Atmospheric
Chemistry
and Physics

Open Access

Aerosols over continental Portugal (1978–1993): their sources and an impact on the regional climate

A. L. Morozova[1] and I. A. Mironova[2]

[1]CITEUC – Center for Earth and Space Research of the University of Coimbra, University of Coimbra, Coimbra, Portugal
[2]Earth Physics Department, Institute and Faculty of Physics, St.Petersburg State University, St.Petersburg, Russia

Correspondence to: A. L. Morozova (annamorozovauc@gmail.com, anna_m@teor.fis.uc.pt)

Abstract. Understanding of aerosol sources that affect climate is an ongoing problem facing scientists as well as policymakers. The role of aerosols in local climate variability depends on a balance between light absorbing and scattering particles as well as on variability of environmental conditions. In this paper we investigate variability of aerosol content (both absorbing and scattering ultraviolet radiation) over continental Portugal in connection with aerosol sources (volcanic eruptions, dust events, wildfires and anthropogenic pollution). The effect of the aerosol on the climate is studied analyzing their contribution to variations of temperature, pressure, sunshine duration and precipitation over Portuguese regions. The present analysis is based on a developed modern multiple regression technique allowing us to build the statistical correlation models to determine both the main local aerosol sources and aerosol's influence on the climate of continental Portugal during 1978–1993. The analysis allows us to conclude that the main sources driving the variations of the aerosol content over studied locations are wildfires, mineral dust intrusions and anthropogenic pollution. The relations between the aerosol content variations and the atmospheric parameters depend on the level of urbanization of the studied region, the type of aerosol and the season. The most significant finding is the decrease of the daily maximum temperature (and diurnal temperature range) related to the decrease of sunshine duration observed during the summer periods of increased content of the absorbing aerosols in the atmosphere.

1 Introduction

Aerosol particles scatter and absorb solar and terrestrial radiations depending on their microphysical and optical characteristics. These characteristics vary significantly due to various aerosol sources, both anthropogenic and natural. Natural sources include volcanic emissions, plant vapors and chemicals released by tiny sea creatures as well as dust from deserts. From the beginning of the industrial period, anthropogenic sources included not only farming and charcoal burning but also emissions from car exhausts, factories and power plants. Some aerosols, such as dust particles or sea spray, are mostly from natural origin. The other aerosols like sulfates and black carbon come from both natural and anthropogenic sources.

One of the important impacts of aerosols is their effect on the Earth's radiation budget. Aerosols affect it in two ways: as a direct and an indirect forcing. The direct aerosol radiative forcing is due to changes in solar irradiance and the indirect one is through aerosol effects on clouds. The report of the Intergovernmental Panel on Climate Change (IPCC, 2013, Ch. 7 "Clouds and Aerosols") indicates that aerosols have given an overall cooling effect on the Earth since pre-industrial times. This effect masked some of the global warming from greenhouse gases that would have occurred in the aerosols absence. Aerosols affect not only global climate but also, due to unevenly spatial distribution, local weather and climate, visibility and human health. The local aerosol influence on the regional weather conditions can be much stronger and lead to local climate changes rather than to global. Normally, aerosol scattering makes the Earth's atmosphere more reflective and leads to a cooling of the cli-

mate system, while aerosol absorption has the opposite effect and leads to warming. The balance between cooling and warming depends on microphysical and optical properties of aerosols.

Most of the previously published papers are dedicated to the analysis of the aerosol content variations over Portugal for relatively short time periods. These periods start mainly in 2002 when the facilities allowing for the in situ measurements of many aerosol parameters were established at the Évora Geophysics Centre Observatory, (38.57° N, 7.91° W; 293 m a.s.l.) – see, e.g., Pereira et al. (2011, 2012). These studies showed the strong dependence of the local aerosol content and its composition both on anthropogenic and natural sources. The latter include mineral dust intrusions and wildfires. The detailed analysis of the properties and time variations of the Portuguese aerosols can be found in Pereira et al. (2005), Pereira et al. (2008, 2011, 2012), Santos et al. (2008, 2013), Catry et al. (2010), Alves et al. (2011), Obregón et al. (2012), Vicente et al. (2012, 2013) and Evtyugina et al. (2013); the analysis of the radiative effect of the aerosols originated from wildfires for the close region in northwestern Spain is presented in Calvo et al. (2010). The impact of the aerosol variations on the local climate variations was studied as well, both with in situ measurements (e.g., Santos et al., 2008; Obregón et al., 2012) and modeling (e.g., Miranda et al., 2002; Santos et al., 2013). For example, a noticeable cooling at the surface level was observed due to aerosols consisting of desert mineral dust and forest fire products (Santos et al., 2008; Calvo et al., 2010).

The present paper is dedicated to understanding the local and global aerosol sources and the effect of the local aerosol content in climate variations of the continental Portuguese region for the time period 1978–1993. Here we take into account a number of different types of aerosols, their local and global sources and their relations with variations of some local climatic parameters: sunshine duration, precipitation, pressure and temperature. The satellite-based TOMS (Total Ozone Mapping Spectrometer) atmospheric aerosol index helps us to obtain information about the aerosol content in the studied region. This approach allows us to minimize the effect of the well-known spatial heterogeneity of the aerosol content. The information about climatic parameters variability is received from the Geophysical Institute of University of Coimbra and the Geophysical Institute of Instituto Dom Luiz of University of Lisbon. As aerosol sources we considered volcanic eruptions, Saharan dust, forest fires and anthropogenic pollution. The length of the studied period (about 15 years) allows us to analyze long-term variations of both aerosol content and resulting climatic effects. The use of two different locations helps us to estimate the differences in the aerosol variations and their climatic consequences between the relatively clean and the industrially polluted regions. The correlation analysis and the multiple regression technique used in our study allow us to build statistical correlation models (1) to specify the main local aerosol sources

and their input into the variations of the local aerosol content; (2) to study aerosols' influence on some local climatic parameters. Concerning the data sources, the preference was given to directly measured data series that have sufficient quality, statistical homogeneity and temporal resolutions, and are available for the whole studied period. In some cases the available measured data series can be considered as well as proxies for those parameters which lack direct measurement during the studied period (such are the cases of SO_2 series used as a proxy for other anthropogenic pollutants or sunshine duration series used as a proxy for cloud amount).

The paper is organized as follows: Sect. 1 presents the modern state of the art and briefly gives an overview of the paper. Section 2 contains the description of the used data sets as well as their statistical properties. Section 3 describes variations of aerosol content over continental Portugal during 1978–1993 and their main sources. In Sect. 4 we show how aerosol variations affect local climate during the studied period. Section 5 gives a summary on the obtained results. Here in Sect. 2 we present only a short description of some data sets used in the study. The detailed analysis of these parameters can be found in the Supplement. Part 1 of the Supplement presents a detailed description of the aerosol data (Part 1.1), comparison of atmospheric parameters from different meteorological stations (Part 1.2) and description of the method used to detect dust events (Part 1.3). Part 2 of the Supplement presents the correlation analysis between the sunshine duration series and other meteorological parameters.

2 Data sets

2.1 Studied locations

We used the aerosol data over two locations of continental Portugal (see Fig. 1a) – the only available TOMS aerosol data for this region. The first one is the site ID 082 over Lisbon (38°46′ N, 9°8′ W; 105 m a.s.l.), the second one is the site ID 288 over Penhas Douradas (40°25′ N, 7°33′ W, 1380 m a.s.l.). In the first case the region around the site is one of the most urbanized and industrial sites in Portugal where the anthropogenic effects are expected to be strong. The second site corresponds to a less populated mountain region affected by the anthropogenic pollution in a lower degree but frequently exposed to forest fire smoke and dust events (Pereira et al., 2005; Pereira et al., 2008; Obregón et al., 2012). Hereafter we use a term "urban" for the site ID 082 and a term "rural" for the site ID 288.

