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Key Points:
- Temporal correlation of aerosol deposition fluxes at three sites in southern Spain separates local from Saharan sources
- Saharan dust increased mass deposition 85% above deposition background levels
- Saharan dust adds 0.07 kg ha^{-1} yr^{-1} of P to ecosystems in this region

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Saharan versus local influence on atmospheric aerosol deposition in the southern Iberian Peninsula: Significance for N and P inputs

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Abstract A novel methodology was used to evaluate the contribution of Saharan dust to the atmospheric deposition of particulate material (PM), total phosphorus (TP), and total nitrogen (TN) in the southeastern Iberian Peninsula. Dry and wet aerosol depositions were measured weekly during two 1 year periods at one site and simultaneously during spring-summer of the same years at two other sites (intersite distance of ~ 40 km). Statistical relationships among depositions at the different sites permitted differentiation of Saharan dust inputs from locally derived dust. PM and TP depositions were synchronous among the three study sites; the synchrony was elevated during periods of Saharan intrusions (evaluated by air mass retrotrajectories analyses), but no temporal correlation was observed during periods without Saharan intrusions. According to analysis of variance results, PM and TP depositions were both significantly affected by Saharan intrusions. During weeks with Saharan intrusions, PM deposition increased around 85% above background levels, with no differences among the three sites, while TP deposition increased by 1.1 μmol TP m^{-2} d^{-1}, i.e., 29% to 81% above background levels depending on the site. There were no correlations or differences in TN deposition among sites or as a function of Saharan intrusion periods. The annual contribution of PM and TP from Saharan dust was 75 kg ha^{-1} and 0.07 kg P ha^{-1}, respectively, which can be considered a genuine input for the ecosystems in this area. This novel approach is likely to be valid in any area in the world under atmospheric deposition of long-range transported material.

1. Introduction

The atmosphere is the main global natural source of nitrogen (N) for ecosystems, through the biological fixation or deposition of reactive nitrogen, even though rock-derived nitrogen (N) can supply biologically available N to ecosystems in regions with sedimentary rocks [*Morford et al.*, 2011]. Atmospheric dynamics and chemistry therefore play a dominant role in the N cycle of ecosystems, whereas parent minerals have traditionally been considered the only significant source of phosphorus (P) [*Walker and Syers*, 1976]. The prevalent paradigm has been that P availability declines with the aging of soils through leaching, erosion, and chemical occlusion, thereby limiting the productivity of terrestrial ecosystems over time [*Walker and Syers*, 1976]. According to this view of the P cycle, external P inputs for aquatic ecosystems are restricted to run off from catchments or, in the case of marine ecosystems, to discharge from rivers.

However, the atmospheric input of P to continental ecosystems can be as high as 1.7 kg ha^{-1} yr^{-1}, although there is wide geographic variability [*Newman*, 1995]. Atmospheric deposition of P transported from a long distance was found to be sufficient to offset soil leaching and erosion losses in the Yucatan Peninsula [*Das et al.*, 2013], and the P content of soils was reported to be rejuvenated by P inputs from active dust deposition [*Eger et al.*, 2013]. Atmospheric P inputs are also known to contribute a fertilizing effect in marine [*Mills et al.*, 2004] and lake [*Morales-Baquero et al.*, 2006] ecosystems. Atmospheric P inputs can be important for Mediterranean soils, especially for degraded soils, which are often deficient in P [*Rodà et al.*, 1999], and for those whose P limitation may increase as a consequence of climate change [*Sardans and Peñuelas*, 2004, 2007]. Hence, there is increasing interest in the role of the atmosphere in the mobilization, transport, and deposition of P.

Dust aerosols are the predominant source of P in the atmosphere [*Mahowald et al.*, 2008] and also transport N compounds over long distances, adsorbed on the surface of the particles [*Dentener et al.*, 1996]. The largest and most persistent dust sources in the world are located in the Northern Hemisphere, mainly in the "dust belt" that extends from the west coast of North Africa, over the Middle East, Central, and South Asia, to China [*Prospero et al.*, 2002]. The Sahara is responsible for the largest amount of dust aerosols worldwide, contributing with $670 \cdot 10^6$ t yr^{-1} [*D'Almeida*, 1986] and accounting for around 50% of global dust production [*Schütz et al.*, 1981]. Aerosols from the Sahara are transported mainly to the Atlantic [*Carlson and Prospero*, 1972; *Swap et al.*, 1992] but also to the Mediterranean, and the amount of Saharan dust mobilized to Europe has been estimated at $80–120 \cdot 10^6$ t yr^{-1} [*D'Almeida*, 1986]. This dust export to the atmosphere has increased exponentially over the past few decades and centuries as a consequence of North Africa droughts [*Prospero and Lamb*, 2003], human-induced desertification [*Moulin and Chiapello*, 2006], and the development of commercial agriculture in the Sahel region [*Mulitza et al.*, 2010].

Much less is known about the deposition of the dust, which can be washed from the atmosphere by rain or snow (wet deposition) or gravitationally deposited (dry deposition). The quantification of dust fluxes is difficult, in particular to differentiate between locally derived or recycled nutrient inputs and aerosols transported over long distances. This is because (a) field sampling is often limited to small spatial and temporal scales and does not adequately represent the complexity and heterogeneity of deposition processes at landscape or regional scale; (b) it is difficult to separate the contribution of dust from that of other atmospheric aerosols (biogenic particles, biomass burning, industrial, or urban); and (c) the measurement of dry deposition is methodologically complex [*Wesely and Hicks*, 2000].

However, understanding of the nutrient budget of an ecosystem requires knowledge of the relative contribution of nutrient input from distant versus local sources. When the nutrient input derives from outside the ecosystem, it constitutes a genuine input from the atmosphere, whereas nutrient input from the same locality should be considered as intrasystem recycling. In comparison to other dust components, atmospheric P deposition appears much more susceptible to the influence of local sources from agricultural and industrial activities [*Kopacek et al.*, 1997], pollen emissions [*Carlisle et al.*, 1966; *Jordan et al.*, 1995], and biomass burning [*Kauffman et al.*, 1994]. It is therefore important to elucidate the sources of P in order to estimate the true inputs of this nutrient to ecosystems and their significance with respect to the rate of mineral weathering of P from parent rock mineral.

