1 Study of the exceptional meteorological conditions, trace gases and particulate matter 2 measured during the 2017 forest fire in Doñana Natural Park, Spain 3 Adame, J.A.^{a,*}, Lope, L.^a, Hidalgo, P. J.^b, Sorribas, M.^a, Gutiérrez-Álvarez, I.^b, del Águila, A.^a, Saiz-Lopez, A.^c, Yela, 4 M.ª 5 ^aAtmospheric Sounding Station – El Arenosillo. Atmospheric Research and Instrumentation Branch. National 6 Institute for Aerospace Technology. INTA. Mazagón-Huelva. Spain. 7 ^bDepartment of Integrated Sciences. Faculty of Experimental Sciences. Huelva University. Campus de El 8 Carmen. Huelva. Spain. 9 ^cDepartment of Atmospheric Chemistry and Climate. Institute of Physical Chemistry Rocasolano. CSIC. Madrid. 10 Spain. 11 *Corresponding author E-mail address: adamecj@inta.es (J.A. Adame). 12 Abstract 13 14 In late June 2017, a forest fire occurred in Doñana Natural Park, which is located in southwestern 15 Europe. Many animal and plant species, some of which are threatened, suffered from the impact of 16 this fire, and important ecosystems in the European Union were seriously affected. This forest fire 17 occurred under exceptional weather conditions. The meteorological situation was studied at both 18 the synoptic scale and the local scale using meteorological fields in the ERA-Interim global model 19 from ECMWF (European Centre for Medium Range Weather Forecasts), the WRF (Weather Research 20 and Forecasting) mesoscale model and ground observations collected at El Arenosillo observatory. 21 Anomalies were obtained using records (observations and simulations) over the last two decades 22 (1996-2016). An anticyclonic system dominated the synoptic meteorological conditions, but a strong 23 pressure gradient was present; positive high pressure anomalies and negative low pressure 24 anomalies resulted in intense NW flows. At the surface, wind gusts of 80 km h⁻¹, temperatures up to 25 35 °C and relative humidity values less than 20 % were observed. In terms of anomalies, these observations corresponded to positive temperature anomalies (differences of 12 °C), positive wind 26 27 speed anomalies (greater than 29 km h⁻¹) and negative relative humidity anomalies (differences of 40

28 %). The forest fire reached El Arenosillo observatory approximately 8 hours after it began. When the fire started, record-setting maximum values were measured for all gases monitored at this site 29 (specifically, peaks of 99995 μ g m⁻³ for CO, 951 μ g m⁻³ for O₃, 478 μ g m⁻³ for NO₂, 116 μ g m⁻³ for SO₂ 30 31 and 1000 µg m⁻³ for PM10). According to the temporal evolution patterns of these species, the 32 atmosphere over a burnt area can recover to initial atmospheric levels between 48 and 96 hours 33 after an event. The impact of the Doñana plume was studied using hourly forward trajectories computed with the HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model to 34 35 analyse the emission source for the burnt area. The Doñana fire plume affected large metropolitan 36 areas near the Mediterranean coast. Air quality stations located in the cities of Seville and Cadiz 37 registered the arrival of the plume based on increases in CO and PM10. Using CO as a tracer, 38 measurements from the AIRS and MOPITT instruments allowed us to observe the transport of the 39 Doñana plume from the Strait of Gibraltar to the Mediterranean. Finally, after two days, the Doñana 40 forest fire plume reached the western Mediterranean basin.

41 Highlights

• A massive forest fire occurred in Doñana Natural Park in late June 2017.

- Biodiversity in this protected area was seriously affected.
- The fire occurred under exceptional synoptic and local meteorological conditions.
- The fire reached El Arenosillo, and record-setting values of trace gases were measured.
- The Doñana plume affected the western Mediterranean basin.
- 47
- 48 Keywords: Forest fire, Doñana Natural Park, biodiversity, trace gases (CO, O₃, NO₂, SO₂), ECMWF49 WRF model.
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54 **1. Introduction**

55 Climate change and air pollution will be the two greatest atmospheric challenges for societies in 56 future decades. These environmental issues are highly connected, and there are a number of factors 57 that link air pollution and climate change, such as emissions, atmospheric processes and chemistry 58 (von Schneidemesser et al., 2015). Climate change has led to an increase in the frequency and 59 intensity of extreme weather events, such as floods, droughts, heat waves and wildfires (Stott, 2016).

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Wildfires occur due to dry weather and the availability of fuel and ignition sources. It is well known that temperature, relative humidity, precipitation and wind speed affect fire spread rates and intensities (Parente et al., 2018). The occurrence of weather extremes, such as hot, dry and windy conditions, leads to the most severe fires (Dupire et al., 2017). Fire season length and duration can affect large areas and have increased across all vegetated continents during the late two decades due to extreme weather events and climate change (Jolly et al., 2015).

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Therefore, there is a clear feedback between climate change and forest fires. Climate change induces weather extremes, which lead to forest fires, while fire emissions contribute to climate change. These emissions increase greenhouse gas concentrations, thereby increasing atmospheric radiative forcing and aerosol concentrations and changing the Earth's albedo by depositing more lightabsorbing particles onto the Earth's surface (Sommers et al., 2014).

