Fire-Pollutant-Atmosphere Interaction and Its Impact on Mortality in Portugal During Wildfire Seasons

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Key Points:

- Air pollution emitted by wildfires combined with heat stress are significantly associated with cause-specific mortality in Portugal during wildfire seasons.
- Months inside the wildfire season with stable atmospheric conditions and cleaner air presented lower cardiorespiratory mortality rates.
- Wildfire population exposure assessment would benefit from forecasting smoke during wildfire season.

Abstract

Wildfires expose populations to increased morbidity and mortality due to the increase of air pollutant emissions. This study assesses the impact of wildfire exposure in Portugal. In this work, we analyze the effects of the wildfire seasons (June-July-August-September-October) on monthly mortality by using data from atmospheric composition reanalysis, air quality stations, remote sensing, and mortality for exposure assessment, cluster analyses and regression models. Cluster analysis separated the months within fire seasons with extreme atmospheric conditions (months with more frequency of lower relative humidity and higher temperature, higher pollutant concentrations and higher wildfire activities), Cluster 1, from months with cleaner air and stable atmospheric conditions, Cluster 2. Linear regression showed statistically significant (*p-value* $<$ 0.05) correlation (r) between Cluster 1 and cardiorespiratory mortalities due to Diseases of the Respiratory System (DRS), Pneumonia (PNEU), Chronic Obstructive Pulmonary Disease (COPD) and Diseases of the Circulatory System (DCS) ($r_{DRS} = 0.49$; $r_{PNEU} = 0.42$; $r_{COPD} = 0.44$; $r_{DCS} = 0.45$). Cluster 2 presented no significant statistical correlation between atmospheric conditions and health outcomes. Results shows epidemiological evidence that heat stress combined with air pollution during wildfire season are contributing to increase disease burden. Besides that, we performed smoke forecasts over Portugal by using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model with satellite-based fire emission. Forecasted PM10 and PM2.5 concentration reproduced the behavior of the observations ($NRMSE_{PM10} = 3.70$, $r_{PM10} = 0.75$; $NRMSE_{PM2.5} = 1.51$, $r_{PM2.5} = 0.46$) during October 15-16th, 2017, fire episode. BC results matches with satellite observations.

Keywords: Air quality, Exposure, Cluster analysis, Linear regression, Dispersion model

Plain Language Summary

We investigated the correlation between five cause-especific mortality (DRS, PNEU, COPD, ASMA and DCS) and Fire-Pollutant-Atmosphere components during wilfire seasons in Portugal. Therefore, we used data from atmospheric composition reanalysis, air quality stations, remote sensing, and mortality for exposure assessment. Through cluster analyses and linear regression models we found that in months with low-relative humidity, high-temperature, high-pollutions concentrations and high-wildfire activities, the incidence of DRS, PNEU, COPD, ASMA and DCS were higher. We also performed smoke forecasts over Portugal using a dispersion model with satellite-based fire emission. The population would benefit from forecasting smoke during wildfire seasons.

1 Introduction

Exposure to poor ambient air quality increases morbidity and mortality and is a leading contributor to global disease burden (Cohen et al., 2017). Air pollution - both household and ambient - remains responsible for 6.7 million deaths in 2019 (Fuller et Al., 2022). In Europe, air pollution is the single largest environmental risk and has significant impacts on the health of the European population (EEA, 2020). A considerable proportion of premature deaths in Europe could be avoided annually by lowering air pollution concentrations, particularly below World Health Organization (WHO) guidelines (Khomenko et al., 2021).

Air pollution has various health effects. WHO reports on six major air pollutants, namely particle matter (PM2.5 and PM10), ground-level ozone (O3), carbon monoxide (CO), sulfur oxides (SOx), nitrogen oxides (NOx), and lead. In this regard, short-term exposure to these pollutants is closely related to COPD (Chronic Obstructive Pulmonary Disease), cough, shortness of breath, wheezing, asthma, respiratory disease, and high rates of hospitalization (a measurement of morbidity) while the long-term effects are associated with chronic asthma, pulmonary insufficiency, cardiovascular diseases, and cardiovascular mortality (Manisalidis et al., 2020).

In Europe, emission from traffic, household heating, energy production and industrial combustion might be the main source of air pollution, but emissions from wildfire events during the fire season in Europe can cause extensive environmental impacts including air pollution. Wildfires emit substantial amounts of air pollutants that can travel over large distances, affecting air quality and human health far from the originating fires (Youssouf et al., 2014; Bowman et al., 2017; Sicard et al., 2019; Machado-Silva et al., 2020; Augusto et al. 2020; Requia et al., 2021; Duarte et al., 2021; Salgueiro et al., 2021, Tarín-Carrasco et al., 2021). Nevertheless, the combination of extreme drought with heat-waves has both been identified as crucial factors in the occurrence of wildfires in Mediterranean forests and shrublands, consequently, bringing large socio-economic impacts (Ruffault et al, 2020). Heat stress and forest fires are often considered highly correlated hazards as extreme temperatures play a key role in both occurrences (Vitolo et al., 2019).