Few published data are available on the relative contribution of different sources to atmospheric P deposition. Various methodologies have been adopted, including determination of soil-specific elements in dust deposition (e.g., Al or Ti) as indicators of soil-derived P deposition [*Bergametti et al.*, 1992]; measurement of dissolved Mo and nss-$SO_4^{=}$ in deposition as indicators of soluble P deposition from fossil fuel combustion [*Tsukuda et al.*, 2005]; application of a sequential P extraction method [*Chen et al.*, 2006]; fractionation of deposition samples to estimate the deposition of long-range transported P from organic material [*Tsukuda et al.*, 2006]; the simultaneous application of three independent methods to evaluate the contribution of long-distance transported dust to P deposition [*Das et al.*, 2013]; and satellite-based multiyear estimate of dust and P deposition [*Yu et al.*, 2015].

In the Iberian Peninsula, the highest atmospheric concentrations of particles sized below 10 μm are found in the Southeast, with almost half of them deriving from Saharan dust [*Pey et al.*, 2013]. This region is less than 1500 km from the Sahara Desert, and 70% of the dust is known to be deposited within the first 2000 km [*Jaenicke and Schütz*, 1978]. Saharan dust inputs have been found to exert a significant influence on the biogeochemistry of lake ecosystems in this area [*Morales-Baquero et al.*, 2006; *Pulido-Villena et al.*, 2006, 2008; *Mladenov et al.*, 2011] and on the chemical signature of aerosol deposition [*Morales-Baquero et al.*, 2013]. However, the relative contribution of atmospheric deposition of Saharan versus local dust has not yet been established.

In the present study, a novel approach was used to differentiate between local and long-range transported deposition. Typically, long-range transported material is mobilized in the free troposphere, above the planetary boundary layer (PBL). The PBL is the layer of the atmosphere adjacent to the ground, and constituents emitted within or entering this layer become vertically dispersed by convection or mechanical turbulence [*Stull*, 1988]. The dispersion of components by the PBL is negatively related to their sedimentation velocity, with a shorter life time and transport range than in the free troposphere [*Seibert et al.*, 2000]. In the case of the Sahara, dust plumes cover thousands of square kilometers in the free troposphere, and it can be expected that particles entering the PBL are homogeneously deposited at ground level on large spatial scales. Therefore, we simultaneously measured the aerosol deposition at three sites using collectors located in the PBL and around 40 km apart, analyzing retrotrajectories of air masses to detect the common signal of long-range transported aerosols in the deposited material as simultaneous increases in the amounts collected under Saharan dust intrusions with respect to those collected without Saharan influence. Local deposition is defined

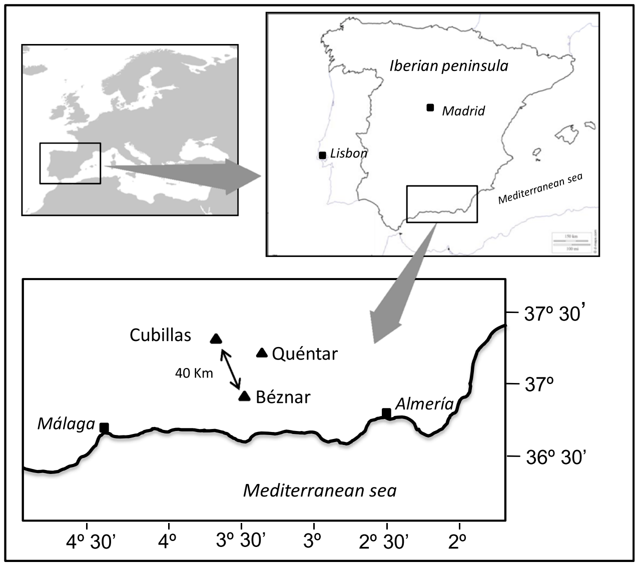

Figure 1. Location of the three sites (triangles) where atmospheric deposition samplers were installed. Mediterranean coastal cities (squares) are indicated for reference

as the dust deposition occurring during weeks without Saharan influence and is mainly supplied by ground sources around collectors and mobilized by turbulence into the PBL.

Our goals were (a) to quantify the relative contributions of Saharan dust versus local sources to atmospheric aerosol deposition in the southeastern Iberian Peninsula and (b) to study the relevance of Saharan dust to atmospheric inputs of N and P in this area.

2. Material and Methods

2.1. Sampling and Chemical Analyses

Sampling of the atmospheric deposition of particulate matter (PM), Total N (TN), and Total P (TP) was conducted at three sites: Quéntar reservoir (1000 m above sea level (asl), 37°12′N, 3°26′W), Cubillas reservoir (640 m asl, 37°16′N, 3°41′W), and Béznar reservoir (500 m asl, 36°54′N, 3°32′W). The sites are in the southeastern Iberian Peninsula at $<$ 25 km from the Mediterranean Sea (Figure 1), in a semiarid region with an annual rainfall of $\approx$ 450 mm.

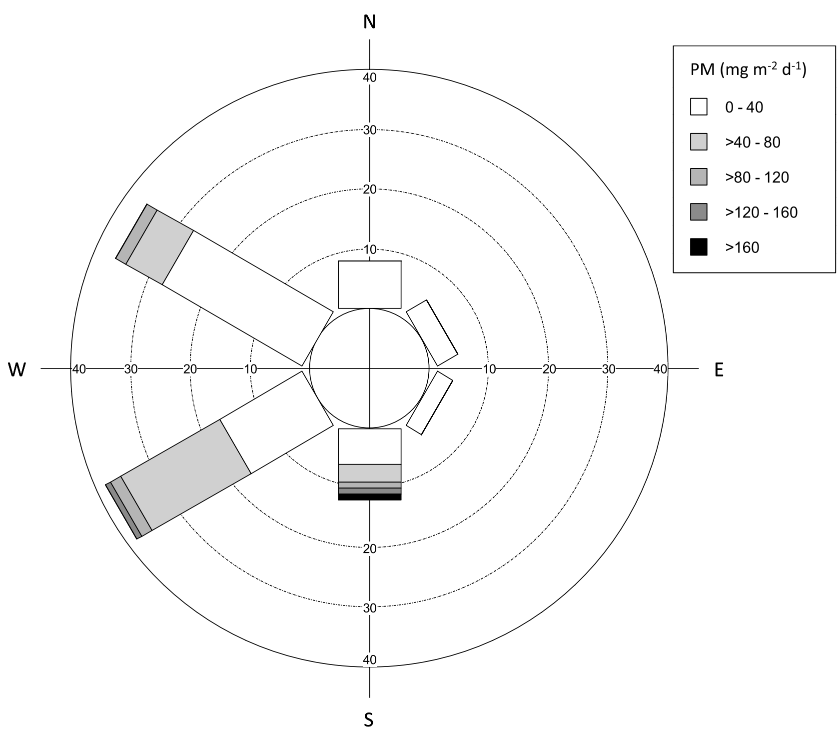

Figure 2. Particulate matter deposition (dry + wet) in Quéntar during the 2 years under study according to the direction of the air masses arriving at the study area. The directions of winds were determined by daily back trajectory analysis using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (NASA). Directions of the air masses were summarized in six categories, represented by the six bars in the figure. The direction of the air masses during each week was estimated as the mean value of the daily directions measured in degrees. The length of bars shows the number of weeks (concentric circle scale) in which the air masses come from the indicated direction. The grey scale refers to the PM amount deposited in the five categories in which the depositions per week are classified.