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Forest fires have impacts on the biosphere-atmosphere interface, atmospheric chemistry, atmospheric circulation, the composition of the ecosystem and its distribution, environmental degradation and air quality (Chen at al., 2017). Chemical species released by wildfires include gases such as CO₂, carbon monoxide (CO), methane (CH₄), non-methane organic compounds, nitrogen oxides (NO_x), nitrous oxide (N₂O) and sulphur dioxide (SO₂). The controlled combustion of various biomass fuels under laboratory conditions has revealed the presence of more than 500 different

volatile organic compounds (VOCs), which differ in terms of reactivity, health effects and the ability
to form active climate constituents (Gilman et al., 2015). Moreover, the simultaneous emission of
nitrogen oxides and reactive VOCs from the combustion of biomass leads to the photochemical
formation of tropospheric ozone (O₃) and secondary organic aerosols (Alvarado et al., 2015).

On the other hand, smoke is the main forest fire-produced atmospheric constituent that affects air quality and climate because the massive plumes can travel thousands of kilometres downwind. The monitoring of these plumes is only possible through satellite measurements, such as AIRS or MOPITT CO measurements, because CO is emitted by forest fires (Ding et al., 2015).

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This work analyses the case of a large forest fire that occurred in June 2017 in an important ecological area (Doñana Natural Park, located in southwestern Europe) under exceptional meteorological conditions because it is a clear example of the connection between extreme weather events, forest fires, air quality degradation and climate change. The meteorological context, impact on biodiversity and CO, O₃, NO₂, SO₂ and PM10 levels are analysed. The regional impact of the fire plume is studied by means of space observations and transport models.

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96 **2.** Area description, instrumentation, atmospheric models and satellite observations

97 2.1. Area description

98 The area of Doñana (116487 ha of a protected park) is considered to be a major hot spot of 99 biodiversity (López-López et al., 2011) and one of the most important wetlands in Europe. It is part of 100 the Guadalquivir River basin and is composed of highly diverse ecosystems: forests, ponds, fossil and 101 mobile dunes, riverbanks and beaches. The Doñana area features rich flora and fauna with many 102 endemic, threatened, and endangered species. Despite this high biodiversity, there are many threats 103 to its conservation, such as summer tourism, the pilgrimage of El Rocio, contamination due to heavy 104 metals and pesticides, intensive agriculture in its surroundings, aquiculture and frequent forest fires 105 (Groom et al., 2005). At least 80 % of the forest fires detected in the area are anthropogenically

106 induced, 50 % of which are directly provoked, and only 4 % are naturally induced (WWF/Adena, 107 2008). The vegetation in the area, which was intensively reforested with pines (*Pinus pinea*) and wide areas of shrublands, makes this territory highly sensitive to forest fires. These pyrophytic species 108 109 facilitate the fast expansion of fires, especially in summer, when high temperatures and drought 110 conditions predominate. Fires are natural disturbances in Mediterranean ecosystems and have 111 contributed to landscape dynamics for thousands of years (Trabaud et al, 1993). A large number of 112 flora and fauna species have adapted to periodic fires by developing strategies that allow them to 113 germinate, resprouting or recolonizing their habitat after fire. However, the delicate structure of the 114 Mediterranean ecosystem is altered when forest fires of anthropogenic origin reoccur frequently.

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116 El Arenosillo observatory is located in the SW Iberian Peninsula in Doñana National Park, 1 km from 117 the Atlantic Ocean coastline (Fig. 1). Surface meteorological conditions, solar radiation, surface 118 chemistry species (CO, O₃, NO₂ and SO₂), and optical, chemical and microphysical aerosol properties 119 are routinely monitored. The urban area of Huelva is located 35 km away from El Arenosillo in the 120 NW direction. The Seville metropolitan area is the largest urban area in SW Europe, with more than 121 1.5 million inhabitants, while the Cadiz urban area has a population greater than 0.5 million 122 inhabitants. These urban areas are near Doñana Natural and National Park (approximately 60-70 km 123 away). The transport of Doñana forest fire plumes to these areas could have potentially impact ~ 2 124 million inhabitants.

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127 2.2. Trace gases and meteorological instrumentation

Trace gases were measured at El Arenosillo observatory. O_3 was collected with an analyser (Dasibi 1008 RS) based on the absorption of ultraviolet radiation at 254 nm using a flow rate of 2 l min⁻¹. NO₂ was recorded with an analyser (TAPI 200E) based on the chemiluminescence method using a flow rate of 0.5 l min⁻¹. SO₂ was measured using an analyser (TAPI T100) based on the ultraviolet

fluorescence technique using a flow rate of 0.65 l min⁻¹. CO measurements were obtained using an analyser (TAPI T300) based on non-dispersive infrared spectroscopy using a flow rate of 0.8 l min⁻¹.
PM10 was measured at the air quality station Mazagon (7.5 km from El Arenosillo observatory) by an instrument based on the attenuation of beta radiation (FAG FH-62-N), which was equipped with a PM10 inlet outside the laboratory.

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These instruments belong to the Survey and Air Quality Control Network of the Environmental 138 139 Department at the Regional Government of Andalusia and follow the quality protocols defined by 140 European Directive (Directive 2008/50/EC). The measurement threshold ranges for instrumentation 141 were as follows: between 1 and 1000 ppb for O_3 (1.96 - 1960 µg m⁻³), between 0.4 and 261 ppb for 142 NO₂ (0.75 - 490 μ g m⁻³), between 0.04 and 88 ppm for CO (45.8 - 100032 μ g m⁻³), between 0.4-376 ppb for SO₂ (1.04 - 985.12 μ g m⁻³) and between 15 and 1000 μ g m⁻³ for PM10. Maintenance (weekly) 143 144 and calibration (monthly) operations were routinely performed to guarantee the accuracy of the 145 instruments. Gas data were collected every 10 min; the inlet was 8 m above the ground and 2 m 146 above the canopy. El Arenosillo observatory has available O₃ concentrations since February 2000, 147 NO₂ concentrations since August 2007, and CO and SO₂ concentrations since November 2015.