In contrast, emissions from wildfires can exacerbate the effects of heat stress on human body, especially on the cardiovascular and respiratory systems (Finlay et al., 2012). Primary emissions from wildfire events that worsen air quality include PM2.5 and PM10, Black Carbon (BC) and gaseous species as CO, methane (CH4), nitrous oxide (N2O), and other combustion pollutants (Urbanski et al., 2008). Fire emissions also contain several trace metals (Cascio, 2018). Air pollution from biomass burning also contribute for the formation of secondary pollutants such as polycyclic aromatic hydrocarbons (PAHs), and volatile organic compounds (VOCs), and O3 generated by the photoreaction of NOx in the atmosphere (Jaffe et al., 2012).

Climate strongly influences global wildfire activity by increasing the frequency and intensity of wildfires in many regions due to climate change (Moritz et al., 2014; Jolly et al., 2015). Wildfires occur at the intersection of dry weather, available fuel, and ignition sources (Moritz et al., 2005). According to Abatzoglou and Kolden (2013), weather is the most variable and largest driver of regional burned areas. Meteorological variables such as temperature, relative humidity, precipitation, and wind speed independently influence wildland fire spread rates and intensities, and the alignment of multiple weather extremes, such as the co-occurrence of hot, dry, and windy conditions leads to the most severe fires (Flannigan and Harrington, 1988). Over mountainous complex terrain, fire weather conditions can be exacerbated by local orography effects that induce intense wind gusts near the surface, increasing fire danger (Couto et al., 2021). According to Turco et al. (2019), enhancing the understanding of how climate change and extreme weather events influence the evolution of the burned area is crucial to assess regional vulnerabilities and and mitigate their impacts.

Nonetheless, climate changes are expected to increase fire season severity over the coming decades (Flannigan et la., 2013). According to Turco et la. (2017), summer fires frequently rage across Mediterranean Europe are often intensified by high temperatures and droughts. Several works suggest that coincident drought conditions and high temperatures promote larger fires in southern Europe (Viegas and Viegas, 1994; Pereira et al., 2005; Pereira et al., 2011; Pausas, 2004; Pausas, 2008; Chuvieco et al., 2009; Turco et al., 2013; Trigo et al., 2016; Turco et al., 2017; Turco et al., 2018; Turco et al., 2019).

Regarding the health impacts of wildfire smoke exposure, Reid et al. (2016) performed a consistent critical review of 53 epidemiological studies indicating that wildfire smoke exposure is associated with respiratory morbidity with growing evidence supporting an association with all-cause mortality. Wildfire pollutants constitute a risk factor for adverse cardiovascular effects, especially among susceptible populations, including the elderly, pregnant women, and those with low socio-economic status (Chen et al., 2021). The young and healthy may also develop biological responses including systemic inflammation and vascular activation (Chen et al., 2021).

In Europe, several studies were performed on population health outcome because of wildfire smoke exposure such as Hänninen et al. (2009), Youssouf et al. (2014), Foustini et al. (2015), Linares et al., (2018), Augusto et al. (2020), European Commission (2020), Chas-Amil et al. (2020), Oliveira et al. (2020), Tarín-Carrasco et al. (2021), Brito et al., (2021), Barbosa et al. (2022). All these works showed, with different methodologies, the importance of wildfires in Europe during the fire season as a public health issue.

Given that Portugal is a very fire-prone region and where the biomass stocks is strongly correlated with wildfire events over space and time, further epidemiological studies in Portugal are essential, which could provide better support for policymakers with the objective of improving the quality of public health. In addition, rural fires that occur in Portugal have a major impact on air quality across Europe as Augusto et al. (2020) showed. Another very important factor is that in Portugal the phenomenon of double aging of the population has worsened with a significant increase in the elderly population – therefore, a group more vulnerable to develop health problems and more sensible to weather extreme events and climate change impacts - and a decrease in the young population according with INE (2022). With uncontrolled wildfires comes the release of large amounts of pollutants into the atmosphere, which may affect local areas but also suffer from long-range transport, contributing to an increase in air pollution in regions far from the source, exposing several people near fire regions or further away to air pollution risk.