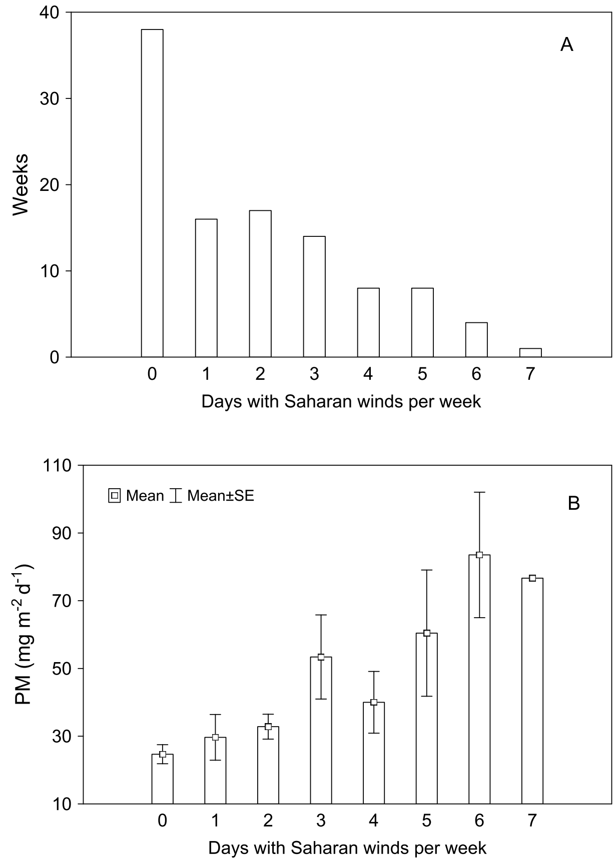

Figure 3. (a) Number of weeks with 0–7 days of winds from the Sahara per week during the 2 years in which atmospheric deposition was measured at the Quéntar site. The presence of Sahara winds was determined by daily back trajectory analysis using the HYSPLIT model. (b) Mean and standard errors of dry + wet particulate matter (PM) collected during these weeks at the Quéntar site.

Separate samples of dry and wet deposition were collected weekly from February 2004 to February 2006 in Quéntar (106 weeks) and simultaneously at Béznar and Cubillas during the spring-summer periods (from May to October in 2004 (21 weeks) and from March 2005 to October in 2005 (28 weeks)). Samples were collected in a MTX® ARS 1010 automatic deposition sampler (100 cm long, 60 cm wide, and 140 cm high) located on a platform at ground level at a meteorological station near each reservoir, closed to public access, and managed by the Spanish water agency "Confederación Hidrográfica del Guadalquivir." On each sampling day, dry and wet deposition buckets were replaced and taken to the laboratory. Dry deposition samples were obtained by rinsing the bucket with 1000 mL of Milli-Q® ultrapure water. The volume of rain in the wet deposition bucket was recorded, and a 1000 mL aliquot was drawn for analysis. When necessary, the rain volume was brought up to 1000 mL with Milli-Q® ultrapure water. The concentration of particulate matter (PM) in dry and wet depositions was determined as dry weight (60°C, 24 h) using Whatman GF/F glass fiber filters. Prior to filtration, 50 mL subsamples were taken to analyze TP and TN concentrations, employing previously published analytical methods [*Morales-Baquero et al.*, 2006].The collection of dry atmospheric deposition is known to be complex and potentially affected by the specific properties of the sampler [*Wesely and Hicks*, 2000]; however, these effects would be minimized due to the relatively large size of the Saharan dust aerosol, which would ensure that sedimentation is the main delivery process [*Alados-Arboledas et al.*, 2003; *Mladenov et al.*, 2010].

The amounts of PM, TP, and TN collected weekly in dry and wet deposition samples were converted to daily deposition units by considering the surface area of the sampler (0.07 m^2) and, in the case of wet deposition, the rain volume. Data were gathered on dry PM, TP, and TN deposition, wet PM, TP, and TN deposition, and the sum of dry and wet (d + w) PM, TP, and TN.

2.2. Remote Sensing and Statistical Analysis

The presence of air masses from Sahara over the study area was determined by daily back trajectory analysis (5 days, 3000 m asl) using the Hybrid Single-Particle Lagrangian Integrated Trajectories Model [*Draxler and Rolph*, 2003; *Rolph*, 2003] of the National Oceanic and Atmospheric Administration Air Resources Laboratory (USA).The height of 3000 m asl was selected for the calculation of back trajectories because it is above the PBL in the study area (annual mean of 1700 ± 500 m asl, [*Granados-Muñoz et al.*, 2012]) and is in the mainstream of atmospheric Saharan dust mobilization (between 1500 and 4000 m asl [*Talbot et al.*, 1986]). Figure 2 shows the frequency of weeks as a function of the average direction of back trajectories during the week (measured in degrees) and the amount of deposition. As can be observed, higher deposition values were obtained when the air masses came from the South or Southwest, i.e., from the direction of the Sahara. Oriana circular statistics [*Kovach*, 2006] were used to construct this figure. The strength of Saharan influence per sampling week was

Table 1. Cumulative Dry, Wet, and Dry + Wet Deposition Values Recorded at the Study Sites During Annual (Quéntar) or Spring and Summer Periods (Quéntar, Béznar, and Cubillas) in 2004 and 2005