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Surface weather conditions were recorded using a Vaisala 520WXT analyser to monitor temperature, relative humidity, pressure, and wind speed and direction; the sensor was installed on the ground at a height of 10 m above ground level (a.g.l.). The data were collected at a time resolution of 10 minutes. Surface meteorological information is available at El Arenosillo site since 1994.

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To investigate the impact of the Doñana fire plume on other areas, observations collected by air quality stations belonging to the previously mentioned Air Quality Network of Andalusia have been used. In Seville, data from the Bermejales and Alcala stations were used; in Cadiz, data from the Cartuja station were used. These stations have been selected because they were suitable locations

downwind of the plumes and were not affected by urban emissions; furthermore, they were under the same meteorological conditions as those when the fire occurred. In all cases, the measurement techniques and maintenance and calibration protocols were similar to those mentioned previously for El Arenosillo observatory.

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163 2.3. Atmospheric models and satellite observations

164 ERA-Interim model meteorological fields from the ECMWF (European Centre for Medium Range Weather Forecasts) (Dee et al., 2011) with a spatial resolution of 0.5° x 0.5° were used to generate 165 166 the historical and anomaly maps. The model had 22 vertical levels from the surface to 250 mb and a 167 temporal resolution of 6 h. To understand the local meteorological fields in detail, the WRF (Weather 168 Research and Forecasting) model was used. Simulations were performed using the default options in 169 the WRF model (version 3.8). The planetary boundary layer was estimated by means of the Yonsei 170 University scheme (Hong et al., 2006), microphysical characteristics were simulated using the WRF 171 single-moment 3-class scheme, and longwave and shortwave radiation parameterizations were 172 carried out by the Rapid Radiative Transfer Model (RRTM) and the Dudhia scheme, respectively. The simulated area consisted of a single domain centred on the Gulf of Cadiz area (37 °N, 7.5 °W), which 173 174 utilized a grid with 100 points and a 9 km spatial resolution. The spatial resolution was chosen 175 following the 1:3 ratio between the inner and outer domains (Dudhia and Wang, 2014). The 176 boundary conditions and initial inputs utilized those from the ERA-Interim fields, with a 0.25° x 0.25° 177 spatial resolution. WRF simulations started at 00:00 UTC on 22 June 2017 and covered seven 178 complete days, which allowed the simulation to stabilize for a few days before reaching the forest 179 fire period.

180

Fig. 1.

181 To investigate the impacts of the Doñana fire plume, the forward trajectories of air masses were 182 computed with the HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model 183 developed by NOAA's Air Resources Laboratory (ARL) (Draxler et al., 2009; Stein et al., 2015). Three-

dimensional kinematic hourly trajectories were computed with a 24-hour pathway at 100 m a.g.l. The
 forward trajectories were calculated using ERA-Interim meteorological fields (spatial resolution of
 0.125° x 0.125°), which were converted into the ARL standard format using the HYSPLIT model.

187 The spatial distribution of the Doñana plume has been investigated using CO measurements from the 188 Atmospheric Infrared Sounder (AIRS) aboard the Aqua satellite, which covers the entire globe from 189 pole to pole twice a day. The AIRS instruments include an infrared spectrometer and a visible 190 light/near-infrared photometer. The standard AIRS products include temperature, water vapour 191 mixing ratios and trace gas concentrations (e.g., O₃, CO, CO₂, and CH₄), which are usually products or 192 by-products of biomass-burning and forest fires (Kumar et al., 2013; Kim and Sarkar, 2017). The AIRS 193 total column amounts of CO are obtained from the product AIRX3STD (level 3). Each file covers a 194 temporal period of 24 hours during either a descending or ascending orbit, with a spatial resolution 195 of 1° x 1°. In addition, continuous measurements of CO are taken by the MOPITT instrument (which 196 measures pollutants in the troposphere) aboard the TERRA satellite (Dekker et al., 2017; Jiang et al., 197 2017). CO data are obtained from the product MOP03J (level 3), which has a 22×22 km² spatial 198 resolution and a global coverage once every 2–3 days.

199

200 **3. Results**

201 *3.1. Doñana forest fire: biodiversity impact*

202 The forest fire started on the afternoon of 24 June 2017 southeast of the village of Moguer. Due to 203 extreme meteorological conditions (Section 3.2), the fire rapidly spread towards Doñana Natural 204 Park, an area that has been intensively reforested with pines. The fire was active for approximately 205 60 hours. The total affected area comprised 10339 ha (88 % of which was protected areas, primarily 206 Doñana Natural Park). A cartographic analysis indicates that a total of 40 endangered plant species 207 were directly affected by the forest fire (see Dávila and Hidalgo, 2017 for further details). In 208 particular, 50 % of the total localities of Linaria tursica, an endangered endemic annual plant in the 209 Doñana area, were impacted. With respect to animals, the habitats of the threatened Iberian lynx 210 (Lynx pardinus), the imperial eagle (Aquila adalberti, Spanish imperial eagle) and Adalbert's eagle 211 (Spanish eagle) were also affected. Reptiles and amphibians, such as a chameleon (Chamaeleo 212 chamaeleon), the red-tailed lizard (Acanthodactylus erythrurus), the Iberian newt (Lissotriton boscai) 213 and the meridional frog (Hyla meridionalis), were among the animals most affected due to their 214 limited capacity for displacement. With respect to the habitat present in Annex I of the Habitat 215 Directive (Council Directive 92/43/EEC on 21 May 1992), a total of 1664 ha of priority habitats were 216 affected (Fig. 1c and Table 1). The most affected habitat was the Atlantic decalcified dunes, of which 217 more than 1000 ha were colonized by heaths in the affected area. These habitats hosted many 218 endangered species and were especially sensitive to alterations. An intensive restoration programme 219 is necessary to recover such extreme fragile environments, especially the Mediterranean temporary ponds and dunes, due to their sensitivity to erosion after the removal of vegetative coverage. 220

221

Table 1.