Our research addresses this gap by forecasting smoke plumes during the October 2017 Portuguese wildfire outbreak by using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) dispersion model at 0.1° (~10 km) horizontal resolution. The HYSPLIT model was conducted to predict surface PM2.5, PM10 and BC and to assess the concentrations of these pollutants at ground level. Another important task of this work was to assess the main interaction among Fire-Pollutants-Atmosphere components and mortality in Portugal between 2012 and 2019 intra-annual wildfire seasons in Portugal. For this purpose, we investigated this impact by using modeled and remote sensing data, meteorological variables, and mortality information to analyze the impact of wildfire exposure in the Portuguese population. This approach allowed a clearer understanding of the magnitude of impact that wildfire seasons can have on human life.

2 Materials and Methods

2.1. Study area

Portugal is located in the southwestern Europe, in the Iberian Peninsula and faces the Atlantic Ocean in its West and South coast, **Figure. 6**, in the transition zone between subtropical and medium latitude types of climate (Turco et al., 2017). The study location was strategically chosen because of the spatiotemporal climatic variability, where the population and the ecosystems suffer frequently from intense natural hazards such as droughts, heatwaves and wildfires that tends to become more intense and common under climate change scenario (Turco et al., 2019). Portugal was also the epicenter of the extraordinarily intense fire season of 2017 with a record total burned area of about 500 000 hectares and more than 120 immediate fatalities (Turco et al., 2019; Augusto et al., 2020). Continental Portugal territory has a temperate type of climate, characterized by a rainy and mild winter and a hot and dry summer (Csa) in the south, and mild and dry (Csb) in a major part of the north (Kottek et al., 2006; Parente et al., 2019).

2.2. Exposure data

2.2.1. Remote sensing data

Monthly Burned Area (Burned_Area) (ha) data from 2011 - 2020 for Portugal were obtained from the Portuguese Institute for Conservation of Nature and Forests, respectively based on ground and satellite measurements and detailed information on ignition and extinction date and time of fire events (Pereira et al., 2011), and assessing changes in the fire regime due to different types of climate and fire management activities (Parente et al., 2016).

Daily Thermal Anomalies/Fire locations for October 15^{th}-16^{th} of 2017 were derived from the Moderate Resolution Imaging Spectroradiometer (MODIS) (Giglio, 2016; Land Atmosphere Near Real-Time Capability for EOS Fire Information for Resource Management System, 2021), and Visible Infrared Imaging Radiometer Suite (VIIRS) (Schroeder et al. 2020). The MODIS Terra (MOD14) and Aqua (MYD14) sensor resolution are 1 km, and the temporal resolution is daily. The fire detection and thermal anomalies are represented as red points and are largely used for studying the spatial and temporal distribution of fire. The 375m I-band from VIIRS data complements the MODIS fire detections (VIIRS S-NPP I Band 375 m Active Fire locations NRT, 2020).

The VIIRS Deep Blue Aerosol Type layer (Sayer et al., 2018; Hsu et al., 2019; VIIRS Atmosphere Science Team, 2021) was used to characterize the aerosol composition over the studied area during on October 15^{th}, 2017, fire outbreak. The VIIRS Deep Blue (DB) dataset product provides total aerosol optical depth (AOD) at 550 nm with a daily global coverage. The DB product includes a the aerosol-type classification - namely dust, smoke, high-altitude smoke, non-smoke fine-dominated, mixed, background, and fine-mode-dominated - and quality-assurance flags over ocean and land from 1 (worst quality) to 3 (best quality) (Sayer et al., 2018; Hsu et al., 2019; Escribano et al., 2022).

Daily Aerosol Index Layer from Ozone Mapping and Profiling Suite (OMPS) and Nadir-Mapper (NM) derived from Suomi-NPP (Torres, 2019) satellite were used to identify areas over Portugal with high amounts of aerosol in the atmosphere on October 15th, 2017. The OMPS Aerosol Index layer indicates the presence of ultraviolet (UV)-absorbing aerosols in the lower troposphere (1-3 km) such as desert dust and soot particles in the atmosphere (Seftor et al., 2014; Torres, 2019). In this work, OMPS Aerosol Index layer was used to identify smoke from Portuguese wildfires.

2.2.2. Air pollution and meteorological data

In this work, surface observational air pollution data obtained from the Online Database on Air Quality (QualAr) of the Portuguese Environment Agency (Agência Portuguesa do Ambiente (APA), at https://qualar.apambiente.pt) were used. QualAr database also provides information of the lacation type (Urban, Suburban and Rural) and the type of emission influence (Background, Traffic and Industrial), following the Commission Decision 2001/752/EC, of October 17, 2001 (APA), 2008).

Data used here refers to levels of PM2.5 and PM10 registered for every hour in 9 background stations (Estarreja, Ilhavo, Aveiro, Fornelo Monte, Fundão, Montemor-o-Velho, Ervedeira, IGC and Fernando Pó) located in different places in Portugal, mainly in rural areas, represented by black triangles in **Figure. 6(c)**. From these nine QualAR stations, all of them measured PM10 concentration and 7 of them measured PM2.5 concentration. The hourly, daily and monthly mean concentrations were considered as two variables named PM10_Obs and PM2.5_Obs.