	2004			2005		
	Dry	Wet	D + W	Dry	Wet	D + W
PM (g m^{-2})						
Quéntar						
annual	10.2	2.5	12.7	10.6	4.1	14.8
spring-summer	6.4	1.3	7.7	6.0	2.0	8.0
Béznar						
spring-summer	7.3	1.1	8.4	7.1	1.0	8.0
Cubillas						
spring-summer	7.5	1.7	9.2	9.2	1.6	10.8
TP (μmol m^{-2})						
Quéntar						
annual	415.7	260.3	676.0	470.2	108.8	579.0
spring-summer	242.6	61.9	304.5	231.9	38.2	270.1
Béznar						
spring-summer	279.3	13.2	292.5	326.8	86.2	413.0
Cubillas						
spring-summer	351.4	78.4	429.8	413.8	84.2	498.0
TN (mmol m^{-2})						
Quéntar						
annual	6.5	15.3	21.8	12.4	14.5	26.9
spring-summer	3.8	7.6	11.4	3.9	6.1	10.0
Béznar						
spring-summer	6.4	1.0	7.3	4.9	2.9	7.8
Cubillas						
spring-summer	7.2	5.2	12.4	8.3	3.2	11.4

defined by the number of days in the week under air masses with a back trajectory that revealed Saharan origin. Weeks with more than 2 days of Saharan winds showed higher deposition values (Figure 3). We examined the influence of Saharan dust intrusions on the amount of PM, TP, and TN deposited during spring-summer at the sites by comparing values between weeks with no Saharan winds (hereafter designated weeks without intrusions) and those with Sahara winds for more than 2 days (hereafter weeks with intrusions). Assuming that the deposition of long-range transported dust occurs at a regional scale (i.e., Saharan dust intrusions would deliver materials simultaneously at the three study sites), the intrusions should synchronize depositions among the sites and increase their intercorrelation. This issue was examined by performing Pearson correlation analyses. Higher mean deposition values can be expected in weeks with versus without Saharan intrusions; the Mann-Whitney *U* test was used to examine this issue, given the nonnormal distribution of the variables. Linear regression analysis was applied to examine the dependence of TP and TN deposition on PM deposition. Finally, three two-way factorial analyses of variances (ANOVAs) were performed to determine the dependence of PM, TP, and TN deposition on the site and/or on the Saharan intrusions, with the deposition rate as dependent variable and the site and presence/absence of Saharan intrusions as factors. Statistica Software [*StatSoft Inc*., 1997] was used for the statistical analyses.

3. Results

3.1. Atmospheric Deposition

In Quéntar reservoir, the contribution of dry deposition to the annual (d + w) deposition of PM and TP was markedly higher than that of wet deposition, whereas the latter made the largest contribution to the annual (d + w) deposition of N (Table 1). During spring-summer periods, the contribution of dry deposition to (d + w) PM deposition and to (d + w) TP deposition was significantly higher than that of wet deposition at all three sites (Table 1). In the case of TN, there was a lesser difference between dry and wet depositions, and no pattern could be detected (Table 1).

The weekly (d + w) deposition of PM and TP in Quéntar varied seasonally, with maximum values during spring-summer and minimum values during winter (Figures 4a and 4b). The range of variation in PM and

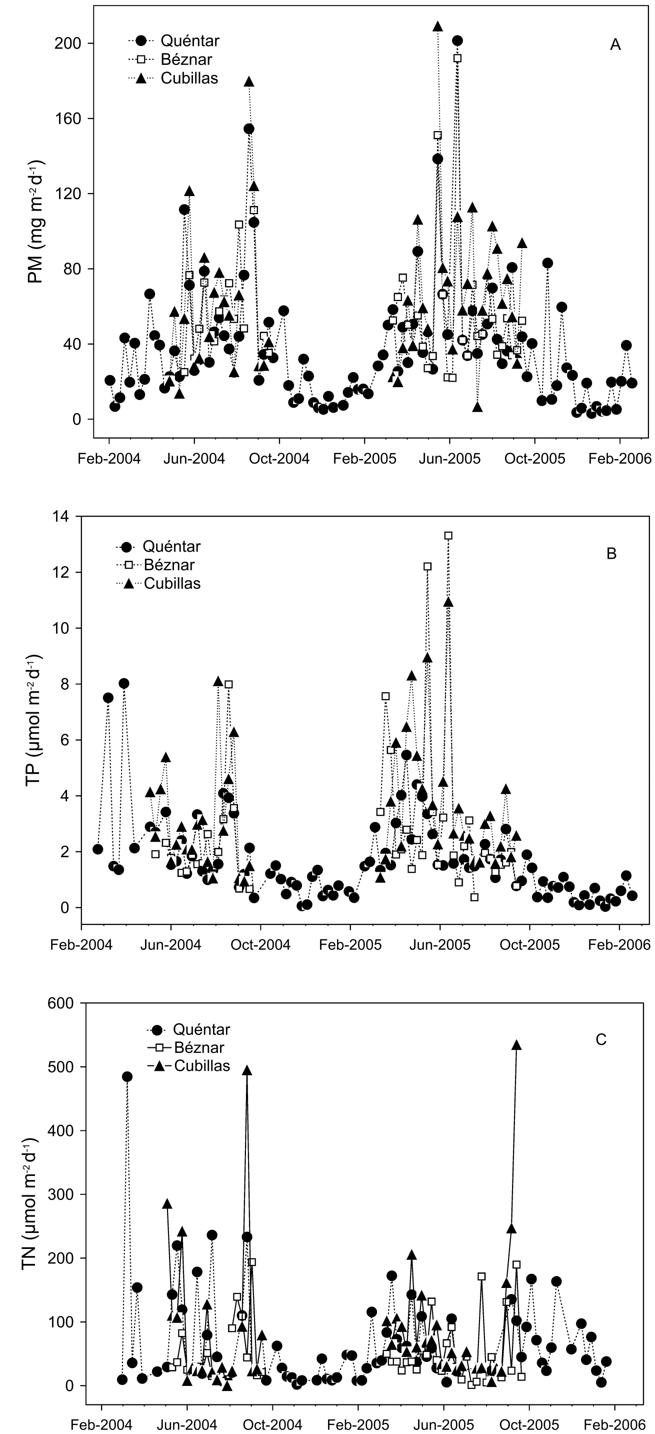

Figure 4. Seasonal dynamics of total (dry + wet) weekly atmospheric deposition of (a) particulate matter (PM), (b) total phosphorus (TP), and (c) total nitrogen (TN) at sites in Quéntar, Béznar, and Cubillas. Results of Pearson correlations between sites are shown in Table 2.

TP weekly (d + w) deposition during spring-summer was of the same order of magnitude across all three sites, which showed significant or marginally significant intersite correlations for PM and TP deposition values (Table 2). TN weekly (d + w) deposition values showed no clear seasonal pattern (Figure 4c); thus, although high values were usually recorded during spring-summer, they were also observed during autumn-winter, especially in 2005. The range of variation in weekly TN (d + w) deposition during spring-summer was of the same order of magnitude across all three sites, although a significant correlation was not observed between all sites (Figure 4c and Table 2).

