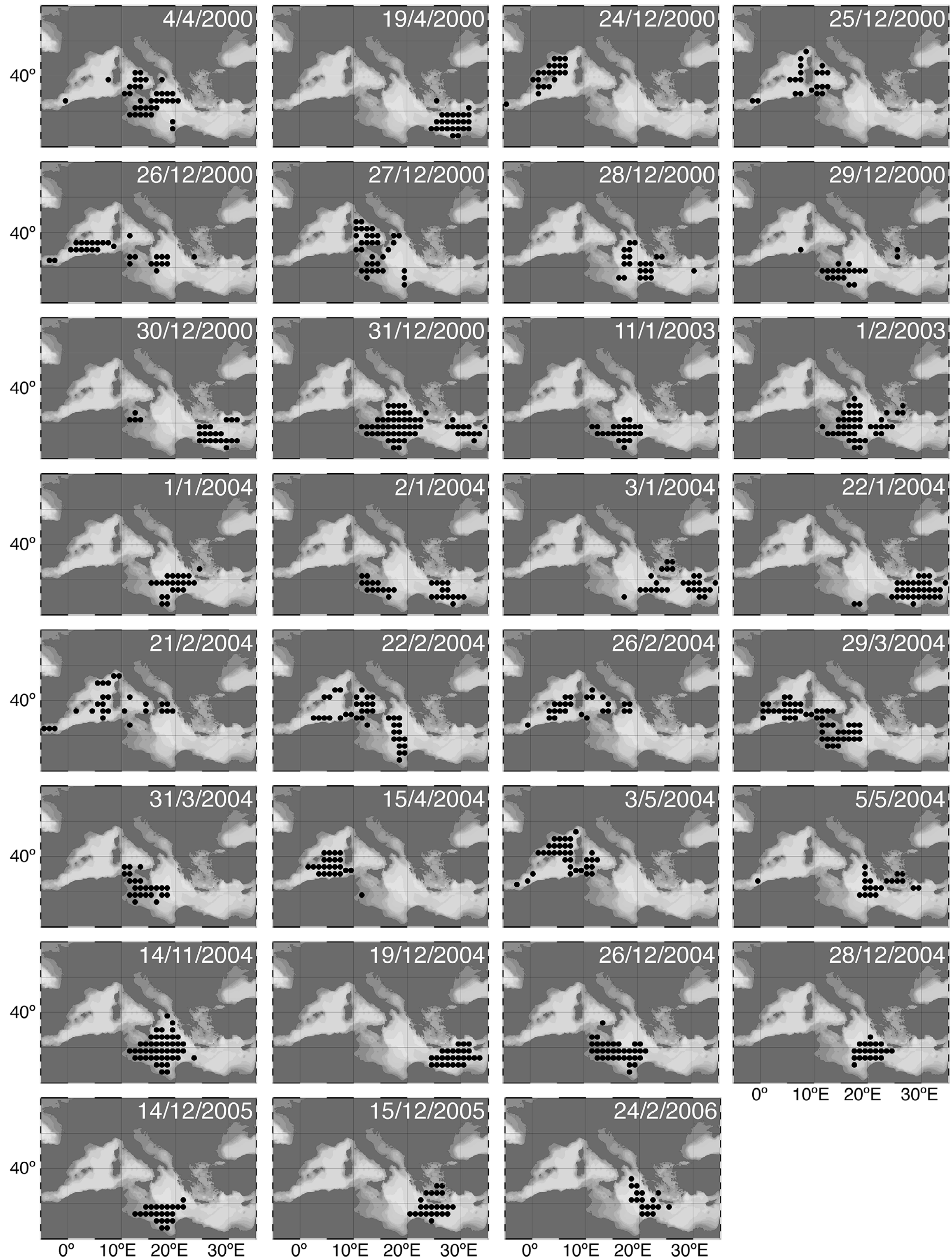

Figure 6. Maps of the geographical distribution of very large deposition events (VLDE) occurred between 2000 and 2007.

Table 1. Chronological list of VLDE events occurred between 2000 and 2007. Season *A* is for autumn, *S* for spring and *W* for winter. *Med* refers to the Mediterranean sub-basin affected by the events (W for west, C for central or E for east). *Dep* shows the amount of modeled dust deposited for each event (kg m^{-2}). For each VLDE event, the mean chlorophyll value of the week previous to the VLDE (Prwk), the chlorophyll peak value (Peak), the maximum chlorophyll value recorded after the VLDE (Max), the mean chlorophyll value of the week after the VLDE event (Mean) (all chlorophyll values are in mg m^{-3}) and the number of days after the event when the chlorophyll peak appeared (Peakd) are shown. The Δ columns correspond to the different increases in percentage. All parameters were calculated for chlorophyll (Chlorophyll) and for residual chlorophyll after a seasonal detrended (detrended Chlorophyll). In the latter case, Δ are calculated from the residuals with respect to Chlorophyll Prewk.