Consequently, we used climatic data measured by two meteorological observatories that are close to the AI sites. The first data set belongs to the Geophysical Institute of University of Coimbra (hereafter, "IGUC series"). The second set belongs to the Geophysical Institute of Instituto Dom Luiz of University of Lisbon (hereafter, "IGIDL series"). Both loca-

tions are shown on the map in Fig. 1a (marked as "Coimbra" and "Lisbon", respectively).

2.2 Aerosol parameters

The TOMS aerosol index (AI) data (http://disc.sci.gsfc.nasa.gov/acdisc/TOMS) from 1 November 1978 to 6 May 1993 were used to study the variations of the aerosol content over Portugal. Under most conditions the AI is positive for the ultraviolet absorbing aerosols (pure absorption) and negative for the ultraviolet non-absorbing aerosols (pure scattering) when two close wavelengths in ultraviolet region near 360 nm are considered (see e.g., Ginoux and Torres, 2003, and http://visibleearth.nasa.gov/view.php?id= 1043). The TOMS aerosol index is calculated in a way that allowed us to separate days with prevailing absorbing (e.g., mineral dust, smoke, volcanic ash) or scattering (e.g., sea salt aerosols in the regions relatively close to ocean and sulfate aerosols in urban areas) particles (Herman et al., 1997; Torres et al., 1998). More details about the AI calculations can be found in the Supplement, Part 1.1. In this study we used daily data only in one case: to identify days with Saharan dust events (SDEs); see Sect. 2.4. For other purposes the monthly, seasonal and annual means of AI (and other parameters) have been calculated. For each of the sites and for each month we calculated three series: $\langle AI \rangle$, $\langle AIpos \rangle$ and $\langle AIneg \rangle$ taking into consideration all, only absorbing or only scattering aerosols, respectively. The seasonal and annual mean series were calculated using corresponding monthly mean series. In the paper these mean series are referenced as monthly, seasonal and annual series, correspondingly. Variations of these three indices for both sites are shown in Fig. 1b–d.

2.3 Atmospheric parameters

The sets of climatic parameters used in this study include monthly and annual means of the following daily variables:

1. minimum (T_{min}), maximum (T_{max}) and average (T_{aver}) temperatures, and daily temperature range (DTR);

2. accumulated precipitation amount (precip);

3. mean atmospheric pressure at station level (p);

4. sunshine duration (SshD).

The temperature and pressure series are part of the historical data set recently presented to the scientific community after the homogenization procedure done in the frame of the FP7 project ERA-CLIM (European Reanalysis of Global Climate Observations) (Morozova and Valente, 2012; Bližňák et al., 2014; Stickler et al., 2014). Other series are still under analysis. The relations between aerosol variations and atmospheric conditions in the region under consideration (see Sect. 4) were studied separately for two sites: the IGIDL series was used together with AI data from the urban site

(ID 082), and the IGUC series were used in pair with the AI series from the rural site (ID 288). The distance between the aerosol detection site and the meteorological station is about 5.5 km in case of the site ID 082 and about 74 km in case of the site ID 288. In the second case the distance between the places of measurement of the aerosols and climatic parameters is quite large. Nevertheless, we used the IGUC series because the other data sets available for this region are of insufficient quality and time resolution (see also a discussion in the Supplement, Part 1.2). The comparison of the IGUC and IGIDL series shows that the climatic conditions in Lisbon and Coimbra are quite similar (correlation coefficients in the range from 0.5 to 0.998 with low p values and meta p values) but not totally identical. Most important differences were found for the April and August series of the precipitation and DTR (correlation coefficients are lower than 0.5). A whole set of correlation coefficients between the IGUC and IGIDL series can be found in the Supplement, Part 1.2.

Please also note that all statistical significance (p values and meta p values) for correlation coefficients presented in this paper is calculated using 10 000 of the Monte-Carlo simulations with the random-phase Fast Fourier Transform as a randomizing procedure (Ebisuzaki, 1997). p value shows the probability for any specific correlation coefficient of the singular comparison to be obtained by chance. In cases when 12 separate monthly plus an annual series were analyzed simultaneously, the multiple comparisons significance (meta p values) was calculated as well.

2.4 Aerosol sources

Spatial and temporal distributions of aerosols as well as their origin are very variable. In this study we take into consideration only main sources responsible for the aerosol content variations over the continental Portuguese region. These sources are mineral dust from Sahara and Sahel regions, wildfires (Alves et al., 2011; Vicente et al., 2012, 2013; Evtyugina et al., 2013), anthropogenic pollution (Pereira et al., 2005; Pereira et al., 2008, 2009, 2011, 2012; Santos et al., 2008; Calvo et al., 2010) and volcanic aerosols. Some other locally important aerosol sources (like sea salt aerosols or anthropogenic aerosols other than SO_2) remain outside the frames of our study due to the absence of reliable (preferably measured) data on their variations for the studied period. Nevertheless, the regression models discussed in Sect. 3.2 (see also Table 2) show that even this limited set of aerosol sources allows us to reconstruct the aerosol content variations with a good accuracy.

2.4.1 Volcanoes

In this study we use the NASA Goddard Institute for Space Studies (GISS) climate simulation (http://data.giss.nasa.gov/modelforce/strataer) data on variability of the stratospheric aerosol optical thickness (AOT) at 550 nm for Northern

Hemisphere as a proxy for the volcanic aerosol content changes. This data set has monthly resolutions from October 1850 to December 2010. The data set is described in Sato et al. (1993) and Bourassa et al. (2012). During the studied period, four volcanic eruptions with the volcanic emissivity index VEI > 4 took places (numbered in Fig. 2c):

1. March 1980 – Mt. St. Helens, tropospheric eruption;

2. March–April 1982 – El Chichon, stratospheric eruption;

3. June and August 1991 – Pinatubo, stratospheric eruption;

4. August–October 1991 – Mt. Hudson, stratospheric eruption.

These eruptions, except the first one, deposited a significant load of sulfate aerosols to stratosphere over the globe. However, during the years following the eruptions the amount of absorbing particles in a zone around $40°$ N latitude did not increased as dramatically as in regions around the equator (see, e.g., Fig. 3 in Torres et al., 2002).

2.4.2 Saharan dust events

Saharan dust events are well-known sources of the dust in the Mediterranean region (Pereira et al., 2008; Obregón et al., 2012). The maximum number of the SDE in the western Mediterranean is observed in the summer period, especially in July–August (Moulin et al., 1998; Fig. 4 in Torres et al., 2002; Rogora et al., 2004; Fiol et al., 2005). These events are characterized by the high amount of the absorbing dust particles in the atmosphere coming from the Sahara and Sahel regions. In this work we identified SDE days using the method fully described in Barkan et al. (2005) and Varga et al. (2013). The main idea is to select days when standardized AI is higher than a threshold value (see Supplement for a brief description of the method and comparison to other published data. The short analysis of the dust events frequency for both AI sites is also presented in the Supplement, Part 1.3). The variations of the monthly mean ⟨AIpos⟩ index averaged over two Portuguese locations increase together with the total monthly number of dust events. ⟨AIneg⟩ variations on contrary have no relations to the SDEs.