When all samples collected at the three sites were considered, regression analyses showed that PM deposition values explained almost half of the variability in TP deposition but only 8% of the variability in TN deposition (Figure 5).

3.2. Saharan Versus Local Influence

Table 2 exhibits the correlations among the three study sites in the weekly (d + w) depositions of PM, TP, and TN, differentiating between weeks with and without Saharan intrusions. PM and TP depositions were synchronous among the three sites during periods of Saharan intrusions, whereas no temporal correlation was found during periods without Saharan intrusions. TN deposition values showed no pattern related to periods with or without Saharan intrusions.

During spring-summer, the mean daily rates of PM and TP (d + w) deposition were significantly or marginally significantly higher during the weeks with Saharan intrusions at all three sites (Figures 6a and 6b). In contrast, mean daily rates of TN (d + w) deposition showed no significant differences between periods with and without Saharan intrusions (Figure 6c). Increases in mean PM deposition values in weeks with Saharan intrusions versus weeks without intrusions were of the same order of magnitude in each site (35.7 mg m^{-2} d^{-1} in Quéntar, 27.2 mg m^{-2} d^{-1} in Béznar, and 47.8 mg m^{-2} d^{-1} in Cubillas), as were the increases in mean TP deposition values (0.8 µmol m^{-2} d^{-1} in Quéntar, 1.7 µmol m^{-2} d^{-1} in Béznar, and 1.0 µmol m^{-2} d^{-1} in Cubillas).

Table 2. Correlation Matrices for PM, TP, and TN (d + w) Deposition Among the Three Sites During Spring/Summer Periods, Considering All Weeks As Well As Weeks With or Without Saharan Intrusions[a]

	All the Weeks			Weeks With Saharan Intrusions			Weeks Without Saharan Intrusions		
	Quéntar	Béznar	Cubillas	Quéntar	Béznar	Cubillas	Quéntar	Béznar	Cubillas
PM									
Quéntar	1	*0.75*	*0.64*	1	*0.77*	*0.64*	1	0.19	0.28
Béznar	$p < 0.000$	1	*0.62*	$p < 0.000$	1	*0.60*	$p = 0.601$	1	0.40
Cubillas	$p < 0.000$	$p < 0.000$	1	$p < 0.000$	$p < 0.000$	1	$p = 0.437$	$p = 0.249$	1
TP									
Quéntar	1	*0.30*	*0.53*	1	*0.36*	*0.53*	1	0.03	0.57
Béznar	$p = 0.058$	1	*0.42*	$p < 0.05$	1	*0.42*	$p = 0.932$	1	0.40
Cubillas	$p < 0.000$	$p < 0.05$	1	$p < 0.01$	$p < 0.05$	1	$p = 0.088$	$p = 0.252$	1
TN									
Quéntar	1	−0.02	*0.47*	1	0.06	*0.44*	1	−0.29	*0.94*
Béznar	$p = 0.886$	1	*0.36*	$p = 0.762$	1	*0.51*	$p = 0.534$	1	−0.33
Cubillas	$p < 0.01$	$p < 0.05$	1	$p < 0.05$	$p < 0.01$	1	$p < 0.01$	$p = 0.469$	1

[a]Weeks without Saharan intrusions are defined as those with no winds from the Sahara and weeks with Saharan intrusions as those with more than 2 days with winds from the Sahara. Statistically significant ($p < 0.05$) or marginally significant values ($p < 0.08$) are in italic. The lower half of the symmetric matrices shows the probability values of corresponding correlations.

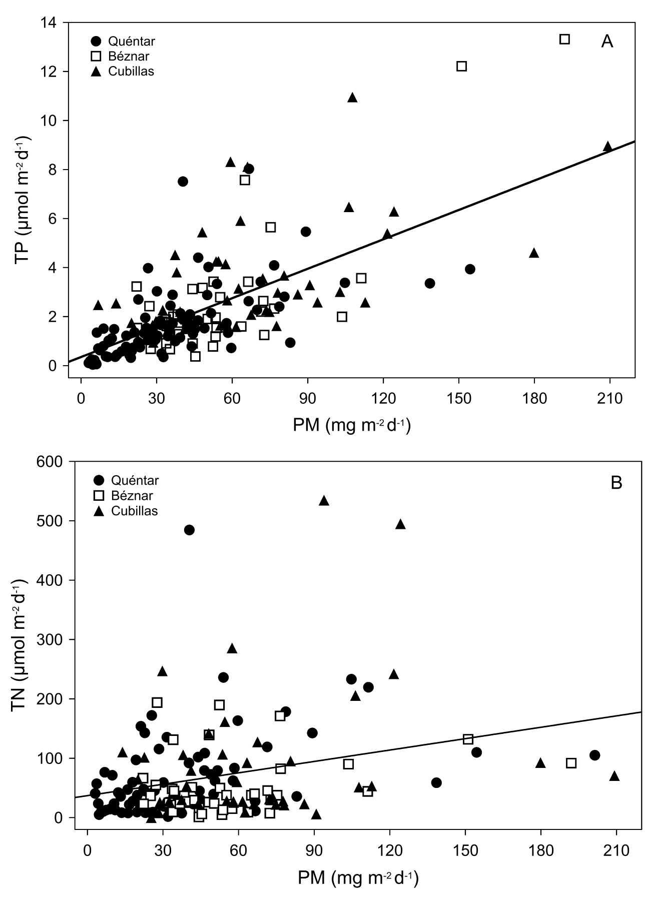

Figure 5. Linear regression between the weekly deposition of particulate matter (PM) and (a) total phosphorus (TP) and (b) total nitrogen (TN) in the three study sites. All samples collected at the three sites were considered in analyses. Linear regression results: (Figure 5a) TP = 0.35 + 0.04 PM, $r^2 = 0.46$, $p < 0.0000$ and (Figure 5b) TN = 37.01 + 0.64 PM, $r^2 = 0.08$, $p < 0.001$.

ANOVA results (Table 3) revealed that Saharan intrusions significantly affected both PM and TP values, whereas the site variable only affected TP deposition values. TN deposition was not affected by site or by Saharan intrusion. In the three ANOVAs, the interaction between sites and intrusions was not significant, indicating the independence of these two factors.