							Chlorophyll								detrended Chlorophyll			
Date	**Season**	**Med**	**Dep**	**Prewk**	**Peak**	**Max**	**Mean**	**ΔPeak**	**ΔMax**	**ΔMean**	**Peakd**	**Peak**	**Max**	**Mean**	**ΔPeak**	**ΔMax**	**ΔMean**	**Peakd**
4/4/00	**S**	**C**	3.10E-04	0.09	0.1	0.25	0.13	19%	181%	46%	3	0.02	0.11	0.03	49%	147%	54%	3
19/4/00	**S**	**E**	3.14E-04	0.05	0.06	0.07	0.06	25%	49%	13%	3	0.02	0.03	0.01	27%	42%	15%	3
24/12/00	**A**	**W**	1.04E-04	0.2	0.33	0.43	0.27	66%	117%	37%	3	0.15	0.28	0.11	51%	117%	29%	2
25/12/00	**A**	**W**	1.19E-04	0.12	0.25	0.27	0.2	103%	119%	61%	5	0.14	0.14	0.04	103%	103%	17%	4
26/12/00	**A**	**W**	2.31E-04	0.11	0.32	0.32	0.21	200%	200%	97%	3	0.24	0.24	0.07	224%	224%	70%	3
27/12/00	**A**	**W**	3.41E-04	0.1	0.43	0.43	0.18	345%	345%	86%	3	0.34	0.34	0.08	343%	343%	83%	3
28/12/00	**A**	**E**	2.36E-04	0.06	0.14	0.14	0.09	113%	113%	39%	2	0.08	0.08	0.02	127%	127%	28%	2
29/12/00	**A**	**C**	4.20E-04	0.1	0.26	0.26	0.13	146%	146%	27%	1	0.27	0.27	0.08	268%	268%	80%	1
30/12/00	**A**	**E**	2.46E-04	0.07	0.14	0.14	0.09	109%	109%	35%	2	0.03	0.03	0.01	49%	49%	18%	2
31/12/00	**A**	**C**	4.63E-04	0.08	0.11	0.11	0.08	33%	33%	-2%	1	0.04	0.04	0.01	27%	27%	-10%	1
11/1/03	**W**	**C**	4.23E-04	0.06	0.09	0.11	0.09	44%	78%	38%	3	0.02	0.04	0.01	20%	60%	8%	4
1/2/03	**W**	**C**	3.17E-04	0.09	0.15	0.15	0.12	65%	65%	37%	4	0.05	0.05	0.03	58%	58%	37%	3
1/1/04	**W**	**C**	3.01E-04	0.07	0.1	0.1	0.08	37%	37%	16%	3	0.03	0.03	0.01	36%	36%	13%	3
2/1/04	**W**	**E**	4.87E-04	0.08	0.18	0.18	0.11	121%	121%	28%	2	0.06	0.06	0.03	48%	48%	15%	2
3/1/04	**W**	**E**	2.83E-04	0.07	0.09	0.09	0.07	28%	28%	7%	5	0.02	0.03	0.01	15%	26%	0%	5
22/1/04	**W**	**E**	5.84E-04	0.07	0.1	0.11	0.09	35%	50%	30%	2	0.04	0.04	0.02	48%	48%	27%	7
21/2/04	**W**	**W**	1.77E-04	0.18	0.31	0.35	0.27	75%	98%	52%	5	0.16	0.16	0.05	91%	91%	26%	1
22/2/04	**W**	**C**	2.45E-04	0.09	0.1	0.25	0.13	19%	181%	46%	3	0.02	0.11	0.03	49%	147%	54%	3
26/2/04	**W**	**C**	1.72E-04	0.68	0.26	0.26	0.19	-61%	-61%	-71%	2	0.1	0.1	0.03	-63%	-63%	-74%	2
29/3/04	**W**	**C**	2.95E-04	0.15	0.17	0.17	0.12	13%	13%	-19%	2	0.05	0.05	0.03	20%	20%	6%	3
31/3/04	**W**	**C**	2.99E-04	0.09	0.12	0.15	0.1	28%	62%	6%	3	0.06	0.07	0.02	51%	65%	19%	3
15/4/04	**S**	**W**	1.62E-04	0.08	0.08	0.12	0.08	-4%	39%	2%	5	-0.01	0.01	-0.01	19%	51%	19%	5
3/5/04	**S**	**W**	9.07E-05	0.12	0.16	0.16	0.12	41%	41%	7%	2	0.04	0.08	0.03	47%	82%	38%	4
5/5/04	**S**	**E**	3.49E-04	0.06	0.08	0.1	0.07	32%	67%	16%	4	0.03	0.07	0.02	24%	93%	14%	5
14/11/04	**A**	**C**	7.31E-04	0.06	0.08	0.08	0.07	20%	20%	6%	6	0.03	0.03	0.02	56%	56%	38%	6
19/12/04	**A**	**E**	3.74E-04	0.05	0.07	0.07	0.05	35%	35%	13%	6	0.01	0.01	0	26%	26%	4%	6
26/12/04	**A**	**C**	5.24E-04	0.1	0.2	0.2	0.1	90%	90%	0%	2	0.06	0.06	0.01	46%	46%	1%	2
28/12/04	**A**	**E**	4.22E-04	0.06	0.07	0.07	0.06	22%	22%	6%	3	0	0	0	12%	12%	0%	3
14/12/05	**A**	**C**	6.23E-04	0.04	0.09	0.09	0.07	101%	101%	50%	3	0.02	0.02	0.01	49%	57%	30%	1
15/12/05	**A**	**E**	3.55E-04	0.04	0.06	0.09	0.06	36%	94%	37%	2	0	0.02	0	29%	65%	18%	2
24/2/06	**W**	**E**	2.69E-04	0.1	0.12	0.12	0.11	18%	18%	8%	5	0.04	0.04	0.03	19%	19%	6%	5

Regarding individual VLDE, chlorophyll concentration peaks were generally observed between days 1 and 6 (with a mean of 3.16 days) after the mineral dust deposition event (Table 1). Similarly, when we analyzed the seasonally detrended chlorophyll data, chlorophyll peaks appeared between days 1 and 7 (mean of 3.20 days). Peak days were not significantly different either across Mediterranean areas or between seasons (Table 3).

µD	Chla	detrended Chla
Peak-Prwk	0.048 (*/***)	0.05 (*/***)
Max-Prwk	0.067 (**/***)	0.066 (**/***)
Mean-Prwk	0.009 (ns/***)	0.007 (ns/***)
Max-Peak	0.019 (**/***)	0.016 (**/***)
Mean-Peak	-0.038 (***/***)	-0.043 (***/***)
Mean-Max	-0.058 (***/***)	-0.059 (***/***)

Table 2.

Matched-pair results between Peak, Max, Mean and Prwk for the VLDE. Differences are shown and in parenthesis the statistical significance for the parametric / non-parametric Wilcoxon signed rank test. Chlorophyll units are mg m^{-3}. The null hypothesis, H_0, is that $\mu_D = 0$, where μ_D is the mean population difference of the matched pair. When significant, p-values are as follows: $p < 0.001$ (***), $p < 0.01$ (**), $p < 0.05$ (*).

Positive DPeak chlorophyll increments range from 13% to 345% for the different outbreaks, with only two events showing a slight decrease (Table 1).

The smallest positive DPeak occurs in the Central Mediterranean during the winter season and in the Eastern Mediterranean in autumn (for detrended data) while the largest occurs in the Western in autumn (Table 1). On the contrary, the minimum chlorophyll concentration in mg·m^{-3} (Peak) was recorded in the Eastern Mediterranean during autumn and the maximum in the Central Mediterranean also during autumn (Table 1). The six largest deposition events (in extension) occurred in the Central Mediterranean with a DPeak ranging from 13% to 65% (Table 1). In terms of dust deposition intensity (kg·m^{-2} d^{-1}), the 12 first largest events were distributed between the Central and Eastern Mediterranean, mainly in autumn, with DPeak ranging from 20% to 46%. Deposition intensity in the Western Mediterranean was lowest with the exception of the VLDE of 27/12/2000 that resulted in the largest DPeak of the set at 345% (Table 1). Using generalized lineal models, the response of chlorophyll (ΔPeak, ΔMax, ΔMean) to VLDE were not statistically related to the magnitude of the dust outbreak, either in deposited dust per unit area or in the extension of the

Mediterranean Region	Chla	detrended Chla	Season	Chla	detrended Chla
Western	3.71 -0.47	3.14 -0.51	Spring	3.33 -0.42	3.83 -0.4
Central	2.77 -0.36	2.69 -0.38	Autumn	3 -0.43	2.71 -0.44
Eastern	3.27 -0.45	3.82 -0.55	Winter	3.27 -0.39	3.45 -0.51

Table 3.

Means and standard error in parenthesis for peak days (number of days) after the VLDE as grouped by geographical area and season. In no case were peak days different using either parametric tests (ANOVA means test) or non-parametric tests (Wilcoxon/Kruskal-Wallis).

affected surface. It was also statistically unrelated to the chlorophyll before the VLDE. Some of the responses were related the time of the year (SEASON) and to a longitudinal gradient (Table 4) SEASON appeared to be significant for ΔPeak with autumn showing larger responses than spring, and winter indistinguishable from either one. Using seasonally detrended data, the importance of seasonality decreases in favor of the longitudinal (LON) gradient of decreasing response intensity from west to east, although ΔPeak is almost significant for SEASON.

