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

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Discussion F

Discussion Paper

Discussion Paper

Discussion Paper

ACPD

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract Introduction

Conclusions References

Tables Figures

 \triangleright

Close

•

Full Screen / Esc

Back

Printer-friendly Version



Understanding of aerosol sources which affect climate is an important problem open in front of scientists as well as policymakers. The role of aerosols in local climate variability depends on a balance between aerosol absorbing and scattering particles as well ₅ as on variability of environmental conditions. In this paper we investigate variability of aerosol content (both absorbing and scattering UV radiation) over Continental Portugal in dependence on 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, 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 the Continental Portugal during 1978-1993 time period. 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 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.

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

Paper

Discussion Paper

Discussion Paper

Discussion

Paper

ACPD

14, 31009–31038, 2014

Aerosols over Continental Portugal (1978 - 1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract Conclusions Introduction References

Tables

Figures





Back



Full Screen / Esc

Printer-friendly Version



Paper

sources include not only farming and charcoal burning but also emissions from car exhausts, factories and power plants. Some aerosols like 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 outcomes 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 on IPCC 2013, Ch. 7 "Clouds and Aerosols" indicates that aerosols give overall a 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 can have much stronger effect on the regional weather conditions and lead rather to local climate changes than to global. Normally, aerosol scattering makes the Earth's atmosphere more reflective and lead to cooling of the climate system, while aerosol absorption has the opposite effect and lead 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 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, 2008, 2011, 2012), Santos et al. (2008, 2013), Catry et al. (2009), Calvo et al. (2010), Obregón et al. (2012). The impact of the aerosol variations on the local climate variations was studied as well, both with in-situ measure-

ACPD

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract Introduction

Conclusions References

Tables Figures

l∢ ≻l

■ Back Close

Full Screen / Esc

Printer-friendly Version



The present paper is dedicated to understanding of the local aerosol sources and the role of the local aerosol content played in variations of the climate of the Continental Portugal region for the 1978–1993 period. This approach allows us to minimize the effect of the well known spatial heterogeneity of the aerosol content. 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 atmospheric aerosol index helps us to obtain information about the aerosol content in the studied region. 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 SO₂ series used as

ACPD

Discussion

Paper

Discussion Paper

Discussion Paper

Discussion

Paper

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract Introduction

Conclusions References

Tables Figures

|d | FI

Back Close

Full Screen / Esc

Printer-friendly Version



The paper is organized as follows: Sect. 1 presents modern state of the art and briefly gives overview of the paper. Section 2 contains the description of the used data sets 5 as well as their statistical properties. Section 3 describes variations of aerosol content over continental Portugal during 1978–1993 years and their main sources. In Sect. 4 we show how aerosol variation affect local climate during the studied period. Section 5 gives summary on the obtained results. Here in Sect. 2 we present only a short description of data sets used in the study. The detailed analysis of all these parameters can be found in the Supplement. Part 1 of the Supplement presents the detailed description of the data sets used in the paper: aerosol data (Part 1.1), atmospheric parameters (Part 1.2) and aerosol forcings (Part 1.3). Part 1.3.3 of the Supplement contains a description of the method used to detect dust events and Part 1.3.4 presents a short analysis of the similarity in the trends of aerosols and anthropogenic sulfates variations. Part 2 of the Supplement presents the correlation analysis between the sunshine duration series and other meteorological parameters.

Data sets

Aerosol parameters

The Total Ozone Mapping Spectrometer (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 aerosol content over Portugal. Under most conditions the AI is positive for the UV absorbing aerosols (pure absorption) and negative for the UV non-absorbing aerosols (pure scattering) when two close wavelengths in UV region near 360 nm are considered (see e.g. Ginouz 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 of absorbing (e.g. mineral dust, smoke, volcanic ash) or

Discussion Paper

Discussion Paper

Discussion Paper

Discussion

Paper

ACPD

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978 - 1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract Conclusions Introduction

References

Tables

Figures

Back

Close

Full Screen / Esc

Printer-friendly Version



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). We use the data on AI variations over two locations in the Continental Portugal (see Fig. 1a). The first one is the site ID 082 over Lisbon (38°46' N, 9°8' W, 105 m a.s.l.), the second 5 one is the site ID 288 over Penhas Douradas (40°25′ N, 7°33′ W, 1380 ma.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 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 smokes and dust events (Pereira et al., 2005, 2008; Obregón et al., 2012). In this study we used daily data only in one case: to indentify days with Saharan Dust Events (SDE) - see Sect. 2.3. For other purposes the monthly, seasonal and annual means of AI (and all other parameters) have been calculated. For each of two sites and for each of the months we calculated three series: <AI>, <Alpos> and <Alneg> 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

2.2 Atmospheric parameters

these three indices for both sites are shown in Fig. 1b-d.