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223 3.2. Meteorological conditions at the synoptic and local scales

224 The meteorological conditions in the lower atmosphere in this region are governed by both synoptic 225 and mesoscale processes. In the warm months (May to September), the synoptic flow can have a 226 maritime origin, and air masses can originate from the Atlantic Ocean or be continental (e.g., 227 originate from the African continent). In this last case, dust can be transported from the Saharan 228 Desert, which is associated with an increase in temperature and a decrease in relative humidity (e.g., 229 Adame et al., 2015). During situations with a low pressure gradient or atmospheric stagnation, 230 mesoscale processes develop, especially in the Atlantic area close to the Iberian Peninsula and the 231 western Mediterranean basin. In this coastal region, two types of sea-land breezes have been 232 identified (Adame et al., 2010): a pure sea-breeze, which has a wind direction perpendicular to the 233 coastline in both sea and land breeze regimes, and non-pure sea-breezes, which result from the flow 234 of pure sea-breezes and northwesterly synoptic forcing.

235 The mean synoptic meteorology was analysed over the last two decades for the week of 23 to 30 236 June (1996 to 2016) using ERA-Interim fields (Fig. 2a-c). At the synoptic scale, surface meteorological 237 conditions were characterized by anticyclonic conditions in late June. A high-pressure system centred 238 over the Azores region, which affects the Iberian Peninsula and western Europe, was associated with 239 a heat low system located over northwestern Africa. The temperature SW of the Iberian Peninsula oscillates between 25 and 30 °C, with values up to 30 °C in northern Africa, while the wind speed at 240 241 the surface only reaches 11-13 km h⁻¹. The highest wind speeds are observed along the coast of 242 North Africa and originate from pressure differences between the Atlantic High and the African Low, 243 which are induced by the Atlas Mountains, which act as an orographic barrier.

244 Using a similar strategy, local meteorological conditions at the surface were studied over the last 245 twenty years using meteorological records at El Arenosillo observatory. The temperature at this time 246 ranged from 17 to 19 °C during the nighttime and a maximum of 25 °C during the daytime. Because it 247 is a coastal region, the relative humidity during the nighttime reached 80 %, which decreased to 55-248 60 % at midday. In regard to wind speed, values of \sim 12.6 km h⁻¹ were obtained during the nighttime 249 and \sim 19.8 km h⁻¹ during the daytime. The atmospheric dynamics in the lowest troposphere were 250 defined by the development of mesoscale processes, Atlantic flows from the W-SW and low-251 frequency flows from the NW.

Several days before the fire event, synoptic conditions had anticyclonic characteristics, with a highpressure system propagating from the Atlantic Ocean to France, which covered the Iberian Peninsula (IP), and a low-pressure system over northwestern Africa. The IP had a weak gradient-baric field, which favoured mesoscale mechanisms in coastal areas, such as the sea-land breeze, which developed in the Gulf of Cadiz area.

The day before the forest fire (24 June), a low-pressure system located over North Africa deepened, and the Azores High moved away from the western Iberian Peninsula, which caused a decrease in the surface pressure in the Gulf of Cadiz and along the western coast of North Africa and an increase in the baric gradient.

These new conditions were not favourable for the development of mesoscale processes, and atmospheric dynamics were governed by synoptic flows from the NW. In the early hours on 24 June, an increase in the wind speed was observed in the coastal area of the Gulf of Cadiz, with gusts reaching 85 km h⁻¹. This change can be observed in the wind fields simulated by the WRF model at 19:00 UTC (Fig. 3a), which was when the fires started.

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Fig. 2.

The observed behaviour was associated with an increase in temperature and a decrease in relative humidity. On 24 June, a maximum temperature of 39 °C and a minimum relative humidity of 15 % were measured at El Arenosillo (Fig. 3d-e). The night of 24 June and throughout the day on 25 June, the wind field showed elevated wind speeds with changing wind directions, mostly in terms of the north component (Fig. 3b).

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Fig. 3.

A channelling effect occurred through the Gibraltar Strait, the Gulf of Cadiz and the Guadalquivir
valley. In the fire zone, there were areas with simultaneous winds from the NW and the SW (Fig. 3c),
which made fire control difficult and facilitated the spread of the fire.

276 On 25 June, a maximum temperature of 36 °C, a minimum relative humidity of 18 %, and wind gusts 277 up to 80 km h⁻¹ were recorded. At 14:00 UTC, the fire reached the El Arenosillo area, with 10-min 278 average temperature and relative humidity values of 45 °C and 15 %, respectively (1-min average values of 52 °C and 10 %, respectively) and wind speeds up to 45 km h⁻¹ with gusts up to 80 km h⁻¹ 279 280 (Fig. 3d-e). On 26 June, the anticyclone weakened, and the Iberian Peninsula was under a weak 281 pressure gradient, which favoured mesoscale mechanisms along the coast. At El Arenosillo, this 282 change was observed by decreases in the wind speed and temperature and an increase in the 283 relative humidity, which were consistent with typical meteorological conditions at this time of year.