Discussion

Large and very large deposition events show patterns of increased frequency in autumn and winter. As for the geographical distribution, the Western Mediterranean is more frequently affected by LDE then the Eastern, although VLDE are more prominent in the Central and Eastern Mediterranean, including the events with the largest dust deposition loads. It is the Central Mediterranean that has the highest proportion (34%) of VLDE to LDE, followed by the Eastern (22%) and the Western (11%) Mediterranean. The fact that the Eastern Mediterranean has less LDE reflects a more continuous and constant supply of dust [*Gallisai et al.*, 2014]. Our results are in agreement with previous studies highlighting that the frequency and the magnitude of dust outbreaks have a wide spatial, seasonal and inter-annual variability across the Mediterranean Sea [*Gallisai et al.*, 2012; *Moulin et al.*, 1998; *Pey et al.*, 2013; *Swap et al.*, 1996; *Varga et al.*, 2014].

	Chlorophyll			detrended Chlorophyll		
	ΔPeak	ΔMax	ΔMean	ΔPeak	ΔMax	ΔMean
Model P value	0.0233*	0.2485	0.1379	0.00194*	0.0296*	0.416
SEASON	0.0173*	0.344	0.0773	0.0505	0.3637	0.7719
Tukey test (SEASON)	AW; WS					
LON	0.2623	0.1672	0.3861	0.0283*	0.0104*	0.1413

Table 4.

Generalized linear model results. The ΔPeak, the ΔMax and the ΔMean of chlorophyll and seasonally detrended chlorophyll data were tested with categorical seasonal (SEASON) and continuous geographical (LON) factors. P-values are reported for the whole model and for each independent variable. The values marked with * show a p-value equal or less than 0.05. A = autumn, W = winter and S = spring.

Part of this variability is linked to the complexities of the atmospheric transport. The dust transport over the Mediterranean Sea is caused by different meteorological situations. In the Western sub-basins, the dust outbreaks are controlled by the summer northward migration of the subtropical high-pressure belt [*Aschmann*, 1973]. In the Eastern basin, they are generated by low-pressure systems as mid-latitude Mediterranean and Sharav cyclones [*Alpert and Ziv*, 1989]. In the Central part, both Western and Eastern outbreaks converge. The area shows characteristics of both sub-basins and becomes the most affected by dust deposition events, followed by the Eastern and Western Mediterranean sub-basins [*Pey et al.*, 2013; *Varga et al.*, 2014]. The main period of the Saharan dust transport, from the North Africa to the Mediterranean Sea, goes from early spring to the end of summer. During this period the air masses cross the basin from east to west coinciding with the presence of thermal convective forces that inject the dust particles high into the atmosphere [*Alpert et al.*, 2004; *Moulin et al.*, 1998]. On the contrary, dust deposition events occur mainly in autumn and winter (Table 1) showing an evident gradient decreasing from south to north in the Mediterranean Sea [*Gallisai et al.*, 2014; *Guerzoni et al.*, 1997]. During the period 2000-2007, the Western Mediterranean was the area least affected by dust deposition in terms of VLDE, as well as in deposition intensity in terms of $kg \cdot m^{-2}$ of dust deposited (Table 1). There, the minimum mass of dust deposited on surface waters was recorded during one VLDE event occurring in spring. The mean amount of dust deposited in the Western Mediterranean Sea is half the amount deposited in the other sub-basins. On the other hand, the relatively intense dust deposition event of 27/12/2000 in the Western Mediterranean produced the largest response of chlorophyll.

The mean deposition and number of episodes were quite similar in the Central and in the Eastern Mediterranean. Since dust may travel at high altitude with little deposition, transport and deposition do not always match. This is a non-trivial issue that points on the importance of data on actual deposition (which is different than the transport inferred from satellite data) in order to study its relationship with the marine ecosystem.

The response of phytoplankton to dust addition in surface Mediterranean Sea waters showed a complex scenario. When all VLDE are taken into account, there are significant chlorophyll peaks after the deposition events, with respect to chlorophyll prior to the events. However, chlorophyll peaks after the events are not sustained over time and recover quickly to levels prior to the events, meaning that the average relative responses to dust addition are rather small as can be seen from the mean differences (Table 2). The main chlorophyll distribution feature of the Mediterranean Sea shows a basin scale gradient from west to east, with a more productive but still oligotrophic Western side and an extremely oligotrophic or ultra-oligotrophic Eastern sub-basin [*D'Ortenzio and Ribera D'Alcalà*, 2009]. A priori one would expect larger responses be observed the larger the amount of deposited dust and the lower levels of nutrients present in resident water, which is in the Eastern Mediterranean and during late spring and summer. We did not observe these expected trends. When geographical (LON) and seasonal (SEASON) variables are assessed, significance surfaces out for SEASON in ΔPeak, but the trend is reversed when ΔPeak is computed using seasonally-detrended chlorophyll (Table 4). However, both variables are not far from significance. That is, we found overall smaller responses with increasing Eastern longitude and larger responses in autumn-winter than in spring.

Reasons for such discrepancy could be related to a rather low number of VLDE. For instance, with the criteria used to define VLDE, we had no events during summer. This season, with the lowest nutrient concentrations in the water is a target for production effects from dust deposition. With a larger data set it is possible that these trends could emerge out of the model error. Part of the error must be related to the heterogeneity of events such as area coverage, intensity and time of the year, combined with the probable heterogeneity of the receiving waters in terms of the amount of nutrients already present and the composition and physiological state of the microbial communities.

Moreover, there are a number of reasons related to the physics, biogeochemistry and biology along the Mediterranean that may conform to the results observed in this study. First, the mixed layer depth increases from west to east [*D'Ortenzio et al.*, 2005]. This implies that for the same amount of nutrients deposited on surface waters, the final concentration in the water column will be higher in the Western basin and lower in the Eastern. Consistent with the evidences of trends in plankton community composition and mixed layer depth, moving from west to east, the maximum phytoplankton stimulation to dust addition we observed shifted from 345% to 121% (Table 1).