2.4.3 Forest fires

Among all southern European countries, Portugal shows the highest density of wildfire ignitions (Catry et al., 2010). Almost all fires occur in summer months (from June to September) due to the dry and hot weather that is common for the region at this time of a year (Pereira et al., 2008; Obregón et al., 2012). Portuguese Institute for the Conservation of the Nature and Forests (Instituto da Conservação da Natureza e das Florestas, ICNF, http://www.icnf.pt) provides the data on the
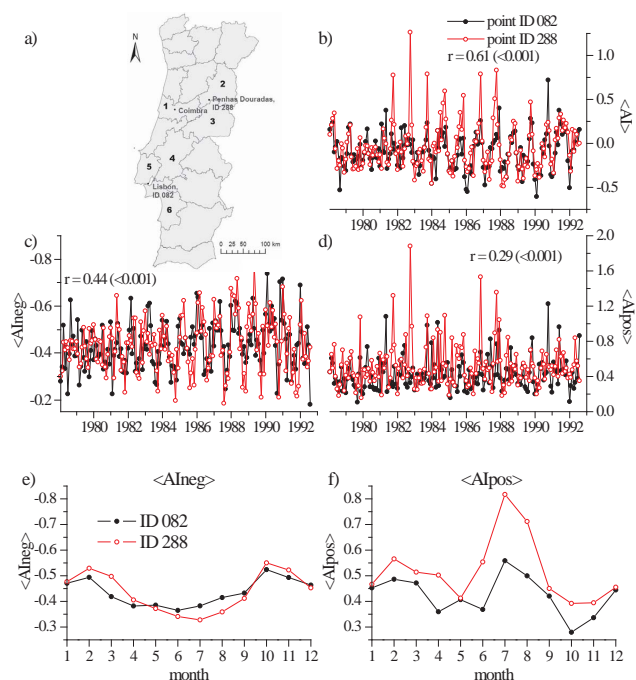

Figure 1. (a) Map of continental Portugal with the locations of the satellite AI observation points (ID 082 and ID 288) and the meteorological stations (Coimbra, Lisbon and Penhas Douradas). Districts used for calculation of the wildfire burned area: 1 – Coimbra, 2 – Guarda, 3 – Castelo Branco, 4 – Santarém, 5 – Lisboa, 6 – Setúbal. **(b–d)** Monthly mean ⟨AI⟩ **(b)**, ⟨AIneg⟩ **(c)** and ⟨AIpos⟩ **(d)** series for two satellite locations: ID 082 and ID 288. **(e–f)** Annual cycle of ⟨AIneg⟩ **(e)** and ⟨AIpos⟩ **(f)** for the sites ID 082 and ID 288 for the time period 1979–1992. Please note the inverted y axes in **(c)** and **(e)**. Correlation coefficients r for the AI series from different sites are shown with p values in brackets.

number of fire occurrences and a total burned area (BA), organized by districts, from 1980 to 2011. In this study we used only the BA data because the fire occurrence series seem to be less reliable (Pereira et al., 2005). The BA data from the Coimbra, Guarda and Castelo Branco districts (marked by numbers 1–3 on the map in Fig. 1a) were used to compare with the AI series from the rural site (ID 288), and the BA data from the Santarém, Lisboa and Setúbal districts (marked by numbers 4–6 on the map in Fig. 1a) were used to compare with the AI series from the urban site (ID 082).

2.4.4 Pollution

Anthropogenic aerosols affect the radiation balance in the atmosphere both through the absorption and the scattering processes (Wang, 2013). We assume that the actual measurements of the air composition give more precise information about the aerosol content than the estimated production of the anthropogenic sulfates and/or nitrates. Therefore, in this work we used the data from the European Monitoring and Evaluation Programme (EMEP) database (http://www.emep.

int), specifically the EBAS (European Monitoring and Evaluation Programme) database (http://ebas.nilu.no) which contains the monthly mean values of SO_2 (in µg S m^{-3}) in five Portuguese stations for the period from August 1979 to December 2009:

1. Bragança (41°49′ N, 6°46′ W; 690 m a.s.l.);

2. Viana do Castelo (41°42′ N, 8°48′ W; 16 m a.s.l.);

3. Monte Velho (38°05′ N, 8°48′ W; 43 m a.s.l.);

4. Fóia (37°19′ N, 8°54′ W; 902 m a.s.l.);

5. Faro (37°01′ N, 7°58′ W; 8 m a.s.l.).

Since the data series contain significant gaps (14 % of the whole data set length), and measurement time intervals are different for different stations, we applied linear interpolation to estimate the missing data and calculated a single mean series.

3 Variations of aerosol content and their sources

The ⟨AIpos⟩ and ⟨AIneg⟩ series (Fig. 1c–d) for the same sites do not correlate with each other: the correlations coefficients between the ⟨AIpos⟩ and ⟨AIneg⟩ monthly series are 0.14 (p value = 0.05) for the site ID 082 and 0.22 (p value < 0.001) for the site ID 288. The spatial correlation for all the types of aerosol indices is more or less strong, which is quite expected for the sites at a distance of about 200 km apart (see Fig. 1b–d). The correlation coefficients for the separate monthly and annual series are shown the Supplement. The analysis of the standard statistical parameters of the ⟨AIpos⟩ and ⟨AIneg⟩ series shows that the absorbing aerosols play a more significant role over the rural site (ID 288) than over the urban site (ID 082). The ⟨AIpos⟩ series has mean, standard deviation and maximum values that are higher in the case of site ID 288 than in case of the site ID 082; on contrary, the same statistical parameters for the ⟨AIneg⟩ series are practically equal for the two sites.

The AI series for both sites show annual cycle mainly due to the well-established seasonal changes of the ⟨AIneg⟩ (see Fig. 1e) – more scattering aerosols are seen from October to March due to the seasonal cycles of nitrate aerosols (see, e.g., Calvo et al., 2013) and/or other anthropogenic pollutants. During the autumn–winter cold period there is an additional input of soot from the domestic heating and, probably, an increase of the local traffic due to the rainy weather conditions (Pereira et al., 2012; Querol et al., 1998). The ⟨AIpos⟩ shows a tendency to bimodal seasonal variations having higher values in July–August with a second (lower) maximum in February–March (Fig. 1f). This bimodality is in agreement with the in situ measurements made in Évora, Portugal (38.5° N, 7.9° W; 300 m a.s.l.) during the time period 2002–2008 (Pereira et al., 2008, 2011). The summer peak is
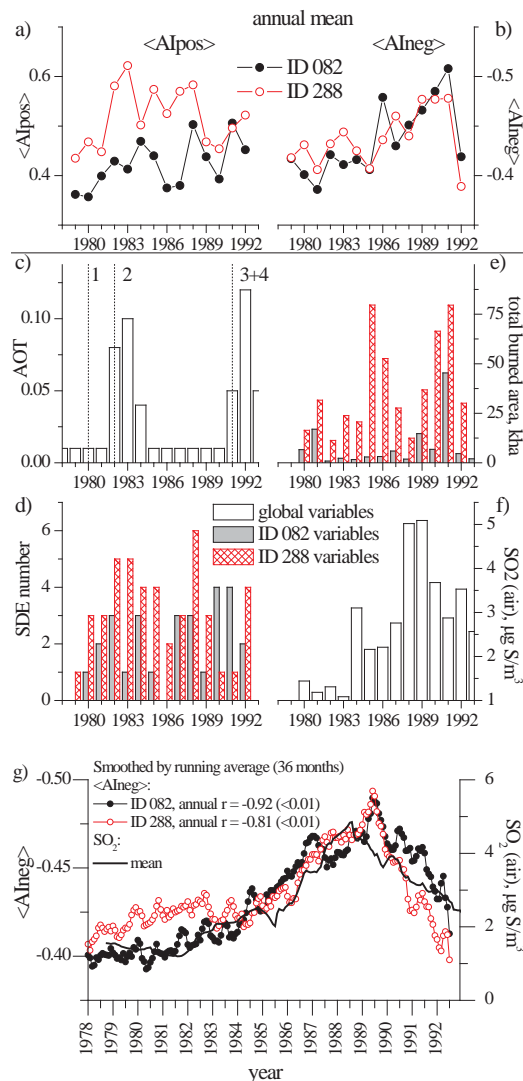