4. Discussion

4.1. PM, TP, and TN Deposition Patterns

Saharan dust is a source of PM and P for ecosystems of the southeastern Iberian Peninsula [*Morales-Baquero et al*., 2006, 2013]. The export of Saharan dust to the Mediterranean basin is controlled by synoptic meteorological scenarios extensively studied by *Escudero et al*. [2005] and *Querol et al*. [2009]. They concluded that Saharan dust mobilization over the Iberian Peninsula occurs mainly between May and October. The results obtained for PM and TP depositions in this study are in accordance with patterns previously described in the southeastern Iberian Peninsula. The increased deposition of PM and TP in spring and summer corroborates the findings of a study conducted in 2001 and 2002 at a location close to the study

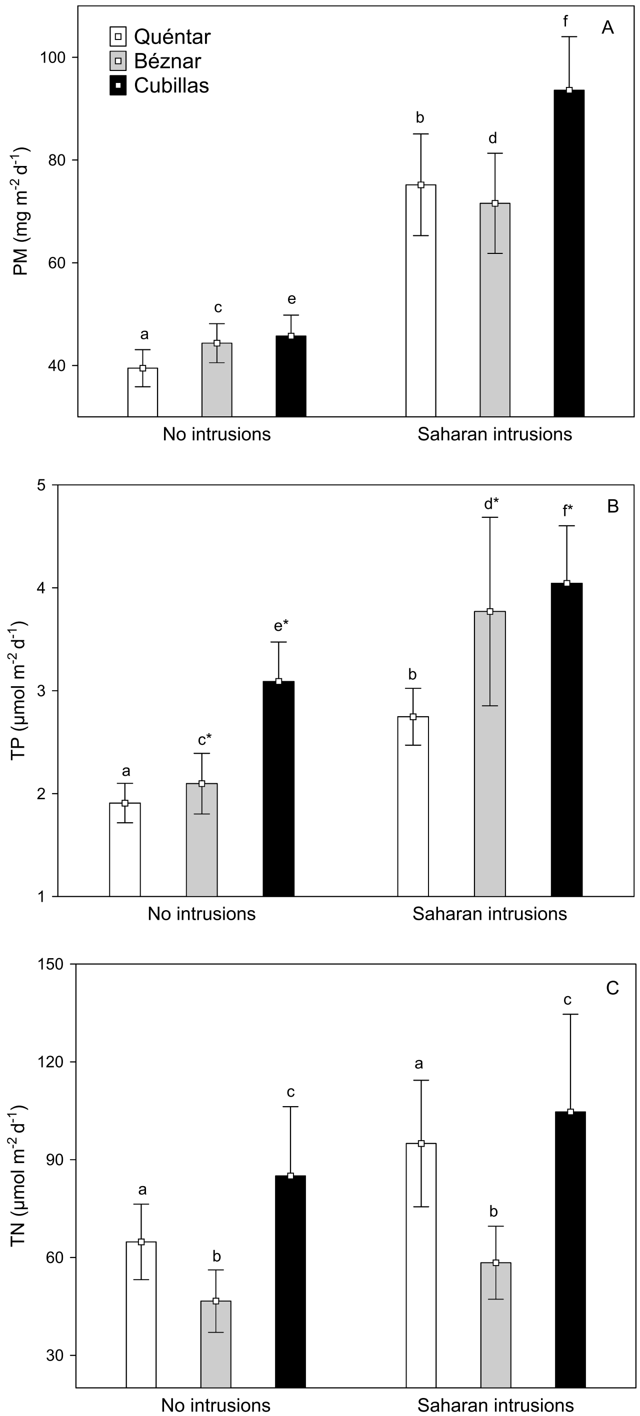

Figure 6. Mean daily deposition rates during weeks without Saharan intrusions and weeks with Saharan intrusions. Weeks without Saharan intrusions are defined as those with no winds from Sahara and weeks with Saharan intrusions as those with more than 2 days of winds from Sahara. Significant ($p < 0.05$) or marginally significant ($p < 0.08$) increases at each site (Mann-Whitney U test) are indicated by a different letter above each pair of columns of the same site (asterisk = marginally significant).

sites [*Morales-Baquero et al.*, 2006]. In the spring and summer, the intense heating and consequent development of the North African thermal low generate a convective system in the western part of the Mediterranean that pumps desert dust up to a height of 5000 m asl. Part of this dust is transported toward the North, with the vast majority of episodes over the Iberian Peninsula occurring without local convective rainfall [*Escudero et al.*, 2005]. Accordingly, the maximum concentration of aerosols over the study area was observed in spring-summer, when rainfall values were lowest (Figure 7) and when the highest deposition values were measured (Figure 4).

The annual PM (d + w) deposition values obtained in this study were slightly higher than those obtained in 2001 and 2002 (10.9 and 11.3 g m^{-2} yr^{-1}, respectively) [*Morales-Baquero et al.*, 2006]. Likewise, annual TP (d + w) deposition values were similar to or slightly higher than those obtained in 2001 and 2002 (635.6 and 390.2 µmol m^{-2} yr^{-1}, respectively). The increased values of PM and TP deposition are consistent with a greater presence of Saharan aerosols over the western Mediterranean during 2004–2005 than during 2001–2002 [*Pey et al.*, 2013]. In addition, the concentration of aerosols over the study area was higher during 2004–2005 than during 2001–2002. Figure 7 shows that there were six months during 2004–2005 with an average aerosol optical depth of more than 0.30 (unitless), whereas this value was only reached in 1 month during 2001–2002 [*Morales-Baquero et al.*, 2013].

When data for the three study sites were pooled in a scatterplot (Figure 5), the relationship between PM and TP depositions indicated a regional coherence consistent with a major contribution of Saharan dust to TP deposition. Thus, linear regression analysis revealed that almost half of the variability in TP deposition was explained by PM deposition, which explained only 8% of the variability in TN deposition. Hence, although dust can transport some N compounds adsorbed on the surface of mineral

Table 3. Results of ANOVAs Performed to Assess the Influence of Site (Quéntar, Béznar, and Cubillas) and Presence of Saharan Intrusions on log(PM), log(TP), and log(TN) Weekly Depositions[a]

Source of Variability	df	Ms	*F*	*p*
PM				
Site	2	0.07	1.78	ns
Intrusions	1	2.27	56.36	0.000
Interaction	2	0.07	1.77	ns
Error	132	0.04		
TP				
Site	2	0.34	5.35	0.006
Intrusions	1	0.87	13.82	0.000
Interaction	2	0.01	0.09	ns
Error	128	0.06		
TN				
Site	2	0.09	2.76	ns
Intrusions	1	0.07	2.05	ns
Interaction	2	0.01	0.16	ns
Error	123	0.03		

[a]Weeks with Saharan intrusions were defined as those with more than 2 days of winds from the Sahara. Samples collected at the three sites during the spring/summer periods were considered in analyses. (df: degrees of freedom; Ms: mean square; ns: not significant)

particles [*Dentener et al*., 1996] and Saharan intrusions significantly augment atmospheric TN deposition during weeks without rain [*Morales-Baquero et al*., 2013], Saharan dust makes a small contribution to N inputs in ecosystems in the studied area. Wet deposition made the predominant contribution to annual TN (w + d) deposition (Table 1), as previously reported [*Morales-Baquero et al*., 2006, 2013]. Gaseous forms of N compounds dissolved in the rain appear to be the most plausible source of N in this area, although it receives lower atmospheric N inputs in comparison to other Mediterranean zones [*Morales-Baquero et al*., 2006] or heavily industrialized areas in central Europe [*Holland et al*., 2005].