The air quality data registered every hour are validated by QualAR and were averaged for the 2011-2020 period in a subsequent analysis for each pollutant. The background stations or rural stations are in geographical areas far from the influence of traffic routes, industrial areas, or any anthropogenic source, which make them a good tool to assess the impact of forest fires. From the hourly data, the daily, monthly, and annual averages were calculated. From the daily concentration of PM2.5 and PM10 corresponding to background stations, it was calculated how many times PM10 and PM2.5 were above WHO (2006) global air quality guidelines 2005 (25 µg/m^3 24-hour mean for PM2.5 and 50 µg/m^3 24-hour mean for PM10). The daily WHO guidelines overpasses of PM10 and PM2.5 were monthly counted and considered as two variables named WHO_PM10 and WHO_PM2.5.

Another important source of data for this work was the European Centre for Medium-Range Weather Forecasts (ECMWF). The ECMWF operates services related to meteorology and atmospheric composition variables and is available through the Copernicus Atmosphere Monitoring Service (CAMS, https://ads.atmosphere.copernicus.eu) on behalf of the European Union including CAMS-Reanalysis. The CAMS-Reanalysis combines models with in-situ and satellite observations through data assimilation technique. Monthly mean data obtained from CAMS-Reanalysis includes Carbon monoxide (CO_CAMs), Black carbon aerosol optical depth at 550 nm (BC_AOD_CAMs), Dust aerosol optical depth at 550 nm (Dust_AOD_CAMs), Particulate matter d < 10 µm (PM10_CAMs), Particulate matter d < 2.5 µm (PM25_CAMs), Relative humidity (RH_CAMs), Surface pressure (Pres_CAMs), Wind velocity (WS_CAMs), 2m temperature (T2m). From T2m we calculate minimum (T2m_Min_CAMs) and maximum (T2m_Max_CAMs) temperature. Data was obtained at a spatial resolution of 0.75° (~80 km). In our analyses, we used CAMS-Reanalysis for the period between 2012 and 2019. A validation of the CAMS global reanalysis can be found in Inness et al. (2019).

2.2.3. Health and population data

Monthly mortality data for Portugal was provided by the National Institute of Statistics (INE) (INE, https://www.ine.pt/) and refer to mortality due to a specific cause in the period 2012-2019, based on the use of administrative data for statistical purposes from the Integrated System of Civil Registration and Identification (SIRIC) and the Information System for Certificates of Death (SICO). Standardized mortality rates (per 100 000 inhabitants - all ages) was selected according to the International Classification of Diseases, version 10 (ICD-10): Diseases of the circulatory system (DCS) (ICD-10: I00-I99); Diseases of the respiratory system (DRS) (ICD-10: J00-J99); Pneumonia (PNEU) (ICD-10: J12-J18); Chronic obstructive pulmonary disease (COPD) (ICD-10: J40-J44); and Asthma (ASMA) (ICD-10: J45-J46). Since most of the data show seasonality, we used monthly data to address intra-annual variability of environmental and health data with focus on the fire seasons in Portugal (June-October) for the period between 2012 and 2019 due to lack of mortality data for years before 2012 and after 2019 at INE database.

2.2.4. Statistical analyses

The influence of fire and meteorological pollutant-atmospheric variables on mortality rates is then studied based on intra-annual analyses performed over the 8 year-period 2012–2019. Standardized anomalies (Z) method was used to ensure that the different variables had equal weights in the statistical analysis process. Accordingly, monthly values of each variable (X) are used to calculate the respective long-term mean () and standard deviation (σ), and standardized anomalies (Z) for each month are then obtained as:

$Z = \frac{(X -)}{\sigma}$ (1)

The strength of the relations among fire and atmospheric variables during the fire season in Portugal is assessed by a multivariate approach called principal component analysis (PCA) based on the correlation matrix. The Pearson correlation coefficient was considered for *p-value* < 0.05. The PCA was applied to monthly fire (Burned_Area), Air quality variables (PM10_Obs; PM2.5_Obs; CO_CAMs; BC_AOD_CAMs; Dust_AOD_CAMs; PM10_CAMs; PM25_CAMs) and meteorological variables (RH_CAMs; Pres_CAMs; WS_CAMs; T2m; T2m_Min_CAMs; T2m_Max_CAMs) to construct two spatial-temporal pollutant-atmosphere interaction index called Pollutant-Atmosphere Index (PAI) and Atmospheric-Pollutant Index (API). PCA converted the actual correlated fire-pollutants-meteorological variables into a new set of orthogonal and uncorrelated components.