Figure 2. (a–b) Annual variations of aerosol indices ⟨AIpos⟩ **(a)** and ⟨AIneg⟩ **(b)** for two locations: ID 082 and ID 288. **(c–f)** Annual values of parameters describing aerosol forcings: volcanic aerosols (**c** – AOT, annual means), Saharan dust events (**d** – annual sums for two AI locations), wildfires (**e** – total annual burned area close to AI locations) and anthropogenic sulfates (**f** – SO_2, annual means). The four most significant volcanic eruptions are marked in *c* by vertical lines: 1 – Mt. St. Helens, 2 – El Chichon, 3 – Pinatubo and 4 – Mt. Hudson. **(g)** Monthly variations of SO_2 and aerosol indices ⟨AIneg⟩ for two sites (ID 082 and ID 288) smoothed by the 36-month running averaging. Correlation coefficients (*r*) are between the AI and the sulfate series (*p* values are shown in brackets). Please note the inverted *y* axes in **(b)** and **(g)** (left).

related to wildfire smoke and intensive SDE events, and the winter maximum is mostly due to the combined effect of local traffic and increased emissions from heating sources.

Table 1. Correlation coefficients between the variations of the AI and different forcing parameters: *annual* series in case of volcanic and pollution forcings, and *summer* series in case of SDE and wildfire forcings. Values in brackets are *p* values (only *p* values ≤ 0.2 are shown).

Forcing	Sites	AI series		
		$\langle AI \rangle$	$\langle AIpos \rangle$	$\langle AIneg \rangle$
Volcanic	ID 082	0.66 (< 0.01)	0.31	0.15
	ID 288	0.62 (< 0.01)	0.43 (< 0.01)	0.27 (0.03)
SDE	ID 082	0.53 (0.05)	0.75 (< 0.01)	-0.11
	ID 288	0.55 (0.05)	0.80 (< 0.01)	< 0.1
Wildfires	ID 082	0.2	0.31	-0.44 (0.12)
	ID 288	< 0.1	-0.39 (0.19)	0.27
Pollution	ID 082	< 0.1	0.51 (0.08)	-0.53 (0.06)
	ID 288	< 0.1	-0.17	-0.37 (0.20)

3.1 Main aerosol sources

3.1.1 Volcanoes

The annual variations of the $\langle AIpos \rangle$ measured over two Portuguese sites (Fig. 2a) show some increase after the eruptions of 1982 and 1991 (Fig. 2c), but these peaks could also be related (at least, partly) to other phenomena such as, e.g., Saharan dust events. On the other hand the correlation analysis (see Table 1) shows (1) a weak but statistically significant dependence of the annual series of the $\langle AI \rangle$ and $\langle AIpos \rangle$ on the AOT variations and (2) no dependence between the $\langle AIneg \rangle$ and AOT annual series.

3.1.2 Saharan dust events (SDE)

Since most of the SDEs take place in summer, we compared not only annual values of aerosol indices and SDE number but also values calculated for the local summer season (from June to September). The correlation coefficients presented in Table 1 as well as comparison of Fig. 2a–b and d clearly show that the high values of the $\langle AIpos \rangle$ in 1982–1983 (at least partly) and in 1988 are caused by the intensive Saharan dust intrusions. The $\langle AIneg \rangle$ series shows no connection to the SDEs, as it has to be expected.

3.1.3 Forest fires

Figure 2e shows variations of the total burned area for both groups of districts. The correlation coefficients between the $\langle AI \rangle$ and BA series (Table 1) are quite low. The reason, probably, is the stronger effect of other forcings (like SDEs) on the aerosol content variations. However, the multiple regression models, which will be discussed later in Sect. 3.2, detect the "forest fire" forcing as a regressor required to explain the AI series variations.

3.1.4 Pollution

The annual values of the SO_2 content are shown in Fig. 2f. As one can see, there is a strong dependence between the variations of the $\langle AIneg \rangle$ (shown in Fig. 2b) and the SO_2 content. The anti-correlation (correlation coefficient $r = -0.53$, p value $= 0.06$) between the curves reflects the increase of the scattering particles in the atmosphere (lower $\langle AIneg \rangle$ values) coinciding with the growth of the measured SO_2 concentration. Unsurprisingly, the $\langle AIneg \rangle$ variations over a highly populated location (ID 082 – Lisbon) show stronger dependence on the SO_2 content (see Table 1). The correlations between the $\langle AIneg \rangle$ and SO_2 variations became more stronger when trends of these two parameters were studied. For example, the comparison of the monthly series of the SO_2 and the $\langle AIneg \rangle$ smoothed by the running averaging procedure (window of 36 months) shows that the satellite measured $\langle AIneg \rangle$ series follows the ground measured sulfate content data with probably a lag of about 5–10 months; see Fig. 2g.

Relatively high correlations between the $\langle AIpos \rangle$ over the urban site (ID 082) and the SO_2 values (Table 1) are probably caused by the similarities in the variations of different pollution gases/aerosols. Since the anthropogenic sulfates are almost totally scattering aerosols, they cannot affect the satellite-measured $\langle AIpos \rangle$ values. However, the amount of other types of aerosols (like light absorbing black carbon) can follow the changes of the SO_2 content due to the same source of origin (e.g., fossil fuel combustion). Unfortunately, we found no measurements of other anthropogenic aerosols/gases for the studied period with an acceptable time resolution and data quality to confirm this suggestion. On the other side, since the pollutants of different types originate from the same sources (such as traffic, coal and biomass burning, industrial activities; see, e.g., Calvo et al., 2013) their temporal variations are more or less similar, and the SO_2 series can be considered in the frame of our study and to a certain degree as a proxy for most of anthropogenic pollutants.

3.2 Multiple regression models of aerosol variations

The analysis of the individual correlations between the AI and a number of natural and anthropogenic aerosol forcings allowed us to find the main sources of the aerosol content variations for this region. Those forcings are the Saharan dust events, the wildfires, the anthropogenic pollution and the volcanic eruptions. Some of these forcings affect both the absorbing and the scattering aerosols (e.g., anthropogenic pollution and forest fires). Other forcings influence only the absorbing part of the aerosol content (e.g., SDE). Linear multiple regression models (MRM) have been constructed to statistically connect the observed variations of the $\langle AI \rangle$, $\langle AIpos \rangle$ and $\langle AIneg \rangle$ due to the changes of the abovementioned forcings.

The models were constructed using a "best subset" technique that finds a subset of regressors (aerosol forcings, in our case) that predicts as much of the variations of the dependent parameter (AI, in our case) as possible. The quality of the MRM is defined by r and $r^2_{\mathrm{adj.}}$ parameters. The first one is a correlation coefficient between the modeled and the original series, and its square multiplied by 100 defines the percent of explained variations. The second parameter is the so-called "adjusted r^2". The adjustment is done using differences between the model and the original data compared to the original data variance and taking into account the number of degrees of freedom. The $r^2_{\mathrm{adj.}}$ was used as a criterion to compare the MRMs with different subsets of regressors: the subset that gives a bigger $r^2_{\mathrm{adj.}}$ value is the "best subset". The role of each of the regressors is estimated by a β coefficient that quantifies how strongly each regressor influences the dependent variable. The β is measured in units of standard deviation σ: the regressors with the highest (absolute) β values have greater impact on the dependent parameter. All parameters for the different MRMs are shown in Table 2 for the annual and summer (June–September) AI series. The MRMs for the annual and summer AI series together with the corresponding original AI data are also presented in Fig. 3. All abovementioned forcings are used as regressors for the $\langle AIpos \rangle$ and $\langle AI \rangle$ series, and only the wildfires and the pollutions are used to model the $\langle AIneg \rangle$ variations.