4.2. Saharan Versus Local Influence

The synchrony of a given parameter among spatially distant ecosystems is considered to indicate a significant (external) climatic control in the region [*Baines et al*., 2000]. The synchronous dynamics among the three study sites for PM deposition (Figure 4 and Table 2) and previous findings at higher altitudes in this area [*Morales-Baquero et al*., 2006; *Pulido-Villena et al*., 2008] indicate that intrusions of Saharan dust over the Iberian Peninsula are a major climate driver of long-range transported PM, whose deposition adds to that from local sources. In fact, when only the weeks without Saharan intrusions are considered, the correlation of PM deposition among sites disappears (Table 2). According to these results, the novel methodology applied in our study appears to be an effective method for estimating the deposition of long-range transported dust. The ability to distinguish this from locally derived dust, based on statistical relationships among different sampling sites, can likely be useful in areas under the influence of the world's dust belt [*Prospero et al*., 2002] and in regions in the Southern Hemisphere subject to high dust aerosol deposition [*Mladenov et al*., 2011].

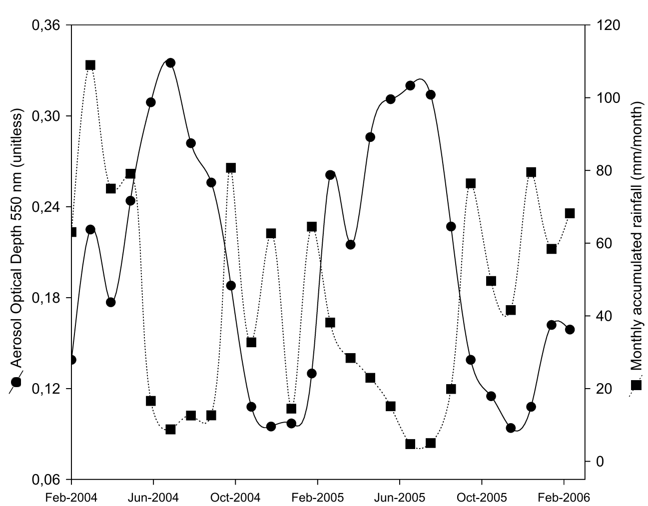

Figure 7. Times series monthly averages of aerosol optical depth (AOD, solid line) provided by NASA Giovanni portal (http://gdata1.sci.gsfc.nasa.gov/daac-in/G3/gui.cgi?instance_idMISR_Monthly_L3) and rainfall (dashed line) (http://gdata1.sci.gsfc.nasa.gov/daac-bin/G3/gui.cgi?instance_idGPCC_Monthly).Values are integrated for the study area shown in the frame of Figure 1.

The average amount of PM collected at each site during spring-summer in the weeks without Saharan intrusions can be considered as their background levels, which are relatively similar to each other (39.5, 44.4, and 45.7 mg m^{-2} d^{-1} in Quéntar, Béznar, and Cubillas, respectively). The increase in PM deposition over these background levels during weeks with Saharan intrusions was similar among the three sites (36.9 mg m^{-2} d^{-1} on average, equivalent to 75 kg h^{-1} yr^{-1}), being about 85% above background levels. The similar increase in PM deposition at the three sites from Saharan intrusions

(Figure 6) suggests that Saharan aerosols are homogeneously deposited at the spatial scale under study. In fact, results of the factorial ANOVAs showed that Saharan intrusions increased PM deposition but that there were no differences in PM deposition among the three sites (Table 3).

Saharan intrusions increased the mean TP deposition at the three sites in a homogeneous manner (Figure 6 and Table 3). The increase in the weeks with Saharan intrusions versus the weeks with no Saharan intrusions reflects the deposition of long-range transported TP and was relatively uniform among the sites (mean of 1.1 μmol TP m^{-2} d^{-1} for the three sites), representing a 29% increase for Cubillas, which showed the highest values of background atmospheric P deposition, 42% for Quéntar, and 81% for Béznar. This Sahara-supplied P, equivalent to 0.07 kg P h^{-1} yr^{-1}, was markedly higher than has been observed at more northern sites in Spain (0.03 kg P ha^{-1} yr^{-1}, [*Avila and Alarcón*, 1998]), consistent with the elevated atmospheric concentration of Saharan dust over the southeastern Iberian Peninsula [*Pey et al*., 2013]. It is also higher than the dust contribution to P inputs in the Yucatan Peninsula (0.05 kg P ha^{-1} yr^{-1}), which is large enough to offset leaching and erosional losses from soils [*Das et al*., 2013], and higher than the dust contribution of P to Amazon Basin (0.023 kg P ha^{-1} yr^{-1}) which is comparable to the hydrological loss of P from the Basin [*Yu et al*., 2015].

The amount of Sahara-supplied P measured can be considered a genuine "new" input for the ecosystems in this region, and its importance for oligotrophic aquatic systems in the area was previously demonstrated [*Morales-Baquero et al*., 2006; *Pulido-Villena et al*., 2008]. The Sahara-supplied P input is within the range of the estimated release of P by weathering calculated by *Newman* [1995] for the world's rocks (0.01 to 1 kg P ha^{-1} yr^{-1}) and may represent an important long-term fertilization for terrestrial ecosystems in this region, because Mediterranean forest soils are usually P deficient [*Hinsinger*, 2001; *Sardans and Peñuelas*, 2004, 2007]. *Di Castri* [1981] found that 0.73–1.26 kg P ha^{-1} yr^{-1} returned to the soil in litter fall in Mediterranean-type shrublands in France. If these figures are valid for shrublands in the southeastern Iberian Peninsula, Saharan P input may represent up to 10% of the P available for plant growth, given that P contribution by cycling is the main source of P for plants in Mediterranean areas [*Menéndez et al*., 2003], while P input by mineral weathering is insignificant in these weathered soils [*Walker and Syers*, 1976; *Campo and Gallardo*, 2012]. In this context, *Moreno-Marcos and Gallardo-Lancho* [2002] reported an atmospheric deposition of 0.16 kg P ha^{-1} yr^{-1}, which matched the annual tree demand for aboveground growth in an oligotrophic *Quercus* forest in central Iberian Peninsula. We measured a similar (Saharan + local) P input in Quéntar of 0.21 kg P ha^{-1} yr^{-1}, with a Saharan P contribution of about 30%.