With K-means cluster analysis was possible to perform a classification of PAI and API indexes in relation to mortality variables (DCS; DRS; PNEU; COPD; ASMA). In this regard, after performing K-means cluster to PAI-API-mortality, the data were separated in two groups, so that samples within the same group are as similar as possible and that the two different groups (clusters) are as different as possible in their constitution.

Finally, the Clusters were submitted individually to a linear regression method to study the statistical association between the independent variables (PAI and API) and the dependent variables (DCS; DRS; PNEU; COPD; ASMA). The linear regression was applied for each dependent variable and the response variable in each group separately. The collinearity of variables was examined by Pearson's correlation test. Significant differences of the values between the groups were tested at the *p-value* < 0.05 level, unless specified. Statistical analyzes were restricted to the years 2012 to 2019.

2.3. HYSPLIT trajectory model

The HYSPLIT model, developed by NOAA's Air Resources Laboratory, is an atmospheric dispersion model widely used to pollutants simulations of forward and backward trajectories (Draxler and Hess, 1998; Draxler, 2006; Stein et al., 2009; Stein et al., 2015; Rolph et al., 2017). The HYSPLIT Version 4.0.1 was employed to simulate the October 15th-16th of 2017 fire episode in Portugal. Overall model configuration details are provided in **Table 1**. The simulation started at 0000 UTC on October 15th and stopped at 0000 UTC October 17th with an output concentration grid resolution of 0.1°. The output concentration grid was centered at 40°N–8°W and extended 10° in each cardinal direction. The model top was set to 10 km above ground level (agl). The simulations were carried out using three-dimensional particle mode with 25 000 computational point particles released per hour — and distributed randomly — within each 0.1° emission grid cell. Smoke particles (PM2.5, PM10, BC) are subject to dry deposition (including turbulent diffusion).

For emission data the GFAS v1.2 (Kaiser et al., 2012) were used. GFAS v1.2 assimilates Fire Radiative Power (FRP) observations were obtained from the National Aeronautics and Space Administration (NASA) through the MODIS instruments onboard the Terra and Aqua satellites and produces daily estimates of biomass burning emissions. Furthermore, it provides information about injection height derived from fire observations and meteorological information using the operational weather forecasts of the ECMWF (Kaiser et al., 2012). The data are gridded to a regular latitude-longitude grid with a horizontal resolution of 0.1°.

The meteorological input used here to calculate 3-D forward trajectories (horizontal wind field plus vertical wind) is the Global Forecast System (GFS) dataset from the National Center for Environmental Prediction (NCEP) (http://www.ready.noaa.gov/archives.php). These data are provided with 0.25° x 0.25° horizontal resolution, 55 vertical levels of hybrid sigma-pressure coordinates, and temporal resolution of 3h.

Table 1.

HYSPLIT Model Configuration and Major Setup Selections.

HYSPLIT Model Configuration and Major Setup Selections	
BB Emission data sets (EMIS)	GFAS v1.2
Plume rise schemes (PR)	Briggs, 1969 (B69)
Meteorology inputs (MET)	GFS 0.25°
Mixing layer depth options (MIXD)	mixing layer height derived from temperature p
Vertical motion (VM) options	Using the meteorological model's vertical veloc
Particle release mode	Full 3-D particle horizontal and vertical
Number of particles	25 000 particles per hour for each source and p
Maximum number of particles	250 000
Horizontal resolution	0.1° (~10 km)
Surface concentration layer	0–100 m
Domain center/radius	40°N–8°W
Model top	10 km (agl)
PM2.5 Average diameter and density	0.8 mm and 2 g/cm^3 (Rolph et al., 2009)
PM10 Average diameter and density	4.0 mm and 2.5 g/cm^3 (Rolph et al., 2009)
BC Average diameter and density	0.0236 mm and 1.0 g/cm^3 (Rolph et al., 2009)
Minimum mixing layer depth	100 m

3 Results and discussion

3.1. Burned area and air pollution

Figure. 1 shows the yearly and monthly burned area (BA), and PM10 and PM2.5 mean concentration in Portuguese background stations from 2011 to 2020. The annual record-breaking wildfire season of 2017 is evident, with more than 500 000 ha of total burned area, **Fig. 1(a)**. For these ten years the BA time series does not show a clear decadal trend, although the annual concentrations of PM10 and PM2.5 show decreasing trends. The Mann-Kendall test confirms that there is a statistically significant decreasing trend (*p-value* < 0.05) for PM10 and PM2.5 and negative but not significant for BA (*p-value* > 0.05). **Fig 1(a)** also makes evident the importance of fires in increasing air pollution concentrations at the surface level as the annual averages of PM10 and PM2.5 concentrations are higher in years of greater burned area.