Two sets of climatic data were used in this study. First one contains parameters measured by the Geophysical Institute of University of Coimbra (hereafter, "IGUC series"). Second one consists of the series measured by the Geophysical Institute of Institute Dom Luiz of University of Lisbon (hereafter, "IGIDL series"). Both locations are shown on the map in Fig. 1a (marked as "Coimbra" and "Lisbon", respectively). The sets of parameters include monthly and annual means of the following daily variables

 minimum (Tmin), maximum (Tmax) and average (averT) temperatures, and daily temperature range (DTR); ACPD

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract Introduction

Conclusions References

Tables Figures

l∢ ≯l

Back Close

Full Screen / Esc

Printer-friendly Version





2. accumulated precipitation amount (precip);

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

4. sunshine duration (SshD).

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 were used in pair with AI data from the site ID 082, and the IGUC series were used in pair with the AI series from the 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 measurements 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).

Aerosol sources

Not only spatial and temporal distributions of aerosols are very variable but also their origin as well. In this study we take into consideration only main sources responsible for the aerosol content variations over Portuguese region. These sources are mineral dust from Sahara and Sahel regions, wildfires, anthropogenic pollution (Pereira et al., 2005, 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 others than SO₂) remains outside the frames of our study due to 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.

ACPD

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978 - 1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract Introduction

Conclusions References

> **Figures Tables**

Printer-friendly Version

Please also note that all significances (*p* values) for correlation coefficients presented in this paper are calculated using 10 000 of the Monte-Carlo simulations with the random-phase FFT 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 significances (meta *p* values) were calculated as well.

2.3.1 Volcanoes

In this study we use the 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 resolution from October 1850 to December 2010. The data set is described in Sato et al. (1993) and Bourassa and Robock (2012). During the studied period four volcanic eruptions with the volcanic emissivity index VEI > 4 took places (numbered in Fig. 2c).

2.3.2 Saharan Dust Events (SDE)

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 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 short analysis of the dust events frequency for both Al sites is presented in the Supplement (Part 1.3.2). The variations of the monthly mean <Alpos> index averaged over two Portuguese locations increase together with the total monthly

ACPD

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I4 ►I

Back Close

Full Screen / Esc

Printer-friendly Version



number of dust events (as shown in the Supplement). <Alneg> variations on contrary have no relations to the SDEs.

2.3.3 Forest fires

Among all southern European countries Portugal shows the highest density of wild-fire ignitions (Catry et al., 2009). 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 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 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 site ID 082.

2.3.4 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 EBAS database (http://ebas.nilu.no) which contain the monthly mean values of SO_2 (in $\mu g S m^{-3}$) for the five Portuguese stations for period from August 1979 to December 2009. These stations are listed in the Supplement (Part 1.3.4). Since the data series contain significant

ACPD

14, 31009–31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I∢ ≻I

Back Close

Full Screen / Esc

Printer-friendly Version



3 Variations of aerosol content and their sources

The <Alpos> and <Alneg> series (Fig. 1c–d) for the same sites do not correlate with each other: the correlations coefficients between the <Alpos> and <Alneg> monthly series are 0.14 (*p* value = 0.05) for the ID 082 and 0.22 (*p* value < 0.001) for the 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 also shown the Supplement. The analysis of the standard statistical parameters of the <Alpos>/<Alneg> series shows that the absorbing aerosols play more significant role over the site ID 288 than over the site ID 082. The <Alpos> series for the site ID 288 has higher values of the mean, standard deviation and maximum values than the similar series for the site ID 082. On contrary, the same statistical parameters for the <Alneg> series are practically equal for these two sites.