In contrast to PM and TP, atmospheric inputs of TN in the three sites showed no synchrony, even when considering only weeks with Saharan intrusions (Table 2). ANOVA results showed no significant increases in TN during weeks with Saharan intrusion and no differences among sites (Table 3), confirming that Saharan dust makes no relevant contribution to the N budget of ecosystems in this area.

However, factorial ANOVA results revealed differences in TP deposition among sites (Table 3). The mean deposition values recorded during weeks without Saharan intrusions can be considered as the background deposition for each site (1.8 to 3.2 μmol TP m^{-2} d^{-1}, Figure 6). The differences found demonstrate the local effects on TP deposition, in agreement with the current view of the greater susceptibility of atmospheric P deposition to the influence of local sources in comparison to other dust components [*Kauffman et al*., 1994; *Newman*, 1995; *Kopacek et al*., 1997].

The homogeneity of the deposition of PM and the associated TP among the sites contrasts with the heterogeneity of background TP deposition. This can be explained by the difference in the sources of atmospheric TP in relation to the PBL. The main known sources of atmospheric TP, besides dust and volcanic ash, include plant pollen and fly ash from the burning of plant material or fossil fuels [*Newman*, 1995]. These sources of TP are produced from the ground below the PBL height, while long-range transported PM and the associated TP fall from above the PBL.

All three study sites are below the PBL height in this region (see section 2), and they may differ in the burning of plant material or fossil fuels. However, the differences in vegetation among Quéntar (forest), Béznar (agriculture + forest), and Cubillas (agricultural management) may be largely responsible for the differences in local TP deposition, because P deposition is sensitive to pollen emissions [*Carlisle et al*., 1966; *Jordan et al*., 1995]. In fact, the pollen aerosol is part of the water insoluble organic matter retained in the filters, which represents an appreciable proportion of the PM deposited in this area (average of 7% of PM in dry deposition [*Mladenov et al*., 2010]). Furthermore, the pollen is rich in P, with values of 2.8 mg g^{-1} being reported

[*Newman*, 1995] and would augment the TP deposition at the study sites. However, the dispersion of constituents by the PBL is negatively related to their sedimentation velocity. Thus, large or heavy particles would have a higher sedimentation velocity, shorter time in the PBL, and lesser dispersion distances. For instance, a common pollen in the area is *Zea mays* (corn), 90 μm in size, which settles at 30.95 cm s^{-1} [*Di-Giovanni et al.*, 1995], whereas the pollen of *Lycopodium* sp., which has the more typical plant pollen size of ~ 30 μm, settles at 2.15 cm s^{-1} [*Di-Giovanni et al.*, 1995]. At any rate, these velocities are considerably higher than those for the dry sedimentation of mineral particles, e.g., 0.018 cm s^{-1} for particles with mean effective radius of 0.73 μm [*Tegen and Fung*, 1994]. In our study area, the mean effective radius of aerosols in the atmosphere under Saharan dust intrusions was reported to range between 0.72 and 1.63 μm (calculated from a distribution in volume) [*Mladenov et al.*, 2010]. Therefore, small mineral particles with low sedimentation velocities that enter the PBL from the mainstream of Saharan dust transportation toward Europe, which reaches up to 5000 m asl [*Escudero et al.*, 2005], may be uniformly distributed among the three study sites by the mixing effects of the layer; however, this would not be the case for pollen particles. Given that the production of pollen presumably differs among the study sites, its low dispersal capacity would lead to intersite variations in pollen deposition, which would explain the different background levels of TP deposition among the sites.

5. Conclusions

The long-term analysis of systematic and simultaneous dust deposition at three sites, covering a smaller surface at ground level than was covered by the plumes of dust transported in the free troposphere, revealed an increase at all sites when they were below these plumes, which can be statistically detected. This novel approach is likely to be useful in any region subject to long-range dust deposition, including those affected by the so-called global dust belt [*Prospero et al.*, 2002].

Our results show that long-range transported Saharan dust is a major source of PM deposition in the southeastern Iberian Peninsula and makes a significant contribution to atmospheric TP inputs in this region. Our results confirm the well-documented association between Saharan dust and P content. The deposition of PM and TP followed the same temporal pattern, with higher values in spring and summer, when the frequency of Saharan dust intrusions is greater. We also observed synchrony in the PM and TP deposited among the sites, which are around 40 km apart; this synchrony was greater when only weeks with Saharan intrusion were considered but disappeared when only those without intrusions were considered. PM deposition explained around half of the variability in TP depositions at the three study sites but only a small percentage of the variability in TN deposition.

No clear local effects were observed on PM or TN deposition, whereas TP deposition showed local background differences with respect to Saharan inputs. Nevertheless, the deposition of Saharan dust and the associated TP appears to be evenly distributed in the area, given that the comparison of deposition values at the study sites between weeks with and without Saharan intrusions revealed similar increases in PM and TP deposition. The increased PM deposition during the weeks with Saharan intrusions (mean of 36.9 mg m^{-2} d^{-1}) was around 85% over local background levels, reflecting a strong impact of Saharan dust on ecosystems in the southeastern Iberian Peninsula. The increased TP deposition (mean of 1.1 μmol TP m^{-2} d^{-1}) during weeks with versus without Saharan intrusions represents a genuinely new input of P for ecosystems in the region. However, their local significance depends on the background P deposition, and the increase in P from Saharan inputs over background levels ranged from 29% to 81% at the three study sites.

The annual input of Saharan P in the southeastern Iberian Peninsula would be somewhat greater than the 0.07 kg P ha^{-1} yr^{-1} measured during the periods of more intense Saharan intrusions. It may represent an important long-term fertilization of these Mediterranean ecosystems, whose P limitation is likely to intensify under the drier conditions forecast for the Mediterranean region [*Intergovernmental Panel on Climate Change*, 2013], when increasing drought could decrease P soil availability for plants [*Sardans and Peñuelas*, 2004].

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