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African dust influence on ambient PM levels in South-Western Europe (Spain and Portugal): A quantitative approach to support implementation of Air Quality Directives

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Abstract. This manuscript proposes and validates a methodology for the quantification of the daily African PM load during dust outbreaks in southern Europe. The daily net dust load in PM_{10} attributable to an African episode can be obtained by subtracting the daily regional background (RB) level from the PM_{10} concentration value at a RB station. The daily RB level can be obtained by applying a monthly moving 30th percentile to the PM_{10} time series at a RB station after a prior extraction of the data coincident with African dust transport. For days with influence of African dust, the dust load is given by the difference between the daily PM_{10} values minus the daily PM_{10} RB levels. This method allows us to quantify the net African dust load without chemical speciation. The comparison between the estimated net load during African dust outbreaks (ADO) and the crustal load determined by chemical speciation of PM_{10} filters at three RB stations in Spain had resulted in a very good correlation (R^2=0.60-0.83), being the equivalence (correlation lines' slopes ~ 1) highly significant in the three cases.

1. Introduction

The determination of the levels of atmospheric particulate matter (PM) is a key parameter in the evaluation of air quality given the proven influence of this pollutant on human health [1] and climate ([2] and references therein). As regards the adverse health effects of PM, the European Commission (EC) issued the Directive 1999/30/EU on air quality. This Directive established PM_{10} as a parameter to be monitored, with annual (40 µg m^{-3}) and daily (50 µg m^{-3}, not to be exceeded more than 35 occasions per year) limit values.

In cases where the exceedances of the limit values can be attributed to natural phenomena such as the transport of natural particles from arid regions, they can be discounted after scientific validation (article 2.15 from the 1999/30/EC Directive).

In southern Europe, the ADO may have a great impact on the PM levels [3,4,5,6,7]. Thus, at the Spanish EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air pollutants in Europe) sites more than 70% of the exceedances of the daily PM_{10}

limit value are attributable to African dust contributions [8]. However, during ADO the PM_{10} fraction is not entirely composed of African crustal aerosols. [4] estimated that between 20-50% of the PM_{10} concentration at RB sites during ADO in NE Spain had a regional or local origin different from mineral dust. In addition, as discussed in [8], the formation of secondary aerosols is enhanced during ADO due to the interaction of local anthropogenic species with mineral dust components.

The most common approach to quantifying dust contribution is the development of chemical speciation studies. The main disadvantages of chemical speciation are the high economic cost and the length of the process.

In the light of the foregoing discussion, we propose a quantitative and simple methodology for estimating the net African dust load in PM_{10} during ADO. This methodology has been validated by chemical speciation data for three RB sites in Spain.

2. Methods

2.1. Measurements and interpretation

The variability of PM_{10} levels from thirteen RB stations was interpreted. The air quality monitoring stations selected are representative of the different climatic areas of the Iberian Peninsula (table 1 and figure 1). PM_{10} measurements at EMEP stations were performed by means of the gravimetric method making use of MCV-PM1025 samplers (30 m^3 h^{-1}). At Valderejo and Izki, PM_{10} measurements have been carried out using β-attenuation instrumentation and, at Monagrega, PM_{10} levels have been obtained with TEOM instrumentation. PM_{10} real time levels were corrected through the application of the correction factors obtained by comparison with the gravimetric measurements.

Table 1. Spanish RB stations providing PM_{10} data.

Station	Air quality monitoring network	Location	Altitude (m.a.s.l.)	PM_{10} monitoring	Sampling period (PM_{10} Chemical Speciation)
O Saviñao	EMEP	42º 38' N 07º 42' W	506	Gravimetric	2001-2003
Niembro-Llanes	EMEP	43º 26' N 04º 51' W	134	Gravimetric	2001-2003
Valderejo	Government of the Basque Country	42º 53' N 03º 14' W	911	β-Attenuation	Since 2000
Izki	Government of the Basque Country	42º 39' N 02º 30' W	835	β-Attenuation	Since 2001
Cabo de Creus	EMEP	42º 19' N 03º 19' E	23	Gravimetric	2001-2003
Els Torms	EMEP	41º 24' N 00º 43' E	470	Gravimetric	2001-2003
Monagrega	ENDESA	40º 59' N 00º 12' W	600	TEOM	Since 1996 (24/03/1999-06/07/2000)
Risco Llano	EMEP	39º 31' N 04º 21' W	1241	Gravimetric	2001-2003
Campisábalos	EMEP	41º 17' N 03º 09' W	1360	Gravimetric	2001-2003
Peñausende	EMEP	41º 17' N 05º 52' W	985	Gravimetric	2001-2003
Zarra	EMEP	39º 05' N 01º 06' W	885	Gravimetric	2001-2003
Barcarrota	EMEP	38º 29' N 06º 55' W	393	Gravimetric	2001-2003
Víznar	EMEP	37º 14' N 03º 28' W	1265	Gravimetric	2001-2003
Montseny	CSIC-Generalitat de Catalunya Goverment	41º 46' N 02º 21' E	720	Laser Spectrometer	Since 2002 (Since 2002)
Castillo de Bellver	Illes Balears Goverment	39º 34' N 02º 37' E	117	β-Attenuation	2004-2005 (08/01/2004-29/97/2005)