The obtained results prove that chosen forcings represent a good set of regressors to explain the $\langle AIpos \rangle$ variations (Fig. 3b, e, h, k). The correlation coefficients between the MRMs and the original AI series are greater than 0.7 (see fourth column of Table 2), and the models explain (taking into account the number of degrees of freedom) at least 35 % of the variations of the original $\langle AIpos \rangle$ series (see sixth column of Table 2). Unsurprisingly, Saharan dust events have the greatest contribution (eighth column of Table 2) to the summer $\langle AIpos \rangle$ variations for both locations and the SO_2 content is an important regressor for the $\langle AIpos \rangle$ series measured above the more populated area (ID 082); see tenth column of Table 2.
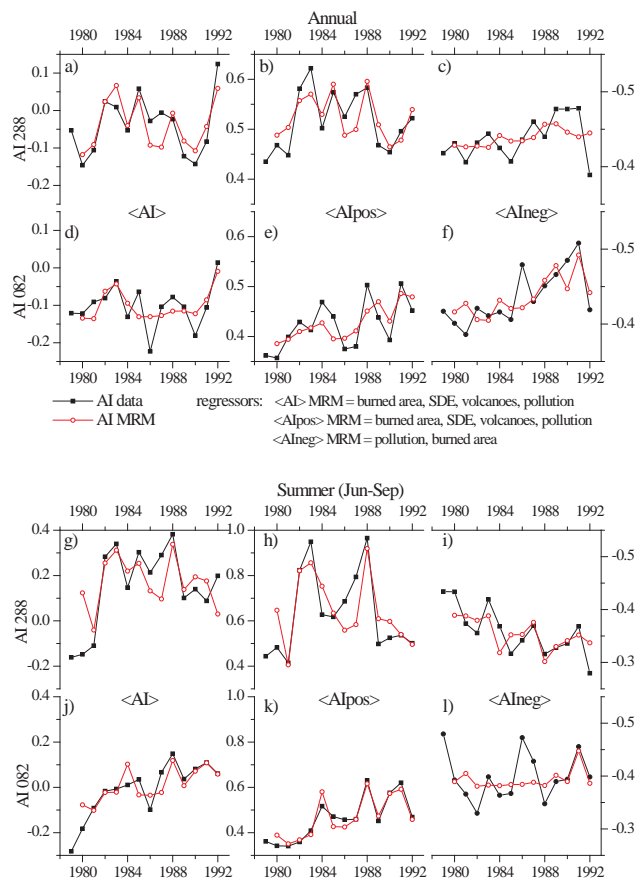

Figure 3. Multiple regression models (MRM) of the annual **(a–f)** and summer (June–September; **g–l)** AI series: **(a, d, g, j)** $\langle AI \rangle$ series; **(b, e, h, k)** $\langle AIpos \rangle$ series; **(c, f, i, l)** $\langle AIneg \rangle$ series (please note the inverted y axis). MRMs are calculated for two locations: ID 288 **(a–c** and **g–i)** and ID 082 **(d–f** and **j–l)**. The sets of regressors are shown for MRMs of $\langle AI \rangle$, $\langle AIpos \rangle$ and $\langle AIneg \rangle$.

Concerning the $\langle AIneg \rangle$ series (Fig. 3c, f, i, l), it is clear that the used regressors cannot sufficiently explain observed variations of the scattering aerosols, especially when the number of degrees of freedom is taken into account. However, the MRMs for the $\langle AIneg \rangle$ are similar to the original series trends: these trends follow the growth of the pollutant (SO_2 content in our case). The discrepancies between the models and the observations can result from the absence in the list of the MRM regressors of some important aerosol sources such as, for example, sea salts or sources other than SO_2 pollutants (e.g., NO_x). Unfortunately, there is no reliable corresponding data series that can be used in the frame of our study.

Finally, the MRMs for the $\langle AI \rangle$ series (Fig. 3a, d, g, j) are well correlated with the original series for both locations and explain 33–55 % of the $\langle AI \rangle$ variations. These results show that the SDEs, the wildfires and possibly the volcanic eruptions significantly affect the aerosol content over the low-populated location (ID 288), while the anthropogenic pollu-

Table 2. Parameters of multiple regression models of AI *annual* and *summer* series (see Sect. 3.2 for parameter descriptions). The correlation coefficients r greater than 0.67 and $r^2_{\text{adj.}}$ equal or greater than 0.45 are in bold, and "×" marks the parameters that were excluded from a particular "best subset".

Time period	AI type	Site ID	r	p value	$r^2_{\text{adj.}}$	β			
						Volcanic AOT	SDE number	Area burned by wildfires	SO_2 content
Annual	⟨AI⟩	082	**0.67**	0.014	0.34	0.69	×	×	0.12
		288	**0.81**	< 0.01	**0.55**	0.5	0.73	0.53	×
	⟨AIpos⟩	082	**0.72**	< 0.01	0.35	0.41	×	0.34	0.57
		288	**0.76**	< 0.01	**0.49**	×	0.98	0.43	×
	⟨AIneg⟩	082	**0.71**	< 0.01	0.40			−0.47	−0.5
		288	0.37	0.21	0.06			×	−0.37
Summer (Jun–Sep)	⟨AI⟩	082	**0.79**	< 0.01	**0.50**	0.28	0.41	×	0.59
		288	**0.67**	0.012	0.33	×	0.97	0.68	×
	⟨AIpos⟩	082	**0.95**	< 0.01	**0.88**	×	0.57	×	0.63
		288	**0.82**	< 0.01	**0.60**	×	1.05	0.37	×
	⟨AIneg⟩	082	0.44	0.12	0.12			−0.44	×
		288	**0.68**	0.012	0.41			×	0.68

tion plays an important role in the variations of the AI over the high-populated location (ID 082).

4 Regional climate variations in relation to aerosol content changes

Here we present the analysis of the relations between the aerosol content and the atmospheric parameters described in Sect. 2. The analysis was done separately for two locations. The analysis of the climatic conditions between the Lisbon and Coimbra (see the Supplement, Part 1.2) showed their strong similarity. This similarity results from the relatively short distance between these locations and their proximity to the ocean. On the other hand, the measured AI monthly means, as was discussed in Sect. 3, are different for these two sites. In our opinion, there are two main reasons for these differences. First of all, the Lisbon area is much more polluted than the region around the rural site (ID 288). Secondly, the more northeastern position of the rural site ID 288 provides the stronger and more frequent effect of the dust intrusions.

4.1 Rural site ID 288

The results of correlation analysis for the pair "AI ID 288 vs IGUC series" are shown in Fig. 4b, d, f. As one can see, the relations between the climatic parameters and the aerosols of absorbing (⟨AIpos⟩) and scattering (⟨AIneg⟩) types strongly depend on the season. The effect of the absorbing aerosols is more prominent during the summer–autumn (Fig. 4d), but the relation between the climatic parameters and the scat-

tering aerosols are stronger during the first half of a year (Fig. 4f). It has to be mentioned that the summer–early autumn period of a year is the dry season in continental Portugal, whereas late autumn, winter and spring seasons often are wet because of the influence of the North Atlantic cyclones (Miranda et al., 2002).