Second, there are reasons related to the plankton community composition. Dinoflagellates dominate the relative abundance of phytoplankton in the Eastern basin, while diatoms dominate the Western side [*Ignatiades et al.*, 2009]. In general, depending on nutrient availability, the different phytoplankton species adopt different competition strategies for resources. The small phytoplankton cells are believed to have a competitive advantage for nutrients in nutrient-poor waters [*Chisholm*, 1992] for their larger surface area to volume ratios. Furthermore, nutrient forms also affect the phytoplankton communities: ammonium is abundant in oligotrophic seas due to remineralization processes, and it favors small cells [*Dortch*, 1990; *Romero et al.*, 2012; *Stolte et al.*, 1994]. On the contrary, the larger cells are more advantaged in nutrient-rich seawater because they have the ability to rapidly take up and temporarily store the "new" nutrients [*Falkowski et al.*, 1998]. Therefore, they have higher photosynthetic efficiency and growth rates in nutrient-rich seawater [*Marañón et al.*, 2007]. Larger responses to dust in the Western Mediterranean when the timing of the chlorophyll peak is not different between geographic areas, means that chlorophyll has increased faster, which tends to conform to a larger dominance of rapidly-responding diatoms in the Western Mediterranean, where background nutrient concentrations are higher and algae may respond rapidly. Furthermore, regarding bacteria, the Western and the Eastern Mediterranean show comparable amounts of depth-integrated phytoplankton and bacteria but in contrast the primary production is 2 or 3 times higher in the west [*Turley et al.*, 2000]. In the Western Mediterranean the competition between bacteria and phytoplankton for resources is not as strong as in the Eastern [*Allen et al.*, 2002]. Nutrient starvation is a notch less pronounced and this fact could allow phytoplankton to have a chance against bacteria to take advantage of the nutrients added in the water column. Thus, when new nutrients arrive to the surface waters, they rapidly stimulate phytoplankton cells, which are ready to respond and this could explain why the highest peak in terms of mg·m^{-3} was recorded in the Western side. When nutrients were added to the ultra-oligotrophic waters of the eastern basin, these were transferred through the food web mostly bypassing the phytoplankton compartment [*Cermeño et al.*, 2005; *Marañón et al.*, 2007]. It seems then that primarily bacteria must have responded to dust addition in ultra-oligotrophic environments [*Krom et al.*, 2016], while phytoplankton stimulation is more efficient in the Western Mediterranean [*Turley et al.*, 2000]. The differential response to dust addition has been shown to depend on the degree of ecosystem oligotrophy before [*Marañón et al.*, 2010]. Heterotrophic bacteria in the ocean have been shown to respond to dust [*Pulido-Villena et al.*, 2008]. Also, in some experiments where both bacteria and primary producers were analyzed, bacteria tended to show primary responses [*Guieu et al.*, 2014; *Herut et al.*, 2005; *Romero et al.*, 2011]. Thus, bacteria and other microheterotrophs have a dominant role in the food web of the Eastern Mediterranean [*Durrieu de Madron et al.*, 2011] and tend to outcompete phytoplankton for nutrients, resulting in smaller phytoplankton responses even when dust deposition, and hence nutrient additions, are larger.

A third set of reasons is related to the dissolved organic matter that bacteria need to grow. The dissolved organic carbon (DOC) concentration in the surface waters has been shown to control both growth and biomass of bacteria [*Thingstad et al.*, 1997]. DOC may accumulate in the surface waters during the productive season and stratification period, and the carbon export process to deep waters only occurs when the total water column is mixed [*Santinelli et al.*, 2013; *Thingstad et al.*, 1997]. In this case the bacterial carbon consumption is restricted due to the nutrient limitation [*Trabelsi and Rassoulzadegan*, 2011] and the biomass rate of bacteria is kept low by the competition for nutrients and by bacterial predators. Thus, at times when dissolved carbon is accumulated in surface waters owing to seasonal production, inorganic nutrients are mostly depleted and water column mixing has not occurred yet, any nutrient stimulation will likely be used and channeled by bacteria. This occurs mostly in summer. In some cases the nutrient availability occurs by dust deposition [*Ternon et al.*, 2010]. Hence, even if we had VLDE in summer it is unlikely that we could detect large chlorophyll peak responses, as bacteria likely respond very fast. This does not mean the system is not responding to the added nutrients but that phytoplankton is not the primary beneficiary.

Recent studies also seem to point to an increasing eastward gradient in surface DOC [*Pujo-Pay et al.*, 2011], which again would favor a trend of increased bacterial responses over phytoplankton responses to nutrient additions.

Overall, we cannot pinpoint a single mechanistic explanation for the observed 70% increase in chlorophyll after VLDE. Chlorophyll response differences between seasons are not clear, as they tend to disappear in seasonally detrended data. Even so, a pattern emerges with an eastward decreasing gradient of response intensity that conforms to other biogeochemical gradients and the relative importance of phytoplankton and heterotrophic bacteria.

In this study we have analyzed the large deposition events. When the whole deposition time series was analyzed in a previous study [*Gallisai et al.*, 2014], 64% of the Mediterranean surface showed significant positive correlation values between chlorophyll concentration and desert dust deposition, mainly in the Central and Eastern Mediterranean. Positive correlation values appeared during all seasons, but the strongest correlations were found in spring. The Central and Eastern Mediterranean seem to share more dynamics than either with the Western Mediterranean, likely because the sill at the Strait of Sicily is the largest water separation feature within the Mediterranean and that the Central and Eastern Mediterranean share similar latitudes. Comparing the results of *Gallisai et al.* [2014] and this study, the impact of large dust deposition events on chlorophyll is complementary both in space and time to a more continuous basal dust input, where the Central Mediterranean is strongly impacted by both dynamics. It appears that Saharan dust has a direct impact on chlorophyll dynamics, indicating an alleviation of nutrient limitation by atmospheric deposition. This has important implications for the nutrient budget of the basin over decadal to geological time scales while at ecological time scales the relative input of atmospheric nutrients is small for a given time and area and the detection of the impact on chlorophyll production will depend on a complex matrix of factors. Although we do see clear chlorophyll peak responses to VLDE, the effects of such inputs are difficult to detect and explain for individual events.

Mahowald et al. [2010] estimated the doubling of desert dust during the 20^{th} century over a large part of the Earth, as well as, a 6% increase in ocean productivity due to the increase in dust deposition trends.

Moreover, future scenarios foresee increases in the presence of Saharan dust particles into the atmosphere [*Ganor et al.*, 2010; *Mahowald et al.*, 2010; *Querol et al.*, 2009] and the climate changes are also foreseen to result in a shallowing of the mixed layer. The unique characteristics of the Mediterranean Sea as a quasi-enclosed basin surrounded by mineral dust and other aerosol production sources, make it a prime target for aerosol-derived nutrient deposition to drive both long-term element budgets as well as to drive ecological alterations, including the response of primary producers and the recycling of organic matter by bacteria, especially in the Eastern Mediterranean. Under global change scenarios, phytoplankton is expected to benefit mostly in the Western Mediterranean, while heterotrophic bacteria will likely increasingly respond towards the Eastern Mediterranean.

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All data using in the present paper are held in a public repository. All Chlorophyll concentration data are available from the CNR database http://www.myocean.eu/web/69-myocean-interactive-catalogue.php. All Dust deposition modelled data are available from the BSC database http://www.bsc.es/earth-sciences/mineral-dust/catalogo-datos-dust.

References

Allen, J. I., P. J. Somerfield, and J. Siddorn (2002), Primary and bacterial production in the Mediterranean Sea: a modelling study, *J. Mar. Syst.*, *33–34*, 473-495, doi:10.1016/S0924-7963(02)00072-6.

Alonso-Perez, S., E. Cuevas, C. Perez, X. Querol, J. M. Baldasano, R. Draxler, and J. J. De Bustos (2011), Trend changes of African airmass intrusions in the marine boundary layer over the subtropical Eastern North Atlantic region in winter, *Tellus (B Chem. Phys. Meteorol.)*, *63*(2), 255-265, doi:10.1111/j.1600-0889.2010.00524.x.

Alpert, P., and B. Ziv (1989), The Sharav Cyclone: Observations and some theoretical considerations, *94*(D15), 18495–18514, doi:10.1029/JD094iD15p18495.