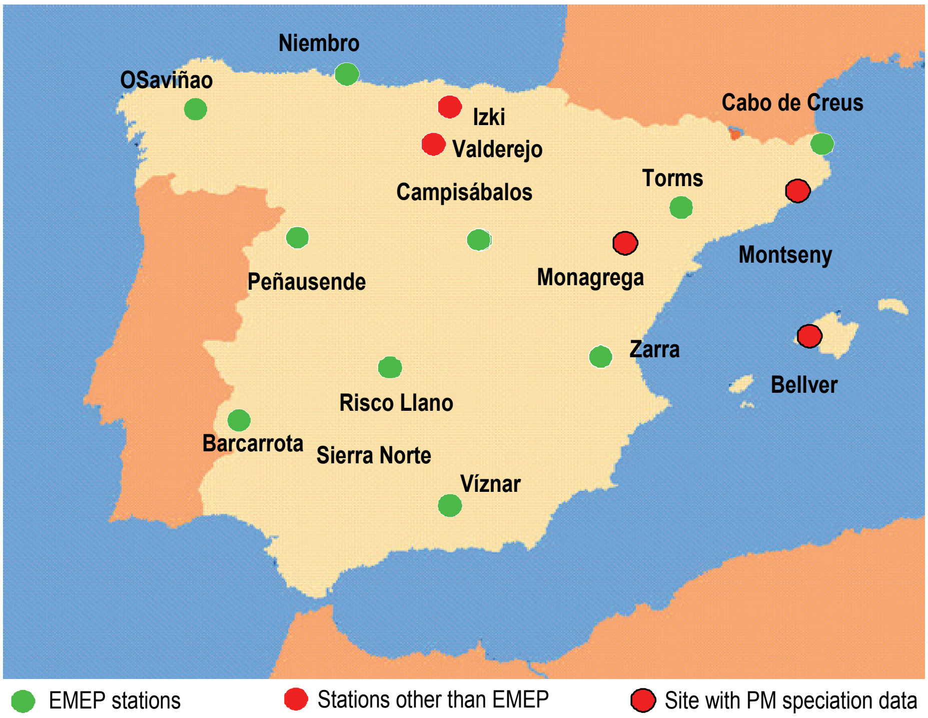

Figure 1. Spanish RB stations providing PM_{10} data and/or chemical speciation data.

The interpretation of the origin of the air masses reaching each of these monitoring sites was performed by calculating back-trajectories with the HYSPLIT4 model [9] accomplished with other tools such as meteorological maps, numerical models, and satellite images (see details in [5]).

2.2. PM_{10} speciation

In order to validate the methodology proposed here, data on chemical speciation of PM_{10} from three RB sites located in the Iberian Peninsula and the Balearic Islands were used (Monagrega, Montseny and Castillo de Bellver, table 1 and figure 1). 24-hour samples of PM_{10} were collected in quartz fibre filters twice per week. The instruments for the gravimetric samplings were high volume samplers working at 30 m^3 h^{-1}. At Monagrega and at Montseny, DIGITEL inlets were used whereas MCV-PM1025 instrumentation was used at Castillo de Bellver.
Once PM_{10} levels were gravimetrically determined, the filters were analyzed by different techniques to quantify the concentrations of approximately 60 PM components [10]. The concentrations of the major crustal components (SiO_2, Al_2O_3, CO_3^{2-}, Ca, Mg, Fe and K) accounted for >99% of the mineral dust load.

3. Results

3.1. Identification of African episodes affecting PM levels

The identification of the origin of the air masses reaching the regional monitoring sites and the interpretation of the origin of PM events was carried out following the above methodology (see details in [5]).