First we examine relations between the ⟨AIpos⟩ and the climatic parameters. One of the most important features is the anti-correlation between the ⟨AIpos⟩ and parameters such as the SshD, T_{max} and DTR during July. Similar relations also take places in June, August and October–November but the magnitude and the statistical significance of correlation coefficients r are smaller. This cooling trend coincides with epochs of frequent SDE events (high ⟨AIpos⟩ values) and is in agreement both with most recent/precise measurement (Santos et al., 2008) and with modeling studies (Santos et al., 2013). In September the correlation coefficients have an opposite sign and are statistically insignificant. As it is shown in Fig. 4d, the increase of the absorbing aerosol amount is accompanied by the decrease of the sunshine duration. The decrease of the SshD leads to the decrease of the amount of the solar radiation reaching the ground, which in turn affects T_{max} (a parameter that can be considered as a measure of the daytime temperature): T_{max} is decreasing. Consequently, the decrease of the T_{max} affects the daily temperature ranges: the DTR decreases also.

The relations between the temperatures over the Iberian Peninsula and SshD during the second half of the 20th century were reported earlier (see del Rio et al., 2012, and references therein). They were attributed, mainly, to the variations
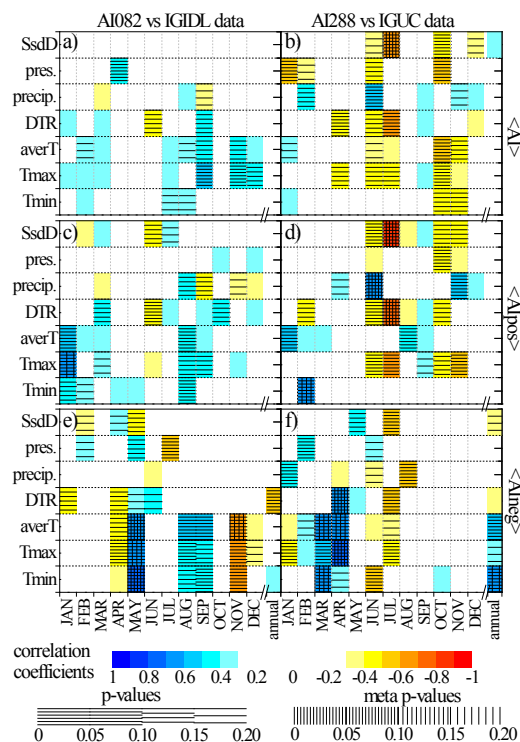
Figure 4. Correlation coefficients (r) between the AI indices ($\langle AI \rangle$ – **a, b**; $\langle AIpos \rangle$ – **c, d**; $\langle AIneg \rangle$ – **e, f**) for sites ID 082 (**a, c, e**) and ID 288 (**b, d, f**) and atmospheric parameters measured in Lisbon (IGIDL series – **a, c, e**) and Coimbra (IGUC series – **b, d, f**). Only correlation coefficients $|r| \geq 0.3$ are shown. The statistical significance for singular (p values) and multiple (meta p values) comparisons are shown by shading. All correlation coefficients are calculated in a way that in case of simultaneous increase/decrease of the amount of any type of aerosols (changes of an absolute value of any AI index) and an atmospheric parameter value the correlation coefficient is positive.

of the circulation patterns over the North Atlantic and consequent changes in the cloudiness. However, accordingly to the data of our analysis, the variations of the SshD can also result from the strong dust intrusions.

This kind of relation between the SshD and temperatures is usually related to the cloud effect on the radiation distribution in the lower atmosphere (IPCC, 2007, 2013, Ch. 7). The T_{max} and DTR correlate very well with the SshD during almost a whole year (the whole set of correlation coefficients can be found in the Supplement, Part 2). The precipitation amount anti-correlates with SshD; therefore, this can be considered as a confirmation of the existence of the clouds that block solar irradiance. However, there is a possibility that such relations between the SshD and $\langle AIpos \rangle$ during the dry summer period, at least partly, are due to the direct aerosol effect. We assume that the change of the radiation balance is also the reason for the correlation between the amount of the absorbing aerosols and the T_{min} (a parameter that can be considered as a measure of the nighttime temperature) found for

the February series. The aerosol particles may play a role in high-level clouds reflecting some of the outgoing infrared radiation back to the ground. The relation between the precipitation amount and the $\langle AIpos \rangle$ found for this site also reflects a process that can be identified as an indirect effect of the aerosols on cloud formation (see, e.g., IPCC, 2007, Ch. 7.5 "Aerosol Particles and the Climate System"). The increase of the $\langle AIpos \rangle$ coincides with the higher amount of the precipitation (Fig. 4d). This effect is more pronounced in April, June and November–December. The aerosol particles may act as seeds for the cloud droplets in relatively dry summer (and sometimes winter) air.

The effect of the scattering aerosols (described by the $\langle AIneg \rangle$ index) on some of the atmospheric parameters in the region is similar to the effect observed for the $\langle AIpos \rangle$. The increase of the aerosol loading coincides with the decrease of the SshD, DTR, T_{max} and T_{aver} in July (and in a weaker form in June and August); see Fig. 4f. The opposite relations take place in early spring (February–April) when $\langle AIneg \rangle$ variations correlate with changes in the T_{min}, T_{max} and T_{aver} (and DTR in April). On the other hand, the precipitation has an opposite dependence on the $\langle AIneg \rangle$ variation compared with those obtained for the $\langle AIpos \rangle$. The precipitation amount decreases when the scattering aerosol loading in the atmosphere increases. This effect can be related to decrease of the cloud droplet size in the polluted air which increases the cloud lifetime and decrease precipitation (see, e.g., IPCC, 2007, Ch. 7.5 in Climate Change 2007). The only exception is January: during this month the $\langle AIneg \rangle$ is correlated with the precipitation.

4.2 Urban site ID 082

The results of correlation analysis for the pair "AI ID 082 vs. IGIDL series" are shown in Fig. 4a, c, e. As one can see there are significant differences in the relation between the variations of the AI and climatic parameters over Lisbon comparing to the Coimbra–Penhas Douradas region. The differences between these two sites can result from different aerosol sources in the more polluted Lisbon area. Furthermore, the Coimbra–Penhas Douradas area is strongly affected by the absorbing aerosols originated from wildfires and Saharan dust. As mentioned in Choobari et al. (2014), the combination of dust and soot particles increase absorption properties of the aerosols, whereas the combination of dust and sulfates from pollution decreases absorption properties.

First of all, the biggest correlation coefficients are obtained for the $\langle AIneg \rangle$ but not for the $\langle AIpos \rangle$ as was found for the other location. As one can see from the comparison of the Fig. 4a, c, e and Fig. 4b, d, f, the similarities in the relations between the AI indices and the climatic parameters exist, mostly, for temperature parameters and $\langle AIpos \rangle$ during January–March, June and August–September periods. The significant seasonal differences are seen only in the
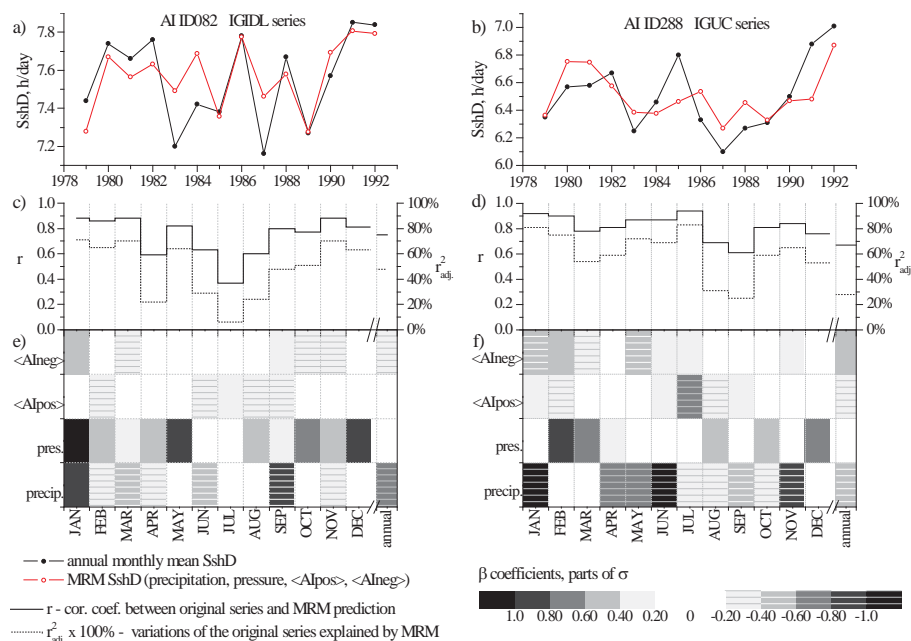
Figure 5. Multiple regression model (MRM) predictions and parameters for the annual and monthly sunshine duration from the IGIDL **(a, c, e)** and IGUC **(b, d, f)** series. **(a–b)** Measured annual SshD series vs. the corresponding MRM predictions. **(c–d)** Correlation coefficients r between the MRM and the original series for the monthly data, and the variance explained by the MRMs ($r^2_{\mathrm{adj.}}$ in per cent). **(e–f)** β coefficients (in parts of standard deviation σ) for each of the regressors for the monthly MRMs.