Regarding the monthly concentrations of PM10 and PM2.5, **Figure. 1(b)**, there is a statistically significant decreasing trend ($p\text{-}value < 0.05$) and no statistically significant trend for BA ($p\text{-}value > 0.05$). However, it is evident that in the months when there were more fires, the concentrations of PM10 and PM2.5 increased, showing that wildfires are an important source of particulate matter.

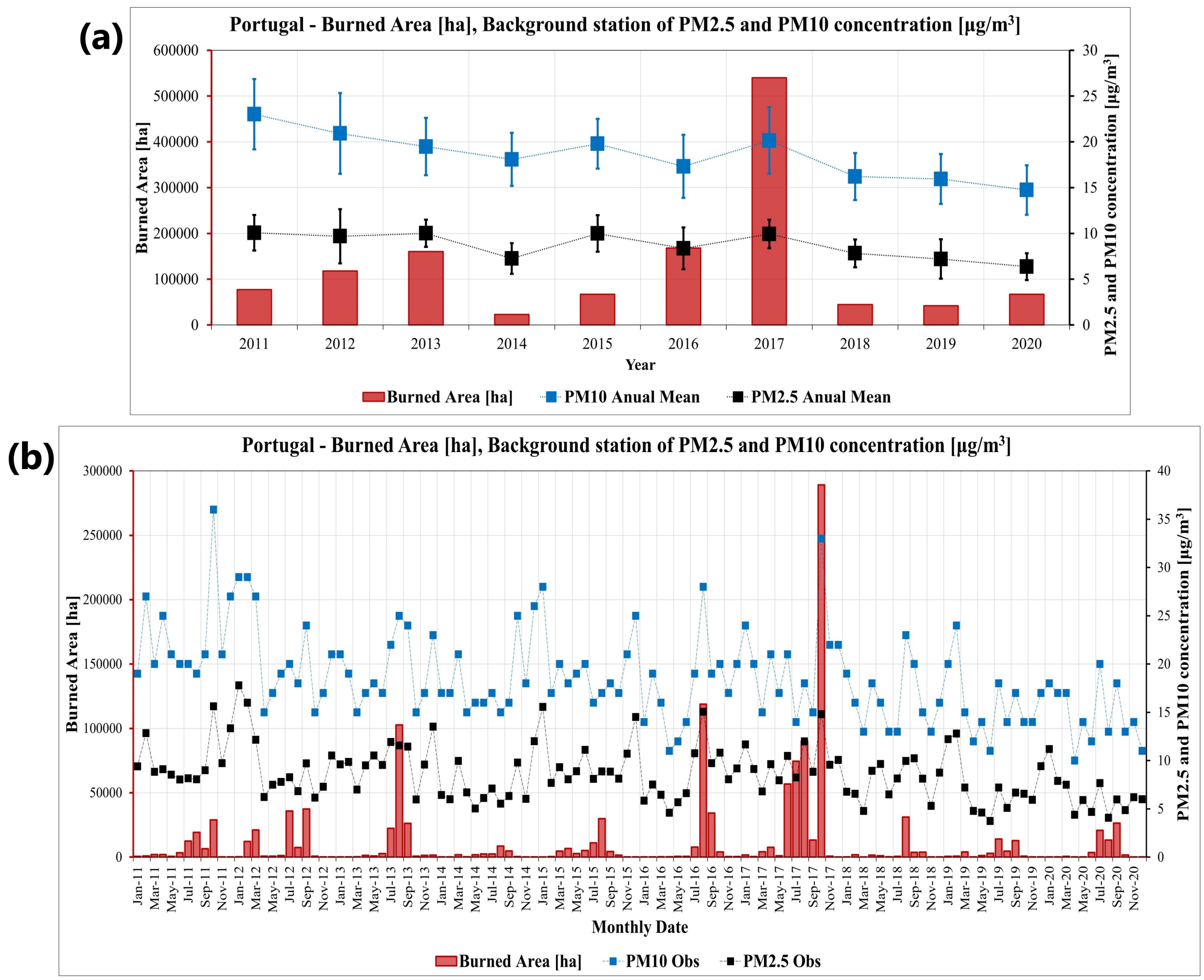


Figure. 1. Mean **(a)** yearly **(b)** monthly total BA, PM10 and PM2.5 concentration with respective standard deviations for 2011-2020 from background air quality monitoring stations in Portugal.