The persistence of some African dust episodes has been taken into account. It should be noted that PM_{10} levels may be high at RB stations one day before and/or two days after the period with proven influence of African dust. This could arise when the African air masses transported towards the Iberian Peninsula are not followed by episodes of intense advection. According to other studies, African dust transported towards southern Europe reaches a minimum altitude of 1500 m.a.s.l. [11]. The deposition of particles between 1 and 10 microns has an average speed of 0.6 cm s^{-1} [12]. Thus, levels of PM could remain high at a given site two days after the episode had ended and, therefore, the African period could also include these dates.

3.2. Calculation of the RB levels

During Atlantic advection episodes in Spain, PM_{10} levels are at their lowest owing to the reduced external contributions. At an specific RB site, the mean moving average of Atlantic advection can be considered as similar to the RB PM_{10} levels. This was performed at the 13 RB sites (table 1 and figure 1) for the period 2001-2003. The execution of this procedure requires daily interpretation of the air masses origin, and therefore, is not a straightforward method to be applied by the air monitoring networks.

In order to simplify the calculation of the RB PM_{10} levels, we propose the application of a monthly moving percentile to the PM_{10} series, after excluding the data corresponding to the days with ADO. This method is easier to apply given that only the days with influence of African dust transport need to be identified. In order to select the most adequate percentile, we performed a sensitivity test using different percentile thresholds (5, 10, 30, 40 and 50th) at the 13 RB stations. As shown in figure 2, the application of the 10 and 30th percentiles (y=1.44x and y=1.07x, respectively) underestimates the RB levels when compared with those calculated after considering only the days with Atlantic advection. By contrast, the use of the 40 and 50th percentiles yielded an overestimation of the RB levels (y=0.96x and y=0.86x, respectively). Thus, the 30th and 40th percentiles proved to be the most suitable. In order to reduce the underestimation of the African dust load we propose the adoption of the 30th percentile.

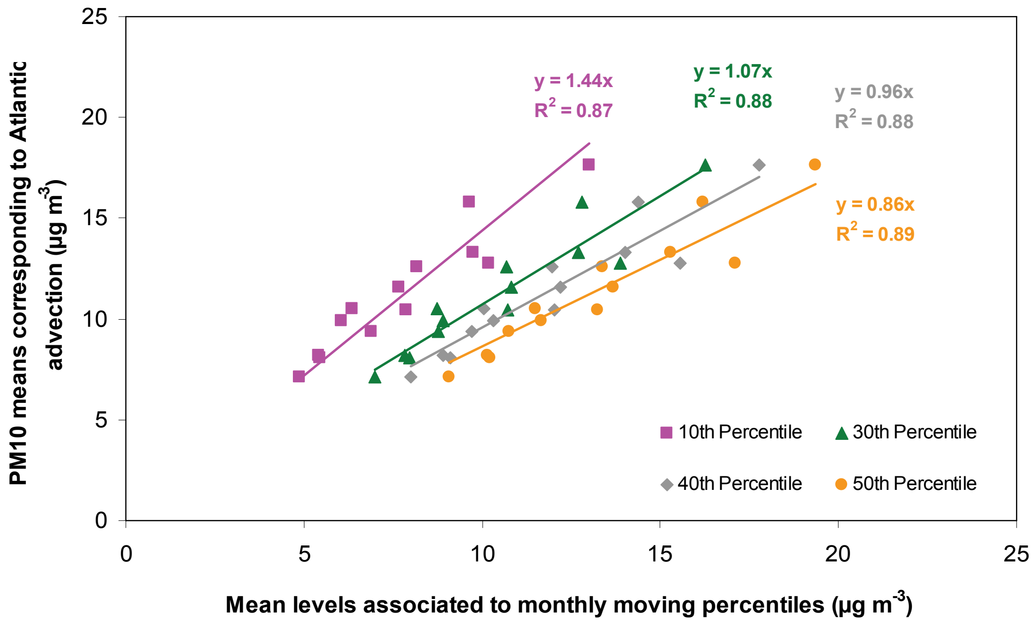

Figure 2. Correlation between the PM_{10} averages calculated only for the days with Atlantic advection (ATL) and the means of the time series obtained after applying different monthly moving percentiles (5th, 10th, 30th, 40th and 50th) to the PM_{10} data from 13 RB stations of the Iberian Peninsula for the period 2001-2003.