3.1 Main aerosol sources

In this study we tried to take into account a number of main aerosol sources identifying their effect on the observed AI variations. These sources are volcanic eruptions, dust from Sahara desert, smoke and soot from the forest fires, and anthropogenic pollutions (sulfate aerosols series is considered as a proxy for most of the anthropogenic pollutant).

4

Discussion

Paper

Discussion Paper

ACPD

14, 31009–31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Discussion Paper

Discussion Paper

Title Page Introduction **Abstract** Conclusions References **Figures Tables** Back Close Full Screen / Esc **Printer-friendly Version** Interactive Discussion



The annual variations of the <Alpos> 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, like 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 <Al> and <Alpos> on the AOT variations and (2) no dependence between the <Alneq> 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 <Alpos> in 1982-1983 (at least, partly) and in 1988 are caused by the intensive Saharan dust intrusions. The <Alneq> series show 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 the both groups of districts. The correlation coefficients between the <AI> and BA series (Table 1) are guite 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 Al series variations.

Discussion

Paper

Discussion Paper

ACPD

14, 31009–31038, 2014

Aerosols over Continental Portugal (1978 - 1993)

A. L. Morozova and I. A. Mironova

Discussion Paper

Discussion Paper

Interactive Discussion



Title Page Introduction Abstract Conclusions References **Figures Tables** Back Close Full Screen / Esc **Printer-friendly Version**

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 <Alneg> (shown in Fig. 2b) and the SO_2 content. The anti-correlation between the curves reflects the increase of the scattering particles in the atmosphere (lower <Alneg> values) coinciding with the growth of the measured SO_2 concentration. Unsurprisingly, the <Alneg> variations over a highly populated location (ID 082 – Lisbon) show stronger dependence on the SO_2 content (see Table 1). The correlations between the <Alneg> and SO_2 variations became even much stronger when trends of these two parameters are studied. For example, the comparison of the monthly series of the SO_2 and the <Alneg> smoothed by the running averaging procedure (window of 36 months) shows that the satellite measured <Alneg> series follows the ground measured sulfate content data with probably a lag of about 5–10 months (see Supplement, Part 1.3.4).

Relatively high correlations between the <Alpos> over the site ID 082 and the SO_2 values (Table 1) probably caused by the similarities in the variations of different pollution gases/aerosols. Since the anthropogenic sulfates are almost totally scattering aerosols, they can not affect the satellite-measured <Alpos> 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 accepted time resolution and data quality to confirm this suggestion. On the other side, since the pollutants of different types are originated from the same sources (like traffic, coal and biomass burning, industrial activities etc., 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.

ACPD

Discussion Paper

Discussion Paper

Discussion Paper

Discussion Paper

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Full Screen / Esc

Close

Back

Printer-friendly Version

Interactive Discussion



31020

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 wild-fires, 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 <AI>, <AIpos> and <AIneg> due to the changes of the above mentioned forcings.

The models were constructed using a "best subset" technique that finds a subset of regressors (aerosol forcings, in our case) that predict 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 $r_{\rm adi.}^2$ parameters. The first one is a correlation coefficient between the modelled 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 comparing to the original data variance and taking into account the number of degrees of freedom. The $r_{\rm adi}^2$ was used as a criterion to compare the MRMs with different subsets of regressors: the subset that gives a bigger r_{adi}^2 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 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) Al series. The MRMs for the annual and summer Al series together with the corresponding original AI data are also presented in Fig. 3. All discussed above forcings are used as regressors for the <Alpos> and <Al> series, and only the wildfires and the pollutions are used to model the <Alneg> variations.

ACPD

Discussion Paper

Discussion Paper

Discussion Paper

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract Introduction

Conclusions References

Tables Figures

Back Close
Full Screen / Esc

Printer-friendly Version

Interactive Discussion



31021

The obtained results prove that chosen forcings represent a good set of regressors to explain the <Alpos> variations (Fig. 3b, e, h, k). The correlation coefficients between the MRMs and the original AI series are greater than 0.7, and the models explain (taking into account the number of degree of freedom) at least 35% of the variations of the original <Alpos> series. Unsurprisingly, Saharan dust events have a greatest contribution to the summer <Alpos> variations for both locations and the SO₂ content is an important regressor for the <Alpos> series measured above the more populated area (ID 082).