3.2 Association between Fire-Pollutants-Meteorological components and mortality

The result of the PCA is presented in **Figure. 2**. It shows the explained variance obtained from Fire-Pollutants-Meteorological variables data. According to the percentage of explained variance criterion, the first two principal components explain more than 68% of the variance in the dataset. The variables contribution for PC1 were PM25_CAMs (9.23 %), PM10_CAMs (9.18 %), BC_AOD_CAMs (9.02 %, PM10_Obs (8.78 %), Burned_Area (8.47 %), WHO_PM10 (8.26 %), PM2.5_Obs (8.04 %), WHO_PM25 (7.84 %), CO_CAMs (7.65 %), AOD_CAMs (7.09 %), RH_CAMs (5.94 %), T2m (3.04 %), T2m_Max_CAMs (2.38 %), Dust_AOD_CAMs (2.38 %), Pres_CAMs (1.78 %), T2m_Min_CAMs (0.90 %), WS_CAMs (0.02 %). For PC2 the variables that contributed with more than 74 % were T2m_Min_CAMs (27.28 %), T2m_Max_CAMs (21.34 %), T2m (18.94 %), and Dust_AOD_CAMs (6.49 %), the other variables had minor contribution.

The contribution of the variables in PC1 and PC2 are shown in **Figure. 2**, where it can be seen that the fire (Burned_Area), air quality (PM10_Obs; PM2.5_Obs; CO_CAMs; BC_AOD_CAMs; Dust_AOD_CAMs; PM10_CAMs; PM25_CAMs) and meteorological (RH_CAMs; Pres_CAMs; T2m; T2m_Min_CAMs; T2m_Max_CAMs) variables are highly correlated variables and strongly correlated with the first PC (represented by the horizontal axis). The high proportion of variability explained by the two-dimensional principal subspace provides solid grounds for these conclusions. In PC2, the variables with the highest correlation and statistically significant (*p-value* < 0.05) were T2m; T2m_Min_CAMs; T2m_Max_CAMs and Dust_AOD_CAMs. Therefore, PC1 and PC2 are the components that best represent the data distribution and the *scores* are the projections of the data towards the main components. In this sense, PC1 and PC2 *scores* were used as two pollutant-atmosphere interaction indexes where PC1 *score* represents pollutant-atmosphere interaction (PAI) because the pollutants and meteorological variables were strongly correlated and had higher weight PC1 formation. PC2 *score* represents the atmosphere-pollutant interaction (API) index because temperature presented higher weight than the pollutants in PC2 formation.