Alpert, P., P. Kishcha, A. Shtivelman, S. O. Krichak, and J. H. Joseph (2004), Vertical distribution of Saharan dust based on 2.5-year model predictions, *Atmospheric Research*, *70*(2), 109-130, doi:10.1016/j.atmosres.2003.11.001.

Amiridis, V., et al. (2009), The potential of the synergistic use of passive and active remote sensing measurements for the validation of a regional dust model, *Ann. Geophys.*, *27*(8), 3155-3164, doi:10.5194/angeo-27-3155-2009.

Aschmann, H. (1973), Distribution and Peculiarity of Mediterranean Ecosystems, in *Mediterranean Type Ecosystems*, edited by F. di Castri and H. A. Mooney, pp. 11-19, Springer Berlin Heidelberg.

Basart, S., C. Perez, S. Nickovic, E. Cuevas, and J. M. Baldasano (2012), Development and evaluation of the BSC-DREAM8b dust regional model over Northern Africa, the Mediterranean and the Middle East, *Tellus (B Chem. Phys. Meteorol.)*, *64*, 18539, doi:10.3402/tellusb.v64i0.18539.

Bonnet, S., C. Guieu, J. Chiaverini, J. Ras, and A. Stock (2005), Effect of atmospheric nutrients on the autotrophic communities in a low nutrient, low chlorophyll system, *Limnol. Oceanogr.*, *50*(6), 1810-1819, doi:10.4319/lo.2005.50.6.1810.

Cermeño, P., E. Marañón, J. Rodriguez, and E. Fernandez (2005), Large-sized phytoplankton sustain higher carbonspecific photosynthesis than smaller cells in a coastal eutrophic ecosystem, *Mar. Ecol. Prog. Ser.*, *297*, 51-60, doi:10.3354/meps297051.

Chisholm, S. W. (1992), Phytoplankton size, in *Primary productivity and biogeochemical cycles in the sea.*, edited by P. G. Falkowski and A. D. Woodhead, pp. 213-237, Plenum, New York.

D'Almeida, G. A. (1987), Desert aerosol characteristics and effects on climate, in *Paleoclimatology and Paleometeorology : Modern and Past Patterns of Global Atmospheric Transport*, edited by M. Leinen and M. Sarnthein, pp. 311-338, Kluwer Academic Publ, Dordrecht.

D'Ortenzio, F., and M. Ribera D'Alcalà (2009), On the trophic regimes of the Mediterranean Sea: A satellite analysis, *Biogeosciences*, *6*(2), 139-148, doi:10.5194/bg-6-139-2009.

D'Ortenzio, F., D. Ludicone, C. de Boyer Montegut, P. Testor, D. Antoine, S. Marullo, R. Santoleri, and G. Madec (2005), Seasonal variability of the mixed layer depth in the Mediterranean Sea as derived from in situ profiles, *Geophys. Res. Lett.*, *32*(12), doi:10.1029/2005gl022463.

Dortch, Q. (1990), The interaction between ammonium and nitrate uptake in phytoplankton *Mar. Ecol. Prog. Ser.*, *61*(1-2), 183-201, doi:10.3354/meps061183.

Durrieu de Madron, X., et al. (2011), Marine ecosystems' responses to climatic and anthropogenic forcings in the Mediterranean, *Prog. Oceanogr.*, *91*(2), 97-166, doi:10.1016/j.pocean.2011.02.003.

Eker-Develi, E., A. E. Kideys, and S. Tugrul (2006), Role of Saharan dust on phytoplankton dynamics in the northeastern Mediterranean, *Mar. Ecol. Prog. Ser.*, *314*, 61-75, doi:10.3354/meps314061.

Falkowski, P. G., R. T. Barber, and V. Smetacek (1998), Biogeochemical controls and feedbacks on ocean primary production, *Science*, *281*(5374), 200-206, doi:10.1126/science.281.5374.200.

Gallisai, R., F. Peters, S. Basart, and J. M. Baldasano (2012), Mediterranean basin-wide correlations between Saharan dust deposition and ocean chlorophyll concentration, *Biogeosci. Disc.*, *9*(7), 28, doi:10.5194/bgd-9-8611-2012.

Gallisai, R., F. Peters, G. Volpe, S. Basart, and J. M. Baldasano (2014), Saharan Dust Deposition May Affect Phytoplankton Growth in the Mediterranean Sea at Ecological Time Scales, *PLoS ONE*, *9*(10), 10, doi: 10.1371/journal.pone.0110762.

Ganor, E., I. Osetinsky, A. Stupp, and P. Alpert (2010), Increasing trend of African dust, over 49 years, in the eastern Mediterranean, *J. Geophys. Res. (Atmos.)*, *115*, D07201, doi:10.1029/2009jd012500.

Ginoux, P., J. M. Prospero, T. E. Gill, N. C. Hsu, and M. Zhao (2012), Global-scale attribution of anthropogenic and natural dust sources and their emission rates based on MODIS Deep Blue aerosol products *Rev. Geophys.*, *50*, doi:10.1029/2012rg000388.

Guerzoni, S., E. Molinaroli, and R. Chester (1997), Saharan dust inputs to the western Mediterranean Sea: Depositional patterns, geochemistry and sedimentological implications, *Deep-Sea Res. Part II.*, *44*(3-4), 631-654, doi:10.1016/S0967-0645(96)00096-3.

Guieu, C., C. Ridame, E. Pulido-Villena, M. Bressac, K. Desboeufs, and F. Dulac (2014), Impact of dust deposition on carbon budget: a tentative assessment from a mesocosm approach, *Biogeosciences*, *11*(19), 5621-5635, doi:10.5194/bg-11-5621-2014.

Herut, B., T. Zohary, M. D. Krom, R. F. C. Mantoura, P. Pitta, S. Psarra, F. Rassoulzadegan, T. Tanaka, and T. F. Thingstad (2005), Response of East Mediterranean surface water to Saharan dust: On-board microcosm experiment and field observations, *Deep-Sea Res. Part II.*, *52*(22-23), 3024-3040, doi:10.1016/j.dsr.2005.09.003.

Ignatiades, L., O. Gotsis-Skretas, K. Pagou, and E. Krasakopoulou (2009), Diversification of phytoplankton community structure and related parameters along a large-scale longitudinal east-west transect of the Mediterranean Sea, *J. Plankton Res.*, *31*(4), 411-428, doi:10.1093/plankt/fbn124.

Krom, M., et al. (2016), Response of the Eastern Mediterranean microbial ecosystem to dust and dust affected by acid processing in the atmosphere, *Frontiers in Marine Science*, *3*, doi: 10.3389/fmars.2016.00133.

Laghdass, M., S. Blain, M. Besseling, P. Catala, C. Guieu, and I. Obernosterer (2011), Effects of Saharan dust on the microbial community during a large in situ mesocosm experiment in the NW Mediterranean Sea, *Aquat. Microb. Ecol.*, *62*(2), 201-213, doi:10.3354/ame01466.

Longhurst, A. (1995), Seasonal cycles of pelagic production and consumption, *Prog. Oceanogr.*, *36*(2), 77-167, doi:10.1016/0079-6611(95)00015-1.