To understand how exceptional these meteorological conditions were during the Doñana forest fire, the synoptic anomalies of mean sea level pressure (MSLP), temperature and wind speed on 24 June 2017 at 18:00 and 25 June 2017 at 12:00 were evaluated and compared against historical data from 1996-2016 (Fig. 2d-I). The local anomalies of daily average temperatures, relative humidity and wind
speeds on 24 and 25 of June 2017 were also evaluated.

The Doñana forest fire occurred under positive pressure anomalies over the Atlantic and negative pressure anomalies over northern Africa (i.e., the high and low were more intense than usual during this time of year). Therefore, a was observed, and NW flows with high wind speeds occurred over areas such as the Guadalquivir valley, the Gulf of Cadiz and the Atlantic Ocean, which were close to North Africa. High anomalies were obtained over the Gulf of Cadiz, with values of 29-36 km h⁻¹ with respect to the historical series.

295 Most of the Iberian Peninsula and North Africa showed positive temperature anomalies, especially 296 the Guadalquivir valley and the Gulf of Cadiz, with differences between 10 and 12 °C. At the local scale, based on the measurements from El Arenosillo observatory, positive temperature anomalies 297 298 were also observed, with nighttime differences from 6 to 8 °C and up to 10 °C during the daytime. 299 The relative humidity presented negative anomalies, specifically between 35 and 45 %. In terms of 300 absolute values, the daytime values were lower than 30 % and reached a minimum of 15 %. An 301 analysis of wind speed anomalies showed positive values during the nighttime (1.8 and 14.4 km h⁻¹) 302 and during the daytime (with differences of 25-29 km h⁻¹).

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304 3.3. CO, O₃, NO₂, SO₂ and PM10 measurements at El Arenosillo

305 Fig. 4 shows the temporal evolution of the CO, O₃, NO₂, SO₂ and PM10 values observed from 22 to 28 306 June 2017. In late June, CO concentrations in this area were characterized by values between 250 and 350 μ g m⁻³. Ozone concentrations varied between 60 and 80 μ g m⁻³ during the nighttime, with a 307 308 minimum occurring at 6:00 UTC. In the early morning, the ozone values increased to 90-100 μ g m⁻³ 309 (from 11:00 to 20:00 UTC). On the other hand, NO₂ values varied between 4 and 7 μ g m⁻³, with peaks in the early morning and during the nighttime of 25 and 40 µg m⁻³, respectively. According to local 310 311 meteorological conditions, air masses with elevated NO₂ levels arrived, which originated from marine emissions or from the industrial area of Huelva, similar to the nighttime observations from 23 to 24 312

June. SO₂ measurements showed background concentrations between 2 and 4 μ g m⁻³ since the observatory is not near an SO₂ emission area. However, under rare atmospheric conditions (i.e., less than 5 % per year), these values can reach 15-20 μ g m⁻³. Daily and hourly SO₂ background values were recorded before the fire. PM10 records in this rural area had values between 35 and 45 μ g m⁻³, indicating a desert dust particle contribution at ground level prior to the Doñana fire. Without the impact of desert dust, PM10 levels decreased to values of 20 μ g m⁻³.

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Fig. 4.

Prior to the fire, typical CO, O_3 , NO_2 and SO_2 values were recorded at El Arenosillo; PM10 was excluded, although it showed an important increase (peaks of ~500 µg m⁻³) on 24 June between 16:00 and 21:00 UTC. This particulate matter event occurred three hours before the Doñana fire began. A study was performed to investigate the origin of this increase in PM10, and it was attributed to the arrival of a plume that originated from forest fires occurring in Portugal.

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At 2:50 UTC on 25 June, approximately 8 hours after the forest fire started, an increase in trace gases and aerosols was observed at El Arenosillo, which remained until ~8:30 UTC (Fig. 4). These increases occurred under NW atmospheric flows and an increase in wind speed, which doubled from 14 to 28 km h⁻¹ with wind gusts of 144 km h⁻¹ (1-minute data). Moreover, the temperature increased to 32 °C at 8:10 UTC, and the relative humidity dropped to values lower than 35 %.

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During these nocturnal hours, the following peaks were reached: a CO peak of 8400 μ g m⁻³, an O₃ peak of 87 μ g m⁻³, a NO₂ peak of 90 μ g m⁻³, a SO₂ peak of 17 μ g m⁻³ and a PM10 peaks of 880 μ g m⁻³. Not all chemical species increased simultaneously since some are primary pollutants, while others, such as ozone, are formed through reactions of fire-emitted precursors. O₃ and NO₂ did not present a continuous increasing trend, as they reached a peak at approximately 3:00 UTC followed by a decrease (Fig. 4). The PM10 nocturnal peak coincided with the relative minimums for O₃ and NO₂, which were less than 20 μ g m⁻³. These values for PM10 are attributed to the arrival of ash, which

339 produced fast O_3 and NO_2 destruction via irreversible dry deposition onto the large ash particle 340 surface area.