Concerning the <Alneg> series (Fig. 3c, f, i, l), it is clear that the used regressors can not sufficiently explain observed variations of the scattering aerosols, especially when the number of degree of freedom is taken into account. However, the MRMs for the <Alneg> show 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 like, for example, sea-salts or others than SO_2 pollutants. Unfortunately, there is no reliable corresponding data series that can be used in the frame of our study.

Finally, the MRMs for the <AI> series (Fig. 3a, d, g, j) are well correlated with the original series for both locations and explain 33–55% of the <AI> 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 pollution 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. For the comparison of the climatic conditions in Lisbon and Coimbra see

ACPD

14, 31009–31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I◀ ►I

→

Close

Full Screen / Esc

Back

Printer-friendly Version



the Supplement, Part 1.2. On contrary to the similarity of the climatic conditions for the studied locations, the measured AI monthly means are different for these two sites, as was discussed in Sect. 3. To our mind, this is a result of the different pollution and circulation conditions over the sites.

4.1 Site ID 288

The results of correlation analysis for the pair "AI ID 288 vs. IGUC series" are shown in Fig. 4 (b, d, f). As one can see, the relations between the climatic parameters and the aerosols of absorbing (<Alpos>) and scattering (<Alneg>) 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 scattering 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 a dry season in the 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 <Alpos> and the climatic parameters. One of the most important features is the anti-correlation between <Alpos> and parameters like SshD, Tmax and DTR during July. Similar relation take also places in June, August and October–November but the magnitude and the statistical significances of correlation coefficients *r* are smaller. This cooling trend coincides with epochs of frequent SDE events (high <Alpos> values) and is in an 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 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 Tmax (a parameter that can be considered as a measure of the day-time temperature): Tmax is decreasing. Consequently, the decrease of the Tmax affects the daily temperature ranges: the DTR decreases also.

ACPD

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I∢ ⊳I

→

Back Close

Full Screen / Esc

Printer-friendly Version



This kind of relations between the SshD and temperatures usually is related to the cloud effect on the radiation distribution in the lower atmosphere (Climate change 2007, IPCC 2013, Ch. 7). As it is shown in the Supplement (Part 2), the Tmax and DTR correlate very well with the SshD during almost a whole year. The precipitation amount 5 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 <Alpos> 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 a reason for the correlation between the amount of the absorbing aerosols and the Tmin (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 of high-level clouds reflecting some of the outgoing IR radiation back to the ground. The relation between the precipitation amount and the <Alpos> found for this site reflects also a process that can be identified as an indirect effect of the aerosols on the cloud formation (see e.g. Climate change 2007 (IPCC AR4), Ch. 7.5 "Aerosol Particles and the Climate System"). The increase of the <Alpos> 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 droplet in a relatively dry summer (and sometimes winter) air.

The effect of the scattering aerosols (described by the <Alneg> index) on some of the atmospheric parameters in the region is similar to the observed for the <Alpos>. The increase of the aerosol loading coincides with the decrease of the SshD, DTR, Tmax and averT in July (and in a weaker form in June and August) – Fig. 4f. The opposite relations take place in early spring season (February–April) when <Alneg> variations correlate with changes of the Tmin, Tmax, averT (and DTR in April). On the other hand, the precipitation has an opposite dependence on the <Alneg> variation compared with obtained for the <Alpos>. 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

ACPD

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I



Back



Full Screen / Esc

Printer-friendly Version



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. Besides, the Coimbra/Penhas Douradas area is stronger affected by the absorbing aerosols originated from wildfires and Saharan dust. As it was 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 decrease absorption properties.