⟨AIneg⟩ variations. All temperature parameters tend to anti-correlate with the amount of ⟨AIneg⟩ in April and November (months of the transient seasons). On the contrary, for the months of May, August and September (hot dry season) there is a tendency towards correlation between the ⟨AIneg⟩ values and the temperatures. As a rule, temperature parameters tend to correlate with the AI (the more aerosol particles of both types, the higher the temperature) with just a number of exceptions (June for ⟨AIpos⟩, and April and November for ⟨AIneg⟩). The relations between the SshD and aerosol content are weak and sporadic. There is just a small number of significant anti-correlations between the ⟨AIpos⟩ and ⟨AIneg⟩ and the SshD series. These are June series for ⟨AIpos⟩ (Fig. 4c) and May series for ⟨AIneg⟩ (Fig. 4e). In the first case the relations are similar to ones obtain for the Coimbra–Penhas Douradas site: the increased amount of the aerosol loading coincides with shorter periods of sunshine duration.

4.3 Multiple regression models of sunshine duration variations

To study further the role played by the aerosols in the climatic variations of this region, we constructed multiple regression models that explain sunshine duration variations depending on the following parameters: precipitation and pressure (proxies for the cloud amount/clear sky conditions), and ⟨AIpos⟩ and ⟨AIneg⟩. The choice of the parameters is defined

by their high correlation coefficients with the SshD series (see, e.g., Fig. 4c–f and the Supplement, Part 2). The MRMs are calculated separately for both sites for the monthly and annual means using the "best subset" technique and parameters described in Sect. 3.2. The results are shown in Fig. 5a–f.

Altogether, the selected regressors allowed us to construct quite good regression models for the SshD series. Figure 5a and b show examples of the MRM predictions (for the annual SshD series for IGIDL and IGUC, correspondingly). The correlation coefficients between the MRMs and the measured data are higher than 0.6 for the IGUC SshD series and higher than 0.4 for the IGIDL SshD series (see Fig. 5d and c, correspondingly). The explained variance taking into account the number of degrees of freedom ($r^2_{\mathrm{adj.}}$) for the IGUC series changes from 25–31 % in August–September to 70–83 % in January–February and May–July; and for the IGIDL series from 14–25 % in August–September to 70–82 % in January and May–July. Overall, the MRMs for the IGUC series have better prediction quality than for the IGIDL series.

The role played by each of the regressors is shown in Fig. 5e–f using the β coefficients. As expected, the precipitation and the pressure series are included in the MRMs for almost all of the months throughout a year. The highest β coefficients (in absolute values) are mostly for the wet season (autumn-to-spring). The AI series are included in the "best subset" of regressors for many of the monthly (and annual) series but with quite low β values. The exceptions are dry summer months between June and September. For these

MRMs the ⟨AIpos⟩ is an important regressor (see Fig. 5f). On contrary, the ⟨AIneg⟩ series are more often included as the regressors into the MRMs for the wet autumn-to-spring season. Thus, the results of the regression analysis confirm the importance of the aerosol loading to explain observed climatic variations.

5 Conclusions

The results of the presented analysis show that the aerosol sources chosen in this study (volcanic aerosols, Saharan dust events (SDE), wildfires and anthropogenic pollution) play an important role in the aerosol content variations over two Portuguese locations (an urban region around Lisbon and a less populated mountain region). Unfortunately, it is impossible to fully separate the effect of the volcanic eruptions, the wildfires and the SDEs. Nevertheless, the regression analysis confirms the relations between the periods of high aerosol content and the periods of more frequently observed wildfires and SDEs. The anthropogenic pollutants also found to affect local aerosol content, especially in the urban region around Lisbon. It was also found that aerosol series averaged over 4 summer months (from June to September) have stronger relation with the SDEs and wildfires than monthly or annual data. Our results confirm the data from previous studies showing the important role of the anthropogenic pollution, wildfires and SDEs as drivers of the aerosol variation over continental Portugal.

The variations of aerosol content were found to be in relations with the changes of atmospheric parameters (temperatures, atmospheric pressure, precipitation amount and sunshine durations). These relations depend on the parameters in questions and change throughout the year. The strongest effect is found for the less urbanized and industrial mountain site. The most significant (both in amplitude and statistically) results were found for the relations between the maximum daily temperature (T_{max}) and daily temperature range (DTR), and absorbing aerosol content during summer months. These temperature and aerosols variations are also in an agreement with sunshine duration changes. The increase of the content of the absorbing aerosols coincides with the decrease of sunshine duration and, consequently, with the decrease of the T_{max} and DTR. This can be related both to the direct (cooling due to the decrease of the solar radiation flux) and indirect (higher cloudiness amount) effect. The response of the atmospheric parameters to aerosol variations is found to be weaker for the more urbanized region.

The Supplement related to this article is available online at doi:10.5194/acp-15-6407-2015-supplement.

References

Alves, C., Vicente, A., Nunes, T., Gonçalves, C., Fernandes A. P., Mirante, F., Tarelho, L., Sanchez de la Campa, A., Querol, X., Caseiro, A., Monteiro, C., Evtyugina, M., and Pio, C.: Summer 2009 wildfires in Portugal: emission of trace gases and aerosol composition, Atmos. Environ., 45, 641–649, 2011.

Barkan, J., Alpert, P., Kutiel, H., and Kishcha, P.: Synoptics of dust transportation days from Africa toward Italy and central Europe, J. Geophys. Res.-Atmos., 110, D07208, doi:10.1029/2004JD005222, 2005.

Bližňák, V., Valente, M. A., and Bethke, J.: Homogenization of time series from Portugal and its former colonies for the period from the late 19th to the early 21st century, Int. J. Climatol., doi:10.1002/joc.4151, online first, 2014.

Bourassa, A. E., Robock, A., Randel, W. J., Deshler, T., Rieger, L. A., Lloyd, N. D., Llewellyn, E. J., and Degenstein, D. A.: Large volcanic aerosol load in the stratosphere linked to Asian monsoon transport, Science, 337, 78–81, 2012.

Calvo, A. I., Pont, V., Castro, A., Mallet, M., Palencia, C., Roger, J. C., Dubuisson, P., and Fraile, R.: Radiative forcing of haze during a forest fire in Spain, J. Geophys. Res.-Atmos., 115, D08206, doi:10.1029/2009JD012172, 2010.

Calvo, A. I., Alves, C., Castro, A., Pont, V., Vicente, A. M., and Fraile, R.: Research on aerosol sources and chemical composition: Past, current and emerging issues, Atmos. Res., 120, 1–28, doi:10.1016/j.atmosres.2012.09.021, 2013.