Ludwig, W., A. F. Bouwman, E. Dumont, and F. Lespinas (2010), Water and nutrient fluxes from major Mediterranean and Black Sea rivers: Past and future trends and their implications for the basin-scale budgets, *Global. Biogeochem. Cy.*, *24*(4), GB0A13 doi:10.1029/2009gb003594.

Mahowald, N. M., et al. (2010), Observed 20th century desert dust variability: impact on climate and biogeochemistry, *Atmos. Chem. Phys.*, *10*(22), 10875-10893, doi:10.5194/acp-10-10875-2010.

Marañón, E., P. Cermeño, J. Rodriguez, M. V. Zubkov, and R. P. Harris (2007), Scaling of phytoplankton photosynthesis and cell size in the ocean, *Limnol. Oceanogr.*, *52*(5), 2190-2198, doi:10.4319/lo.2007.52.5.2190.

Marañón, E., et al. (2010), Degree of oligotrophy controls the response of microbial plankton to Saharan dust, *Limnol. Oceanogr.*, *55*(6), 2339-2352, doi:10.4319/lo.2010.55.6.2339.

Moulin, C., H. R. Gordon, V. F. Banzon, and R. H. Evans (2001), Assessment of Saharan dust absorption in the visible from SeaWiFS imagery, *J. Geophys. Res. (Atmos.)*, *106*(D16), 18239-18249, doi:10.1029/2000jd900812.

Moulin, C., et al. (1998), Satellite climatology of African dust transport in the Mediterranean atmosphere, *J. Geophys. Res. (Atmos.)*, *103*(D11), 13137-13144, doi:10.1029/98JD00171.

Nickovic, S., G. Kallos, A. Papadopoulos, and O. Kakaliagou (2001), A model for prediction of desert dust cycle in the atmosphere, *J. Geophys. Res. (Atmos.)*, *106*(D16), 18113-18129, doi:10.1029/2000jd900794.

Paerl, H. W. (1997), Coastal eutrophication and harmful algal blooms: Importance of atmospheric deposition and groundwater as ‘’new’’ nitrogen and other nutrient sources, *Limnol. Oceanogr.*, *42*(5), 1154-1165, doi:10.4319/lo.1997.42.5_part_2.1154.

Papanastasiou, D. K., A. Poupkou, E. Katragkou, V. Amiridis, D. Melas, N. Mihalopoulos, S. Basart, C. Pérez, and J. M. Baldasano (2010), An Assessment of the Efficiency of Dust Regional Modelling to Predict Saharan Dust Transport Episodes, *Adv. Meteo.*, *2010*, 154368 doi:10.1155/2010/154368.

Pay, M. T., P. Jiménez-Guerrero, O. Jorba, S. Basart, X. Querol, M. Pandolfi, and J. M. Baldasano (2012), Spatio-temporal variability of concentrations and speciation of particulate matter across Spain in the CALIOPE modeling system, *Atmos. Environ.*, *46*, 376-396, doi:10.1016/j.atmosenv.2011.09.049.

Pérez, C., S. Nickovic, G. Pejanovic, J. M. Baldasano, and E. Özsoy (2006a), Interactive dust-radiation modeling: A step to improve weather forecasts, *J. Geophys. Res. (Atmos.)*, *111*(D16), D16206, doi:10.1029/2005JD006717.

Pérez, C., S. Nickovic, J. M. Baldasano, M. Sicard, F. Rocadenbosch, and V. E. Cachorro (2006b), A long Saharan dust event over the western Mediterranean: Lidar, Sun photometer observations, and regional dust modeling, *J. Geophys. Res. (Atmos.)*, *111*(D15), D15214, doi:10.1029/2005JD006579.

Pey, J., X. Querol, A. Alastuey, F. Forastiere, and M. Stafoggia (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*(3), 1395-1410, doi:10.5194/acp-13-1395-2013.

Pujo-Pay, M., P. Conan, L. Oriol, V. Cornet-Barthaux, C. Falco, J. F. Ghiglione, C. Goyet, T. Moutin, and L. Prieur (2011), Integrated survey of elemental stoichiometry (C, N, P) from the western to eastern Mediterranean Sea, *Biogeosciences*, *8*(4), 883-899, doi:10.5194/bg-8-883-2011.

Pulido-Villena, E., T. Wagener, and C. Guieu (2008), Bacterial response to dust pulses in the western Mediterranean: Implications for carbon cycling in the oligotropic ocean, *Global. Biogeochem. Cy.*, *22*(1), GB1020, doi:10.1029/2007GB003091.

Querol, X., A. Alastuey, J. Pey, M. Cusack, N. Perez, N. Mihalopoulos, C. Theodosi, E. Gerasopoulos, N. Kubilay, and M. Kocak (2009), Variability in regional background aerosols within the Mediterranean, *Atmos. Chem. Phys.*, *9*(14), 4575-4591, doi:10.5194/acp-9-4575-2009.

Ridame, C., J. Dekaezemacker, C. Guieu, S. Bonnet, S. L'Helguen, and F. Malien (2014), Contrasted Saharan dust events in LNLC environments: impact on nutrient dynamics and primary production, *Biogeosciences*, *11*(17), 4783-4800, doi: 10.5194/bg-11-4783-2014.

Romero, E., F. Peters, C. Marrasé, T. Guadayol, J. M. Gasol, and M. G. Weinbauer (2011), Coastal Mediterranean plankton stimulation dynamics through a dust storm event: An experimental simulation, *Estuar. Coast. Shelf Sci.*, *93*(1), 27-39, doi:10.1016/j.ecss.2011.03.019.

Romero, E., J. Garnier, L. Lassaletta, G. Billen, R. Gendre, P. Riou, and P. Cugier (2012), Large-scale patterns of river inputs in southwestern Europe: seasonal and interannual variations and potential eutrophication effects at the coastal zone, *Biogeochemistry*, 1-25, doi:10.1007/s10533-012-9778-0.

Santinelli, C., D. A. Hansell, and M. Ribera D'Alcalá (2013), Influence of stratification on marine dissolved organic carbon (DOC) dynamics: The Mediterranean Sea case, *Prog. Oceanogr.*, *119*, 68-77, doi:10.1016/j.pocean.2013.06.001.

Stolte, W., T. McCollin, A. A. M. Noordeloos, and R. Riegman (1994), Effect of nitroge-source on the size distribution within marine phytoplankton populations, *J. Exp. Mar. Biol. Ecol.*, *184*(1), 83-97, doi:10.1016/0022-0981(94)90167-8.

Swap, R., S. Ulanski, M. Cobbett, and M. Garstang (1996), Temporal and spatial characteristics of Saharan dust outbreaks, *Journal of Geophysical Research*, *101*(D2), 4205, doi:10.1029/95jd03236.

Ternon, E., C. Guieu, C. Ridame, S. L'Helguen, and P. Catala (2011), Longitudinal variability of the biogeochemical role of Mediterranean aerosols in the Mediterranean Sea, *Biogeosciences*, *8*(5), 1067-1080, doi:10.5194/bg-8-1067-2011.