First of all, the biggest correlation coefficients are obtained for the <Alneg> but not for the <Alpos> as for another location. As one can see from the comparison of the Fig. 4a, c, e and b, d, f, the similarities in the relations between the Al indices and the climatic parameters exist, mostly, for temperature parameters and <Alpos> during January–March, June and August–September periods. The significant seasonal differences are seen only in the <Alneg> variations. All temperature parameters tend to anti-correlate with <Alneg> amount in April and November (months of the transient seasons). On contrary for the May, August and September months (hot dry season) there is a tendency to correlation between the <Alneg> values and the temperatures. As a rule, temperature parameters tend to correlate with the Al (the more aerosol particles of both types, the higher the temperature) with just a number of exceptions (June for <Alpos>, and April and November for <Alneg>). The relations between the SshD and aerosol content are weak and sporadic. There is just a small number of significant anti-correlations between the <Alpos> and <Alneg> and the SshD series. These are June series for <Alpos> (Fig. 4c) and May series for <Alneg> (Fig. 4e). In the first

ACPD

Discussion

Discussion

Paper

Discussion Paper

Discussion Paper

14, 31009–31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

■







Back



Full Screen / Esc

Printer-friendly Version

Interactive Discussion



31025

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 the studied region we constructed the 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 <Alpos> and <Alneg>. 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 shows 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 ($r_{\rm adj}^2$) 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 Septem-

ACPD

14, 31009–31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I∢ ≻I

•

Close

Full Screen / Esc

Back

Printer-friendly Version



5 Conclusions

The analysis of the aerosol content changes over two sites in the Continental Portugal, their sources and their role in the local climate variations were studied using both ground and satellite measurements for the 1978–1993 period. One site is an urban region around Lisbon; another is located in a less populated mountain region. The aerosol content was obtained from the data of TOMS instrument on board of Nimbus-7 satellite. Absorbing and scattering aerosols were studied separately. Four main aerosol sources were considered: volcanic aerosols, Saharan dust events (SDE), wildfires and anthropogenic pollution. The effect of the aerosol on the atmospheric conditions was studied using the direct ground measurements by two meteorological stations close to the aerosol measurement sites. The set of atmospheric parameters includes four temperature parameters (minimum, maximum and average daily temperatures, and daily temperature range – DTR), atmospheric pressure, precipitation amount and sunshine durations. All data are of monthly and annual time resolution (except wildfire burned area series which are of annual resolution only).

The results of the analysis show that the aerosol sources chosen in this study play an important role in the local aerosol content variations. 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 four

Discussion

Paper

Discussion

Paper

Discussion Paper

Discussion Paper

14, 31009-31038, 2014

ACPD

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract

Conclusions References

Tables

Figures

Introduction

•



Back

Close

Full Screen / Esc

Printer-friendly Version



The variations of aerosol content were found to be in relations with the changes of atmospheric parameters. 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 (and, consequently, DTR) and absorbing aerosol content during summer months. These temperature and aerosols variations are also in 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 Tmax 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 urban region.

The Supplement related to this article is available online at doi:10.5194/acpd-14-31009-2014-supplement.

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Discussion Paper

Discussion Paper

Discussion Paper

Discussion Paper

ACPD

14, 31009–31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract Introduction

Conclusions References

Tables Figures

I4 ►I

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



31028

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Paper

Discussion Paper

Discussion Paper

Discussion Paper

Back

Printer-friendly Version

Interactive Discussion



ACPD 14, 31009–31038, 2014

Aerosols over Continental Portugal (1978 - 1993)

A. L. Morozova and I. A. Mironova

Title Page Introduction **Abstract**

Conclusions References

> **Tables Figures**

Close

Full Screen / Esc

Paper

Paper

Interactive Discussion



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ACPD

14, 31009–31038, 2014

Aerosols over Continental Portugal (1978-1993)

A. L. Morozova and I. A. Mironova

Title Page

Introduction Abstract

Conclusions References

> **Tables Figures**

Close

Back

Full Screen / Esc

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15

ACPD

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova



Table 1. Correlation coefficients between the variations of the AI and different forcing parameters: *annual* series in case of volcanic and pollution, 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	Al series						
		<ai></ai>	<alpos></alpos>	<alneg></alneg>				
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)				