3.3. Calculation of the net African dust load and validation

The daily net African dust load was calculated by subtracting the daily RB level (obtained with the 30th percentile) from the daily PM_{10} value registered during an ADO.

In order to validate this method, we compared the mineral matter (sum of SiO_2, Al_2O_3, CO_3^{2-}, Ca, Mg, Fe and K) load for days with ADO from three RB monitoring stations (Monagrega, Montseny and Castillo de Bellver, Figure 1) with the net dust contribution calculated by using the proposed method. As shown in figure 3, the crustal load determined by chemical speciation strongly correlated ($0.60<R^2<0.69$) with the net African dust load calculated. Therefore, the net African dust load calculated is mainly composed of mineral matter, which corroborates the major natural origin of the PM_{10} fraction, although sulphation and nitration of dust is evidenced in specific cases.

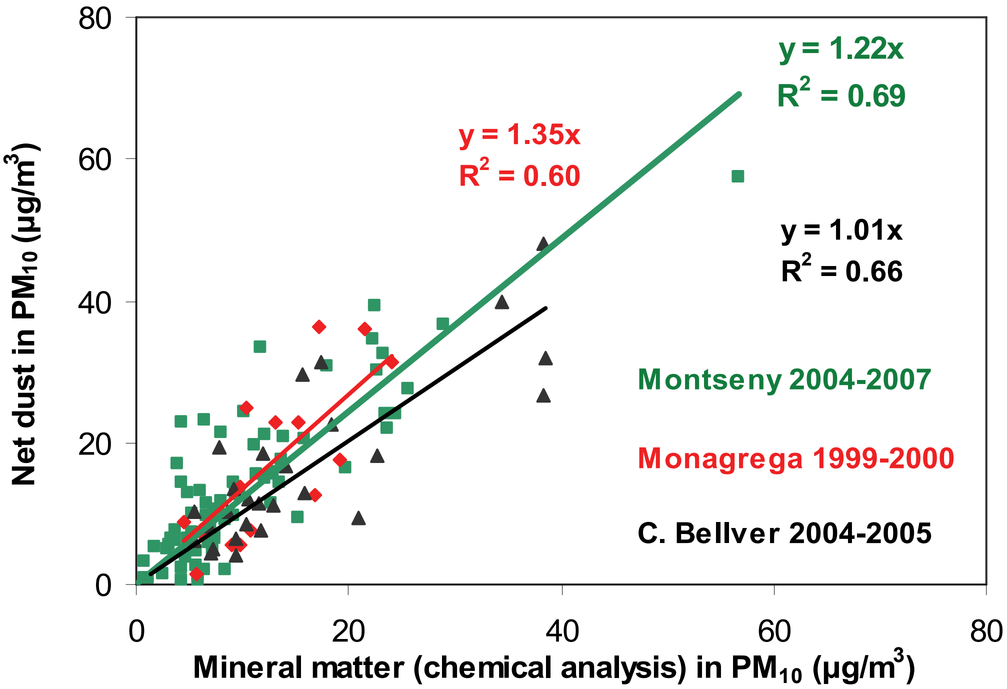

Figure 3. Correlation between the net African dust load obtained with the proposed methodology and the crustal load in PM_{10} determined by chemical speciation at Monagrega, Montseny and Castillo de Bellver.

3.4. Application of this method during exceedences of the daily limit value of PM_{10}

When the PM_{10} daily limit value is exceeded at urban or industrial stations during an ADO, the net African dust load can be used to evaluate the impact of the natural contribution to that exceedance. For example, at two stations, urban background and kerbside, 60 and 100 $\mu gPM_{10}\ m^{-3}$ are measured, respectively, on a given day during an ADO. Simultaneously, at the closest RB site, a daily value of 41 $\mu gPM_{10}\ m^{-3}$ is measured, and the monthly moving 30th percentile value is 10 $\mu g\ m^{-3}$ for this day. Therefore, the net African dust load in this region, for this day, is 31 $\mu gPM_{10}\ m^{-3}$. In this case, the exceedance at the urban background site can be attributed to the dust contribution (60-31 = 29 $\mu gPM_{10}\ m^{-3}$ as anthropogenic load), whereas the at the kerbside station must be ascribed to anthropogenic contributions (100-31 = 69 $\mu gPM_{10}\ m^{-3}$).

In some cases, negative values could be obtained when subtracting the net African dust load. In these situations, the net African dust load should be calculated at another RB station in the vicinity of the urban or industrial station at approximately the same altitude.

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