Ternon, E., C. Guieu, M. D. Loye-Pilot, N. Leblond, E. Bosc, B. Gasser, J. C. Miquel, and J. Martin (2010), The impact of Saharan dust on the particulate export in the water column of the North Western Mediterranean Sea, *Biogeosciences*, *7*(3), 809-826, doi:10.5194/bg-7-809-2010.

Thingstad, T. F., A. Hagstrom, and F. Rassoulzadegan (1997), Accumulation of degradable DOC in surface waters: Is it caused by a malfunctioning microbial loop?, *Limnol. Oceanogr.*, *42*(2), 398-404, doi:10.4319/lo.1997.42.2.0398.

Trabelsi, A., and F. Rassoulzadegan (2011), Inorganic nutrient control of dissolved organic carbon (DOC) dynamics in NW Mediterranean waters: An experimental approach, *Mar. Biol. Res.*, *7*(7), 667-676, doi:10.1080/17451000.2011.554557.

Turley, C. M., M. Bianchi, U. Christaki, P. Conan, J. R. W. Harris, S. Psarra, G. Ruddy, E. D. Stutt, A. Tselepides, and F. Van Wambeke (2000), Relationship between primary producers and bacteria in an oligotrophic sea - the Mediterranean and biogeochemical implications, *Mar. Ecol. Prog. Ser.*, *193*, 11-18, doi:10.3354/meps193011.

UNEP (1989), State of the Mediterranean marine environment, edited, pp. 1-62, UNEP, Athens.

UNEP/FAO/WHO (1996), Assessment of the state of eutrophication in the Mediterranean Sea, edited, p. MAP technical report series nº106, UNEP, Athens.

Varga, G., G. Ujvari, and J. Kovacs (2014), Spatiotemporal patterns of Saharan dust outbreaks in the Mediterranean Basin, *Aeolian Research*, *15*, 151-160, doi:10.1016/j.aeolia.2014.06.005.

Volpe, G., B. B. Nardelli, P. Cipollini, R. Santoleri, and I. S. Robinson (2012), Seasonal to interannual phytoplankton response to physical processes in the Mediterranean Sea from satellite observations, *Remote Sens. Environ.*, *117*, 223-235, doi:10.1016/j.rse.2011.09.020.

Volpe, G., R. Santoleri, V. Vellucci, M. R. d'Alcala, S. Marullo, and F. D'Ortenzio (2007), The colour of the Mediterranean Sea: Global versus regional bio-optical algorithms evaluation and implication for satellite chlorophyll estimates, *Remote Sens. Environ.*, *107*(4), 625-638, doi:10.1016/j.rse.2006.10.017.

Volpe, G., V. F. Banzon, R. H. Evans, R. Santoleri, A. J. Mariano, and R. Sciarra (2009), Satellite observations of the impact of dust in a low-nutrient, low-chlorophyll region: Fertilization or artifact?, *Global. Biogeochem. Cy.*, *23*(3), GB3007, doi:10.1029/2008gb003216.

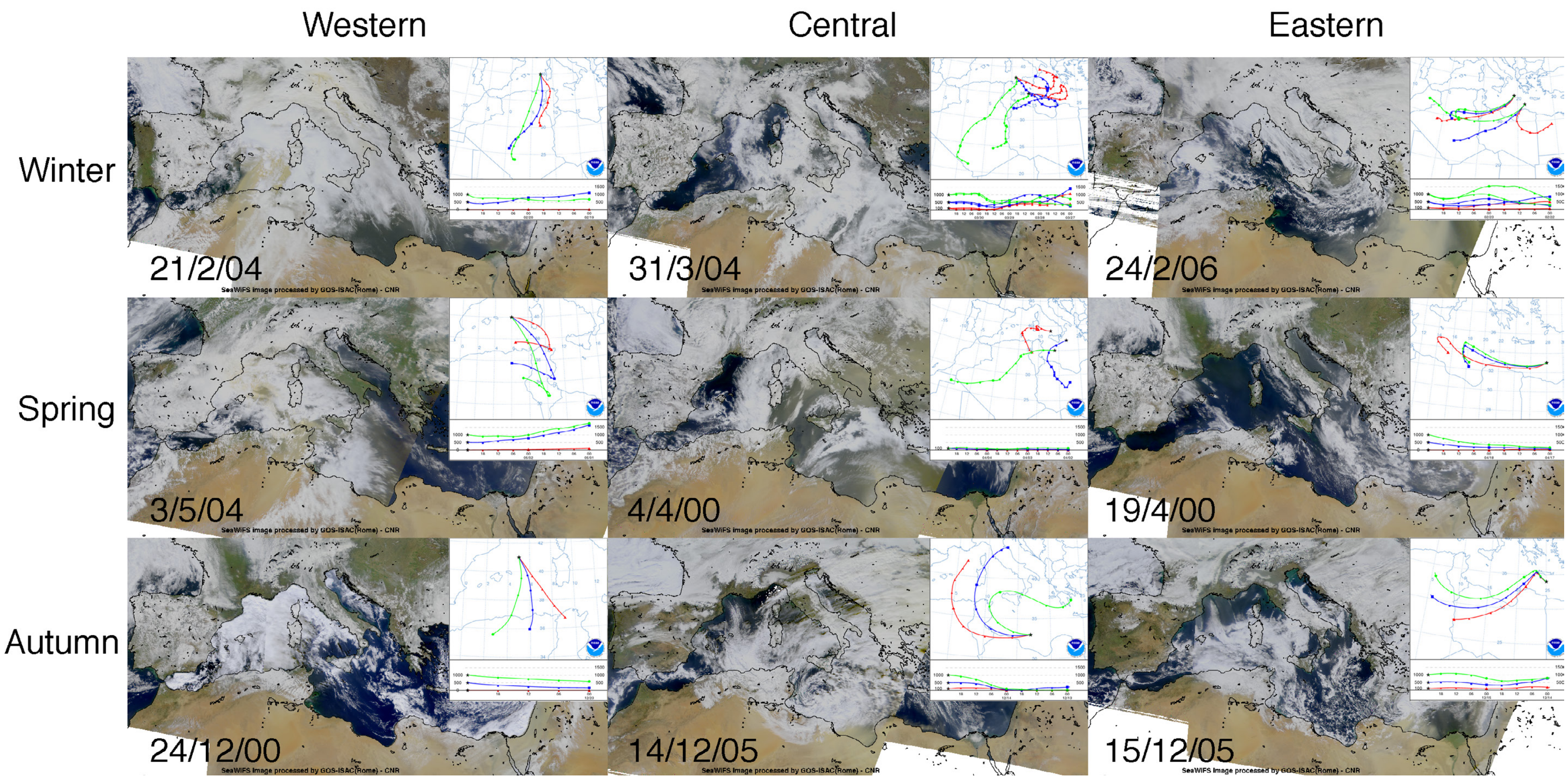

Figure S1. True-color satellite images for some VLDE representative of each season and geographical area. In the inset we show wind back-trajectories from the HYSPLIT model.