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Title Page

Introduction

Abstract

Conclusions References

Tables Figures

l∢ ≯l



Full Screen / Esc

Printer-friendly Version



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

Time	Al	site	r	р	$r_{\rm adj.}^2$			β	
period	Туре			value		volcanic AOT	SDE number	area burned by wildfires	SO ₂ content
Annual	<ai></ai>	082 288	0.67 0.81	0.014	0.34 0.55	0.69 0.5	× 0.73	× 0.53	0.12 ×
	<alpos></alpos>	082 288	0.72 0.76	<0.01 <0.01	0.35 0.49	0.41 ×	× 0.98	0.34 0.43	0.57 ×
	<alneg></alneg>	082 288	0.71 0.37	<0.01 0.21	0.40 0.06			−0.47 ×	-0.5 -0.37
Summer (Jun–Sep)	<ai></ai>	082 288	0.79 0.67	<0.01 0.012	0.50 0.33	0.28 ×	0.41 0.97	× 0.68	0.59 ×
	<alpos></alpos>	082 288	0.95 0.82	<0.01 <0.01	0.88 0.60	×	0.57 1.05	× 0.37	0.63 ×
	<alneg></alneg>	082 288	0.44 0.68	0.12 0.012	0.12 0.41			-0.44 ×	× 0.68

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978–1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract Introduction

Conclusions References

Tables Figures

Back Close

Full Screen / Esc

Printer-friendly Version



Interactive Discussion



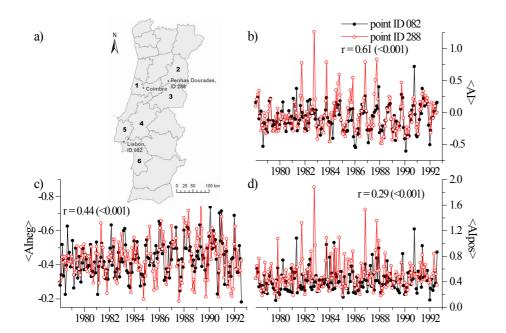


Figure 1. (a) - Map of Continental Portugal with the locations of the satellite Al observation points (ID 082 and ID 288) and the meteorological stations (Coimbra, Lisbon and Penhas Douradas). Districts used for calculation of the wildfire burned area: 1 – Coimbra, 2 – Guarda, 3 - Castelo Branco, 4 - Santarém, 5 - Lisboa, 6 - Setúbal. (b-d) - Monthly mean <AI> (b), <Alneg> (c) and <Alpos> (d) series for two satellite locations: ID 082 (black lines filled dots) and ID 288 (red lines open dots). Please note the inverted Y axis in c. Correlation coefficients r for the AI series from different sites are shown with p values in brackets.

ACPD

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978 - 1993)

A. L. Morozova and I. A. Mironova

Title Page Introduction **Abstract**

Conclusions References

> **Figures Tables**

 \triangleright \blacktriangleright

Close

Back

Printer-friendly Version

Interactive Discussion



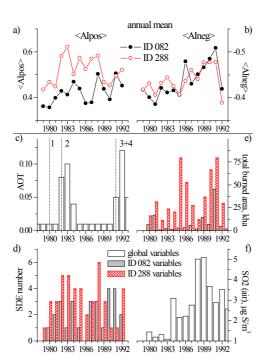


Figure 2. (a, b) – Annual variations of aerosol indices <Alpos> (a) and <Alneg> (b) for two locations: ID 082 (black lines filled dots) and ID 288 (red lines open dots). (c-f) - Annual values of parameters describing aerosol forcings: volcanic aerosols (c - AOT, annual means), Saharan dust events (**d** – annual sums for two Al locations), wildfires (**e** – total annual burned area close to AI locations) and anthropogenic sulfates (f - SO₂, annual means). White bars show global (AOT, mean values for the Northern Hemisphere) or averaged for a number of locations over whole Portugal (SO₂, means for five measurement locations) data; gray bars show data related to the site ID 082; red-white crossed bars show data related to the site ID 288. Four most significant volcanic eruptions are marked in (c) by vertical lines: 1 - Mt. St. Helens, 2 - El Chichon, 3 - Pinatubo and <math>4 - Mt. Hudson. Please note the inverted Y axis in **(b)**.