From the early morning hours to 13:00 UTC, these concentrations decreased, and they resumed their 341 342 usual values. Although the wind continued to blow from the NW, a decrease in temperature was 343 recorded (Fig. 4). After 13:00 UTC, the wind speed and temperature began to increase, and the 344 humidity began to decrease as the fire surrounded El Arenosillo. Under the meteorological 345 conditions mentioned in Section 3.2., the fire reached the observatory, and our facilities suffered 346 from the fire during the next 1.5 hours (approximately until 15:10 UTC). Due to our autonomous 347 power system, the atmospheric instrumentation continued to work and measured the direct 348 emissions of the forest fire. Specifically, at 14:10 UTC on 25 June 2017, record-setting peaks for all gases monitored at El Arenosillo were measured: 99995 µg m⁻³ of CO, 951 µg m⁻³ of O₃, 478 µg m⁻³ of 349 NO₂, 116 µg m⁻³ of SO₂ and 1000 µg m⁻³ of PM10 (at Mazagon station) (Fig. 4). Note that in the cases 350 of CO, NO₂ and PM10, the instruments reached their upper limits of operation; hence, the 351 352 concentrations could have been even higher. The fire directly emitted chemical species such as CO, 353 NO₂, SO₂ and PM10. However, the presence of high levels of VOCs and NO_x associated with the fire 354 can promote additional atmospheric pathways for ozone production (Parrington et al., 2013). O₃ 355 production in wild fires includes the net effect of aerosols on chemical and photochemical reactions 356 within a fire plume and the impact of oxygenated VOCs and nitrous acid on O₃ production.

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The beginning of the post-fire period at El Arenosillo started at 15:30 on 25 June. The O₃ values decreased abruptly (only 7-10 μ g m⁻³ between 16:00-17:00 UTC), with values below 20 μ g m⁻³ during the following night. This decrease could be caused by several effects: i) due to the high ash concentrations, the solar radiation decreased, and the local photochemical activity ceased; ii) the high aerosol concentrations eliminated O₃ via dry deposition; and/or iii) the emission of VOCs and the conversion of radicals initiated the oxidation chains and removed O₃. In the following days (26 to 28 June), O₃ values did not recover; a maximum of 60 μ g m⁻³ was measured, with lower values than

usual during this time of year. Even though there was no smoke around the observatory, aerosol levels remained elevated, and hence, O_3 could have been removed by dry deposition on airborne particles. The O_3 concentrations did not start to exhibit normal values until 29 June (four days later), with peaks of 80 µg m⁻³ (not shown).

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370 Once the fire passed, CO concentrations remained elevated during the following eight hours, with 371 values between 5000 and 10000 μ g m⁻³. Almost 24 hours after the fire reached the observatory (approximately 14:30 UTC on 26 June), CO recovered to its usual values of ~300 µg m⁻³. After the fire, 372 373 NO_2 decreased, but over the next few hours, values remained between 10 and 25 µg m⁻³, which were 374 elevated for this region. After 1:00 UTC on 26 June (less than 24 hours later), the observed NO_2 375 values were below 10 μ g m⁻³, which are typical in this area and indicate recovery. SO₂ did not present 376 an abrupt decrease similar to that of O₃ and NO₂, but a continued decrease during the next 48 hours 377 was observed. This can be explained by the longer photochemical lifetime of SO_2 with respect to NO_2 . 378 SO₂ values below 5 µg m⁻³ were not recorded until 16:00 on 27 June. Later, a PM10 peak was 379 observed (a second peak of 560 µg m⁻³ between 0:00 and 1:00 UTC on 26 June); this showed a decreasing trend during the next eight hours. At 9:00 UTC, the PM10 concentrations were 60 µg m⁻³. 380

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382 3.4. Regional atmospheric impacts of the Doñana forest fire

383 To investigate the widespread impacts of the Doñana fire emissions, hourly forward trajectories were 384 computed for 25 to 27 June 2017 at a height of 100 m. For the analysis, a source region was defined 385 with five points, which covered the burnt area. The results obtained can be observed in Fig. 6. At 386 first, on 25 June, the forward trajectories showed atmospheric transport towards the Gulf of Cadiz, 387 but later, the plumes were transported towards the SE, affecting the Seville and Cadiz areas as well 388 as the Mediterranean area near the Strait of Gibraltar, which is in agreement with the wind fields 389 obtained with the WRF model (Section 3.2). On 25 and 26 June, the Doñana plume continued to 390 move toward the SE, reaching the western Mediterranean area.

391

Fire plumes impacted the Seville and Cadiz urban areas, which are located 60-70 km away from the 392 393 Doñana National Park (Fig. 1), and the arrival of plumes was registered by air quality stations, which 394 recorded increases in hourly CO, O₃ and PM10 values (Fig. 5). During the hours prior to the beginning 395 of the forest fire, increases in CO and O_3 (14:00-16:30 UTC) were observed in the Cadiz area, and an 396 increase in PM10 (18:30-20:30 UTC) was observed in both Seville and Cadiz, with a peak of 140 μ g m⁻ 397 ³. Lower values were observed at this time in El Arenosillo. This increase over Seville and Cadiz could 398 be attributed to the arrival of an air mass that originated from a forest fire that occurred in Portugal 399 during the second half of 24 June 2017, as mentioned in Section 3.3. Therefore, this plume from 400 Portugal not only affected the coastal area of the Gulf of Cadiz (Mazagon station) but also inland 401 areas.

402

The Cadiz urban area was affected by Doñana forest fire emissions at 10:30 UTC on 25 June, which 403 increased PM10 values to 100 µg m⁻³. However, a later impact was more intense and occurred 404 between 14:30 and 16:20 UTC. This later event featured a CO peak of 1600 μ g m⁻³ and PM10 values 405 406 higher than 240 μ g m⁻³. In addition, an increase in O₃ was recorded, reaching values of 135 μ g m⁻³. 407 Twelve hours later, at approximately 2:30-3:00 UTC on 26 June, the plume reached both the Seville 408 metropolitan area and the Cadiz urban area and affected this inland region over the next 8 hours until approximately 10:30 UTC on 26 June 2017. During this period, CO peaks with values of 2032 μg 409 m⁻³ in Seville (Bermejales station) and 1300 µg m⁻³ in Cadiz (Cartuja station) were recorded, and 410 411 maximum PM10 values of 110 μ g m⁻³ were recorded in the Seville area (Fig. 5).