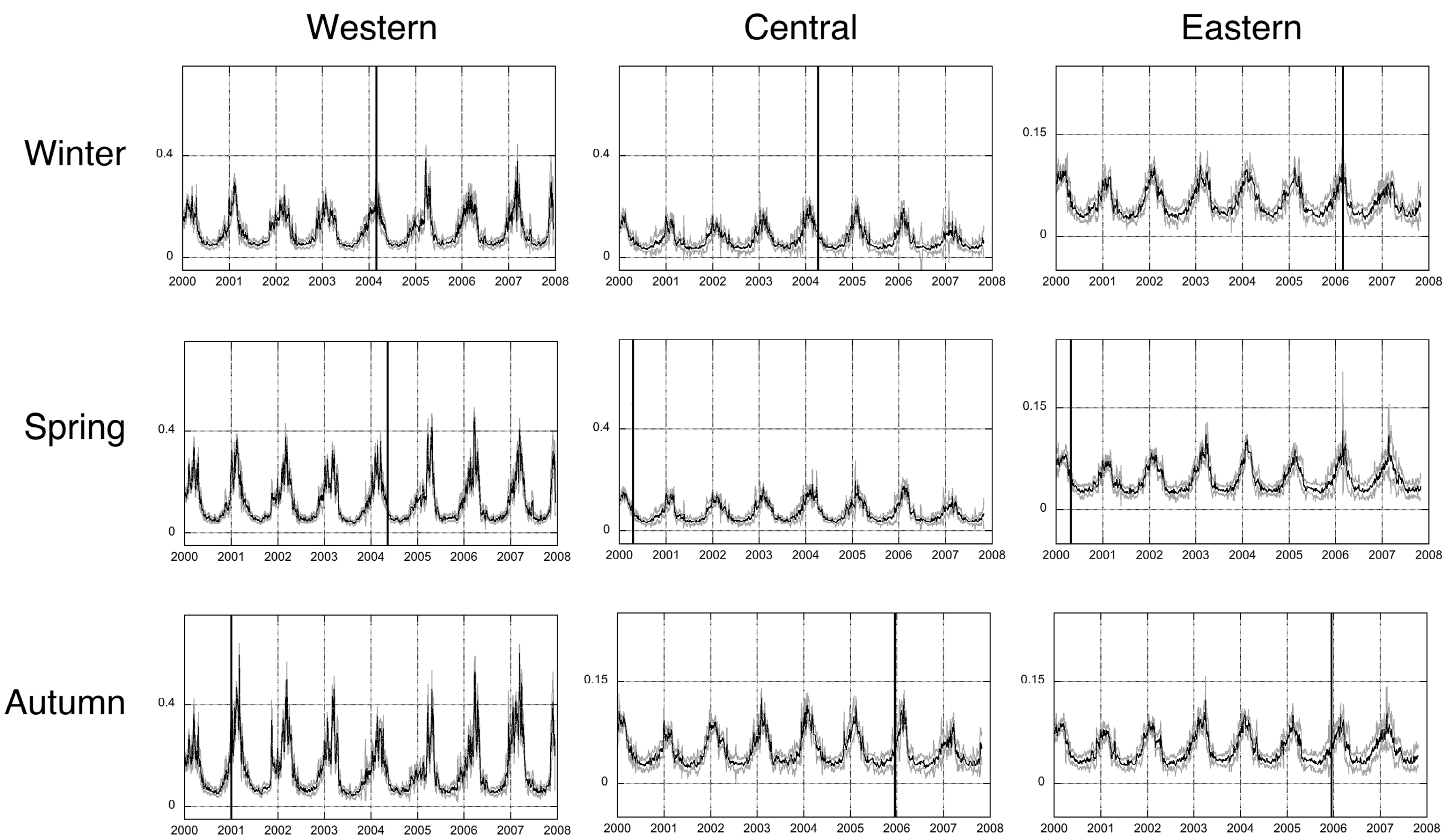

Figure S2. Chlorophyll dynamics (average in black and ±standard error in grey) for the cell-grids affected by some VLDE, representative of each season and geographical area. The strong vertical black line indicates the date of the VLDE.

Conclusions

1- The seasonal dynamics of modeled dust deposition from 2000 to 2007 shows highest values in late autumn and winter in the Central and Eastern Mediterranean, albeit with a lower overall annual variability than in the Northern-Northwestern Mediterranean. The spatial distribution shows high deposition close to Africa with a decreasing gradient from South to North of the basin. By contrast, the ocean surface chlorophyll distribution shows decreasing gradients both from West to East and from North to South. In addition, the broadest variability in surface chlorophyll was found in the Western Mediterranean, coinciding with the highest chlorophyll concentration on average.

2- Modeled dust deposition correlates with satellite chlorophyll concentration in large areas of the Mediterranean Sea, with a clear South to North decreasing gradient in correlation coefficient. This is especially true for the Central ($r=0.65$) and the Eastern ($r=0.3$) Mediterranean, where dust deposition dynamics matches with chlorophyll annual dynamics. When seasonally detrended values of both variables were used, the correlation coefficients decreased somewhat. The areas with significant correlations were located mainly in the Central and Southwestern Mediterranean with correlation values between 0.3 and 0.4.

3- Positive correlations between deposition and surface chlorophyll can be found during all seasons, although it is in spring (AMJ) when we see the largest correlation coefficients mainly in the Central, Eastern and Southwestern Mediterranean. The Western and Central Mediterranean also show regions with positive correlations in summer (JAS), while in autumn (OND) there are some positive correlations in the Central and Eastern Mediterranean. Furthermore, small areas of significant negative correlations are observed in the Alboran Sea, in the eastern coast of Spain and in the Aegean Sea.

4- Large dust deposition events (above average deposition load in 5 or more 1°X1° areas) were identified during the period between 2000 and 2007. Many of the 153 events occurred in years 2000 and 2004, especially in autumn and winter. In general, the occurrence of large dust deposition events showed a high variability. Depending on the year and season they affected different parts of the Mediterranean Sea. In terms of the extension of the affected surface, the most affected region was the Eastern Mediterranean in winter, followed by the Central Mediterranean in autumn and the Western Mediterranean in spring. Among all large dust deposition events, 31 were considered very large dust deposition events (affecting 20 or more 1°X1° areas). They were distributed unequally over seasons: 12 in the winter, 5 in spring and 14 in autumn. Geographically, 7 very large dust deposition events affected the Western, 13 the Central and 11 the Eastern Mediterranean.

5- Surface chlorophyll peaked significantly after most of the very large dust deposition events. Chlorophyll concentration peaks were observed between days 1 and 6 after the deposition event, with chlorophyll increments ranging from 13% to 345% for the different outbreaks.

6- The amount of dust deposited ($kg \cdot m^{-2}$) onto surface waters was larger in the Eastern than in the Western Mediterranean. The response of chlorophyll to very large dust deposition events was not statistically related to the magnitude of the dust outbreak, either in deposited dust per unit area or in the extension of the surface affected. It was also statistically unrelated to the chlorophyll before the VLDE. Chlorophyll responses were related to the time of the year (SEASON) with higher responses in autumn than in spring. Using seasonally detrended data, the importance of seasonality decreases in favor of a longitudinal (LON) gradient of decreasing response intensity from west to east. We relate this gradient to the relative importance of heterotrophic bacteria to phytoplankton, which increases eastward.

7- In an ocean heavily marked by seasonality where the annual cycle explains an average of 77% of the variability in the chlorophyll signal, deposition may explain another 4% in certain areas.