Catry, F. X., Rego, F. C., Bação, F. L., and Moreira, F.: Modeling and mapping wildfire ignition risk in Portugal, Int. J. Wildland Fire, 18, 921–931, doi:10.1071/WF07123, 2010.

Choobari, O. A., Zawar-Reza, P., and Sturman, A.: The global distribution of mineral dust and its impacts on the climate system: A review, Atmos. Res., 138, 152–165, 2014.

del Río, S., Cano-Ortiz, A., Herrero, L., and Penas, A.: Recent trends in mean maximum and minimum air temperatures over Spain (1961–2006), Theor. Appl. Climatol., 109, 605–626, doi:10.1007/s00704-012-0593-2, 2012.

Ebisuzaki, W.: A method to estimate the statistical significance of a correlation when the data are serially correlated, J. Climate, 10, 2147–2153, 1997.

Evtyugina, M., Calvo, A., Nunes, T., Alves, C., Fernandes, P., Tarelho, L., Vicente, A., and Pio, C.: VOC emissions of smouldering combustion from Mediterranean wildfires in central Portugal, Atmos. Environ., 64, 339–348, 2013.

Fiol, L. A., Fornós, J. J., Gelabert, B., and Guijarro, J. A:. Dust rains in Mallorca (Western Mediterranean): Their occurrence and role in some recent geological processes, Catena, 63, 64–84, 2005.

Ginoux, P. and Torres, O.: Empirical TOMS index for dust aerosol: Applications to model validation and source characterization, J. Geophys. Res.-Atmos. 108, 4534, doi:10.1029/2003JD003470, 2003.

Herman, J. R., Bhartia, P. K., Torres, O., Hsu, C., Seftor, C., and Celarier, E.: Global distribution of UV-absorbing aerosols from Nimbus 7/TOMS data, J. Geophys. Res., 102, 16911–16922, 1997.

IPCC: Climate Change 2007 – The physical science basis: Working Group I Contribution to the Fourth Assessment Report of the IPCC (Vol. 4), in: IPCC Fourth Assessment Report: Climate Change 2007 (AR4), Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M., and Miller, H. L., Cambridge University Press, Cambridge, UK and New York, NY, USA, 2007.

IPCC: Climate Change 2013 – The Physical Science Basis: Working Group I Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, IPCC, Cambridge University Press, 2013.

Miranda, P., Coelho, F. E. S., Tomé, A. R., Valente, M. A., Carvalho, A., Pires, C., Pires, H. O., Pires, V. C., and Ramalho, C.: 20th century Portuguese climate and climate scenarios, in: Climate Change in Portugal. Scenarios, Impacts and Adaptation Measures – SIAM Project, edited by: Santos, F. D., Forbes, K., and Moita, R., Gradiva Publishers, Lisbon, 23–83, 2002.

Morozova, A. L. and Valente, M. A.: Homogenization of Portuguese long-term temperature data series: Lisbon, Coimbra and Porto, Earth Syst. Sci. Data, 4, 187–213, doi:10.5194/essd-4-187-2012, 2012.

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.: Satellite climatology of African dust transport in the Mediterranean atmosphere, J. Geophys. Res.-Atmos., 103, 13137–13144, 1998.

Obregón, M. A., Pereira, S., Wagner, F., Serrano, A., Cancillo, M. L., and Silva, A. M.: Regional differences of column aerosol parameters in western Iberian Peninsula, Atmos. Environ., 62, 208–219, 2012.

Pereira, M. G., Trigo, R. M., da Camara, C. C., Pereira, J. M. C., and Leite, S. M.: Synoptic patterns associated with large summer forest fires in Portugal, Agr. Forest Meteorol., 129, 11–25, doi:10.1016/j.agrformet.2004.12.007, 2005.

Pereira, S., Wagner, F., and Silva, A. M.: Scattering properties and mass concentration of local and long-range transported aerosols over the South Western Iberia Peninsula, Atmos. Environ., 42, 7623–7631, 2008.

Pereira, S. N., Wagner, F., and Silva, A. M.: Continuous measurements of near surface aerosols in the south-western European (Portugal) region in 2006–2008, Adv. Sci. Res., 3, 1–4, 2009.

Pereira, S. N., Wagner, F., and Silva, A. M.: Seven years of measurements of aerosol scattering properties, near the surface, in the southwestern Iberia Peninsula, Atmos. Chem. Phys., 11, 17–29, doi:10.5194/acp-11-17-2011, 2011.

Pereira, S. N., Wagner, F., and Silva, A.M.: Long term black carbon measurements in the southwestern Iberia Peninsula, Atmos. Environ., 57, 63–71, 2012.

Querol, X., Alastuey, A., Puicercus, J. A., Mantilla, E., Miro, J. V., Lopez-Soler, A., Plana, F., and Artiñano, B.: Seasonal evolution of suspended particles around a large coal-fired power station. Particulate levels and sources, Atmos. Environ., 32, 1963–1978, 1998.

Rogora, M., Mosello, R., and Marchetto, A.: Long-term trends in the chemistry of atmospheric deposition in Northwestern Italy: the role of increasing Saharan dust deposition, Tellus B, 56, 426–434, 2004.

Santos, D., Costa, M. J., and Silva, A. M.: Direct SW aerosol radiative forcing over Portugal, Atmos. Chem. Phys., 8, 5771–5786, doi:10.5194/acp-8-5771-2008, 2008.

Santos, D., Costa, M. J., Silva, A. M., and Salgado, R.: Modeling Saharan desert dust radiative effects on clouds, Atmos. Res., 127, 178–194, 2013.

Sato, M., Hansen, J. E., McCormick, M. P., and Pollack, J. B.: Stratospheric aerosol optical depth, 1850–1990, J. Geophys. Res., 98, 22987–22994, 1993.

Stickler, A., Brönnimann, S., Valente, M. A., Bethke, J., Sterin, A., Jourdain, S., Roucaute, E., Vasquez, M. V., Reyes, D. A., Allan, R., and Dee, D.: ERA-CLIM: historical surface and upper-air data for future reanalyses, B. Am. Meteorol. Soc., 95, 1419–1430, doi:10.1175/BAMS-D-13-00147.1, 2014.

Torres, O., Bhartia, P. K., Herman, J. R., Ahmad, Z., and Gleason, J.: Derivation of aerosol properties from satellite measurements of backscattered ultraviolet radiation: Theoretical basis, J. Geophys. Res., 103, 17099–17110, 1998.

Torres, O., Bhartia, P. K., Herman, J. R., Sinyuk, A., Ginoux, P., and Holben, B. A.: Long-Term Record of Aerosol Optical Depth from TOMS Observations and Comparison to AERONET Measurements, J. Geophys. Res., 59, 398–413, 2002.

Varga, G., Kovács, J., and Újvári, G.: Analysis of Saharan dust intrusions into the Carpathian Basin (Central Europe) over the period of 1979–2011, Global Planet. Change, 100, 333–342, 2013.

Vicente, A., Alves, C., Monteiro, C., Nunes, T., Mirante, F., Cerqueira, M., Calvo, A., and Pio, C.: Organic speciation of aerosols from wildfires in central Portugal during summer 2009, Atmos. Environ., 57, 186–196, 2012.

Vicente, A., Alves, C., Calvo, A. I., Fernandes, A. P., Nunes, T., Monteiro, C., Almeida, S. M., and Pio, C.: Emission factors and detailed chemical composition of smoke particles from the 2010 wildfire season, Atmos. Environ., 71, 295–303, 2013.

Wang, C.: Impact of anthropogenic absorbing aerosols on clouds and precipitation: A review of recent progresses, Atmos. Res., 122, 237–249, 2013.