412

Fig. 5.

413 On the first night of the fire (24 to 25 June 2017), the typical nocturnal removal of O_3 was not 414 observed, but the O_3 concentration did slowly decrease, with a minimum of 60-70 µg m⁻³. Since the 415 Doñana plume had not yet affected the Seville and Cadiz areas, this nocturnal behaviour could have 416 been associated with O_3 -rich aged air from Portugal. On 25 June, when the Doñana plume impacted

Seville and Cadiz, an increase in O₃ concentrations was observed, with daily maxima of 30-40 μg m⁻³, 417 higher than those measured on previous days (values of 125-130 µg m⁻³ compared to 90-100 µg m⁻³). 418 Because the Doñana plume was transported towards the east on 26 and 27 June 2017, surface 419 420 observations at other air quality stations located to the east (e.g., the Malaga metropolitan area) 421 were analysed. The recorded increases in O_3 , CO, NO_2 and PM10 were not attributed to the impact of 422 the Doñana plume. Hence, based on meteorological conditions, such as forward trajectories, wind 423 fields, and the absence of gas concentration increases in ground observations, the Doñana plume 424 was transported towards the Mediterranean Sea.

425

426 To understand the spatial distribution of the Doñana plume, CO was used as a tracer for space 427 observations from the AIRS and MOPITT instruments aboard the Aqua and Terra satellites, 428 respectively (Fig. 6). The day before the Doñana fire (24 June), the AIRS instrument measured high 429 CO total concentrations over the maritime area of the Gulf of Cadiz, which were also observed by 430 MOPITT on 25 June. Both instruments measured CO transport from the south of Portugal to SW 431 Spain. These observations support the arrival of the Portuguese plume to this area. However, the air quality stations measured an increase in the PM10 concentration on the evening of 24 June but not 432 433 an increase in surface CO. Based on these observations, it was determined that the CO plume was 434 transported at upper levels; both satellite instruments measured the CO column, and it was observed 435 that the increase in PM10 at the surface was likely due to gravitational settling processes.

436

Fig. 6.

437 On 25 June, AIRS observed a CO column of 1.7-1.8 x 10¹⁸ mol cm⁻² in the Doñana area and 438 Mediterranean area close to the Strait of Gibraltar. This column was transported towards the interior 439 of the Mediterranean in agreement with the forward trajectories and the wind fields (Fig 3. a-c). On 440 27 June, both the AIRS and MOPITT instruments showed an increase in CO in the Mediterranean area 441 near the east coast of the Iberian Peninsula. Atmospheric transport occurred on these days (24 to 27 442 June) in the western Mediterranean from the west to the east, which is confirmed by the forward

trajectories and ECMWF wind fields. Therefore, the arrival of emissions from Europe, North Africa orthe eastern Mediterranean should be neglected.

445

446 4. Conclusions

A forest fire affected the Doñana Natural Park from 24 to 26 June 2017. This fire had a strong impact on biodiversity and affected many endangered plant and animal species. Similarly, many priority habitats considered by the Habitat Directive were seriously damaged. An intensive restoration programme is necessary to recover the biodiversity in the region and avoid erosion.

451

452 The Doñana forest fire occurred under an exceptional meteorological scenario, which consisted of a 453 strong pressure gradient, positive high pressure anomalies in the Atlantic and negative anomalies in 454 North Africa, and areas of low pressure. The following atmospheric characteristics were also 455 observed: intense atmospheric flows from the NW; positive wind speed anomalies, with differences 456 greater than 8 m s⁻¹; wind gusts of 80 km h⁻¹; positive temperature anomalies, with differences 457 between 10 and 12 °C and an absolute maximum of 35 °C; and negative humidity anomalies with a difference of ~40 % and an absolute minimum of 15 %. Unfortunately, these meteorological 458 459 conditions, i.e., high temperatures (>30°), low humidity (<15 %) and high wind speeds, with gusts up 460 to 80 km h⁻¹, were suitable for the occurrence and spread of a forest fire.

461

The Doñana forest fire affected El Arenosillo observatory approximately 8 hours after it started. Despite these extreme conditions, the instrumentation continued to work, and record-setting values were observed for all gases measured at our observatory. After the fire ceased (i.e., during the postfire period), the chemical species recovered to their usual concentration values, although some species, such as CO, SO₂ and PM10, remained at high levels for several hours. The temporal evolution shown at El Arenosillo indicated that a burnt area should recover to its initial atmospheric levels between 48 and 96 hours after the event.

469

Hourly forward trajectories were computed with the HYSPLIT model to understand how the Doñana
plume developed. Forest fire emissions affected the Seville and Cadiz metropolitan areas and
subsequently moved towards the Mediterranean Sea. One day after the fires started, the air quality
stations in these urban areas observed increases in the CO and PM10 values. CO peaks of 1600 µg m⁻³
and 2032 µg m⁻³ and PM10 values of 240 µg m⁻³ and 100 µg m⁻³ were measured in Cadiz and Seville,
respectively.

476

After these metropolitan areas were affected, the Doñana plume continued moving toward the Mediterranean. To investigate its dispersion, CO was used as a tracer via observations from the AIRS and MOPITT instruments aboard the Aqua and Terra satellites. The increase in the total column of CO was measured in the Gibraltar Strait area and later in areas farther away from the Mediterranean.

481

Therefore, the Doñana plume had a local impact in the first 24 hours, reaching the Seville and Cadiz
metropolitan areas, and was later transported toward the Mediterranean and the Strait of Gibraltar.
Two days later, the Doñana plume had a regional impact, as it affected the western Mediterranean
basin (800-1000 km from emission source).