ACPD

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978 - 1993)

A. L. Morozova and I. A. Mironova

Title Page **Abstract** Introduction

Conclusions References

> **Tables Figures**

Þ Back Close

Full Screen / Esc

Printer-friendly Version

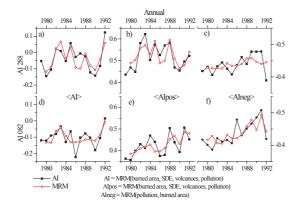




Printer-friendly Version

Interactive Discussion





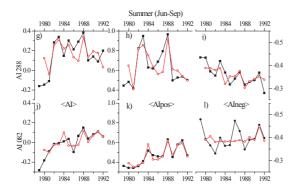


Figure 3. Multiple regression models (MRM) of the annual (a-f) and summer (June-September, g-I) Al series: $(a, d, g, j) - \langle AI \rangle$ series; $(b, e, h, k) - \langle AIpos \rangle$ series; $(c, f, i, k) - \langle AIpos \rangle$ 1) - <Alneq> series (please note the inverted Y axis). Original AI series are shown by black lines with dots; MRMs are shown by red lines with open dots. 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 <AI>, <Alpos> and <Alneg>.

Conclusions

Abstract



Introduction

References

ACPD

14, 31009-31038, 2014

Aerosols over

Continental Portugal

(1978 - 1993)

A. L. Morozova and I. A. Mironova

Title Page





Full Screen / Esc

Printer-friendly Version

Interactive Discussion



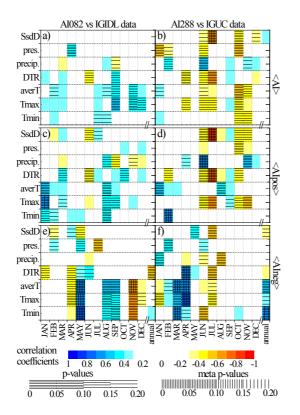


Figure 4. Correlation coefficients (r) between the AI indices ($\langle AI \rangle - a$, b; $\langle AIpos \rangle - c$, d; <Alneg> - e, f) for sites ID 082 (a, c, e) and ID 288 (b, d, f) and atmospheric parameters measured in Lisbon (IGIDL series -a, c, e) and Coimbra (IGUC series -b, d, f). Only correlation coefficients |r| > 0.3 are shown. The statistical significances 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.

ACPD

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978 - 1993)

A. L. Morozova and I. A. Mironova

Title Page

Abstract

Conclusions References

Introduction

Close

Tables

Figures

Þ

Back

Full Screen / Esc

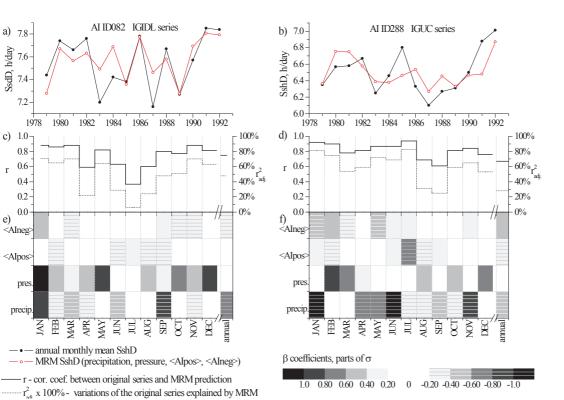


Figure 5. Multiple regression model (MRM) predictions and parameters for the annual and monthly sunshine duration from the IGIDL (a, c, e) and IGUC (b, d, f) series. (a, b) - the measured annual SshD series (black lines filled dots) vs. the corresponding MRM predictions (red lines, open circles). (c, d) – correlation coefficients r (black lines) between the MRM and the original series for the monthly data, and the variance explained by the MRMs (r_{adi}^2) in per cent, dotted lines). (e, f) - β coefficients (in parts of standard deviation σ) for each of the regressors for the monthly MRMs.

AI ID082 IGIDL series

1986

1988 1990

SE

AUG

a) 7.8

SsdD, h/day

7.2

c) 1.0 -

0.8

0.6

0.4

0.2

0.0 <AIneg>

<Alpos>

pres

precip

1980 1982

MAR APR MAY

annual monthly mean SshD

Z.

JUL.

1984

ACPD

14, 31009-31038, 2014

Aerosols over Continental Portugal (1978 - 1993)

A. L. Morozova and I. A. Mironova

Title Page

Introduction **Abstract**

Conclusions References

Figures Tables

Þ Back Close

Full Screen / Esc

Printer-friendly Version