486

This forest fire was one of many fires that occur every year around the planet. If the fire close to Doñana Natural Park was small and had occurred under normal atmospheric conditions, it would not have caused the impacts that it had in this exceptional meteorological situation. When a forest fire occurs under extreme conditions, control of the fire is more difficult, the burnt area is greater, and the atmospheric emissions are greater. This forest fire had the potential to have a great impact on radiative forcing at a regional level. Forest fires similar to this event contribute to the occurrence of new unusual atmospheric events, leading to feedback processes.

495 Acknowledgments.

496 This work was supported by the Spanish Ministerio de Economía y Competitividad (MINECO) under grant CGL2014-55230-R (AVATAR project). The authors wish to thank the Survey and Air Quality 497 498 Control Network belonging to the Environmental Department of the Regional Government of 499 Andalusia (Spain) for access to its database. We acknowledge the NOAA (National Oceanic and 500 Atmospheric Administration) for the provision of HYSPLIT (Hybrid Single Particle Lagrangian 501 Integrated Trajectory Model). The authors acknowledge NCAR (National Center for Atmospheric 502 Research) and NOAA for the development of the WRF (Weather Research and Forecasting) model. 503 The authors thank AEMET (State Meteorological Agency - Spanish Government) and ECMWF 504 (European Centre for Medium-Range Weather Forecasts) for the ERA-Interim meteorological fields. 505 The authors also thank the Atmospheric Science Data Center at NASA Langley Research Center for 506 the provision of total CO measurements from the AIRS and MOPITT instruments.

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622 Table.

Table 1. Priority habitats (according to Habitat Directive, 1992) present in the area and total area affected.

Priority habitats	Area (ha/km ²)
3170-Mediterranean temporary ponds	289.9/2.899
2250-Coastal dunes with Juniperus spp.	52.1/0.521
2130-Fixed coastal dunes with herbaceous vegetation (grey dunes)	102.6/1.026
2150-Atlantic decalcified fixed dunes (Calluno-Ulicetea)	1206.64/12.066
4020-Temperate Atlantic wet heaths with Erica ciliaris and E. tetralix	12.55/0.125
Total Area	1664.00/16.64

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624

625 Figure captions.

Fig. 1. Iberian Peninsula in Europe (a); location of El Arenosillo observatory and the Seville and Cadiz
metropolitan areas (b); forest fire and different priority habitat locations (Directive, 1992) (c).

Fig. 2. Mean sea level pressure (MSLP), temperature and wind speed maps for the week of 23 to 30 June using
the period 1996-2016 (a-c). MSLP, temperature and wind speed anomalies for 24 June at 18:00 UTC and 25
June 2017 at 00:00 and 12:00 UTC compared to the historical averages over the last two decades (d-l).
Meteorological fields obtained from the ECMWF model using ERA-Interim files with a 0.5° x 0.5° spatial
resolution.

Fig. 3. Wind fields obtained from the WRF model with a 9 x 9 km spatial resolution for 24 June at 19:00 UTC and
25 June at 8:00 and 17:00 UTC (a-c). Wind (direction and speed) and wind gusts (d); temperature and relative
humidity (e) recorded at El Arenosillo (INTA; Huelva) from 22 to 28 June 2017. The shaded area represents the
forest fire period.

Fig. 4. Temporal evolution trends for CO, O₃, NO₂ and SO₂ measured at El Arenosillo and PM10 measured at the
Mazagon station from 22 to 28 June 2017 using 10-minute data.

639	Fig. 5. Temporal evolution trends for CO, O_3 and PM10 measured at Seville (CO and O_3 at the Bermejales
640	station and PM10 at the Alcala station) and Cadiz (Cartuja station) from 22 to 28 June 2017 using 10-minute
641	data.
642	Fig. 6. Total daytime CO columns observed by the AIRS and MOPITT satellite instruments from 24 to 27 June
643	2017 and forward trajectories obtained with the HYSPLIT model using ERA-Interim meteorological fields (0.125°
644	x 0.125°).
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Wind speed anomaly for 24/06/17 at 18:00 UTC.



20°W 15°W 10°W 5°W 0° 5°E 10° Wind speed anomaly for 25/06/17 at 00:00 UTC.



20[°]W 15[°]W 10[°]W 5[°]W 0[°] 5[°]E 10[°] Wind speed anomaly for 25/06/17 at 12:00 UTC.





20°W 15°W 10°W 5°W 0° 5°E 10 Temperature anomaly for 24/06/17 at 18:00 UTC.



20[°]W 15[°]W 10[°]W 5[°]W 0[°] 5[°]E 10[°] Temperature anomaly for 25/06/17 at 00:00 UTC.



20[°]W 15[°]W 10[°]W 5[°]W 0[°] 5[°]E 10[°] Temperature anomaly for 25/06/17 at 12:00 UTC.





MSLP anomaly for 24/06/17 at 18:00 UTC.



40° W 30° W 20° W 10° W 0° 10° E 20° MSLP anomaly for 25/06/17.at 00:00 UTC.



40° w 30° w 20° w 10° w 0° 10° E 20° MSLP anomaly for 25/06/17 at 12:00 UTC.













38[°] N

36[°] N

34[°] N

1.8

8°W 6°W

2

4°w

2°W 0° 2°E

2.2 x 10¹⁸ molec cm⁻²

38 1

36[°] N

34[°] N

8[°]W 6[°]W 4[°]W 2[°]W 0[°] 2[°]E

СО

1.4

1.6