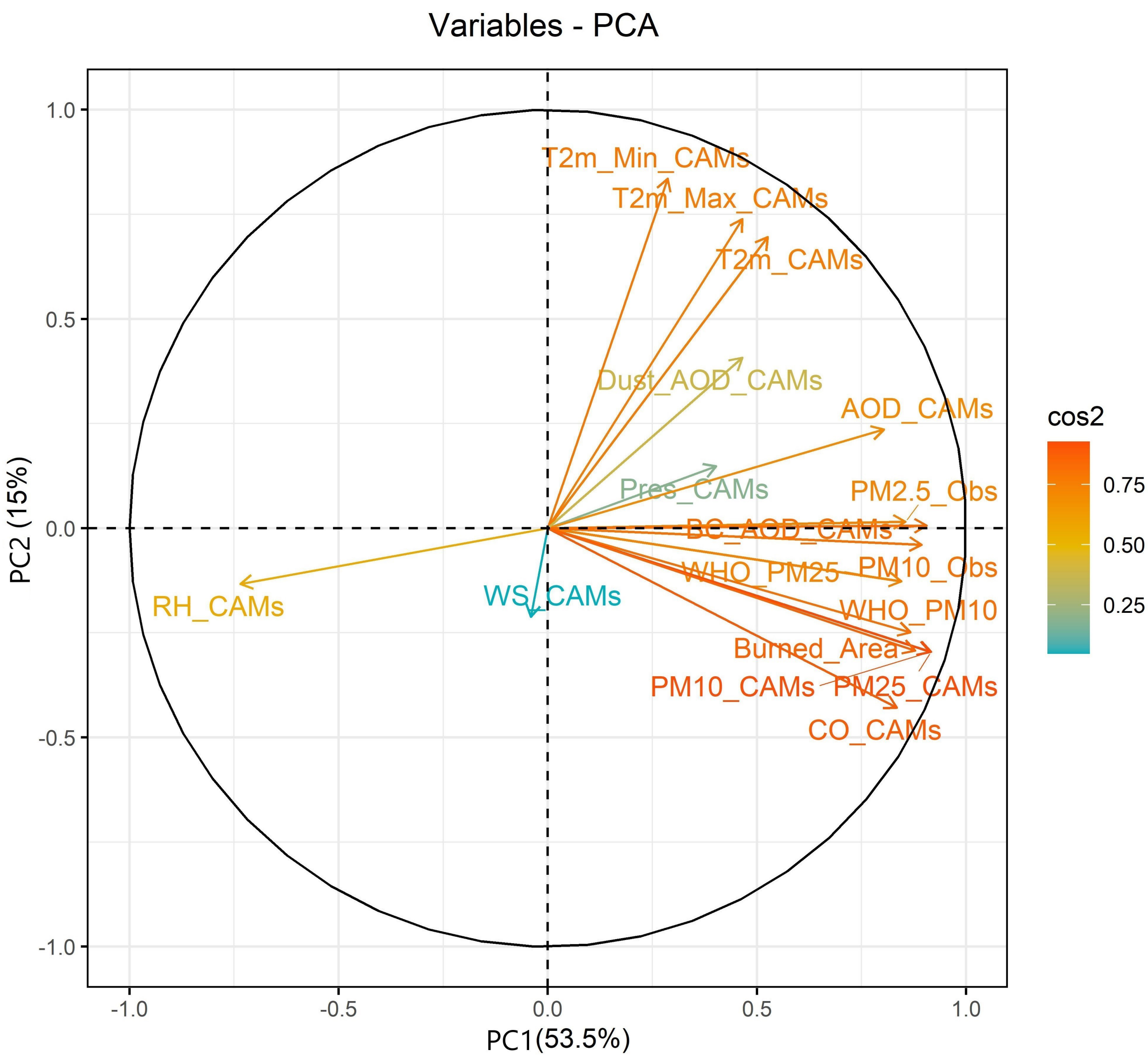
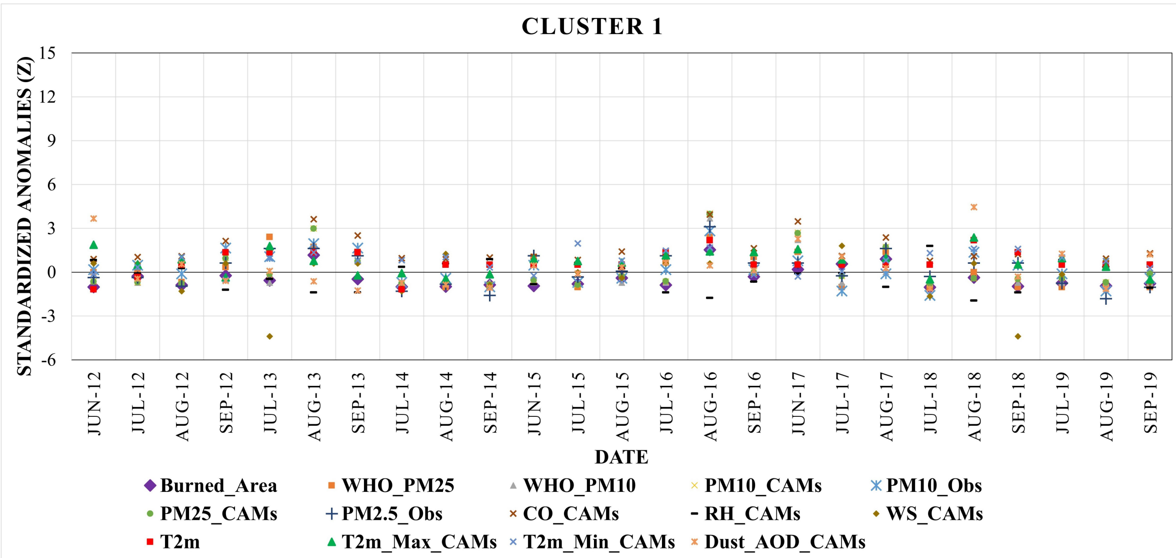


Figure. 2. Principal Component Analysis (PCA) for monthly data. Vectors indicate the contribution of each Fire-Pollutants-Meteorological variable to the respective PC1 and PC2.

K-means cluster analysis applied to PAI-API-Mortality data separated the dataset in two clusters called Cluster 1 and Cluster 2, **Figure. 3**. Cluster 1, **Figure. 3(a)**, are related to extreme atmospheric conditions and encompasses the months with more frequency of lower relative humidity and higher temperature (heat stress), and higher pollutant concentrations as well as higher wildfire activities. Cluster 2, **Figure. 3(b)**, are related to more stable atmospheric conditions characterized by the months with higher relative humidity, lower temperature, and lower pollutant concentration. October 2017 is an outstanding month, **Figure. 3(b)**. The October 2017 wildfire episode that happened on October 15-16th, 2017, can be classified as an exception. As described by Turco et al., (2019), the case of October 2017 was marked by strong and persistent southerly winds caused by the close passage of hurricane Ophelia moving northward. Together with dry vegetation and soil due to dry and high temperatures throughout 2017, this meteorological event created the conditions for the extreme fire events of October 15-16th (Turco et al., 2019).

(a)



(b)

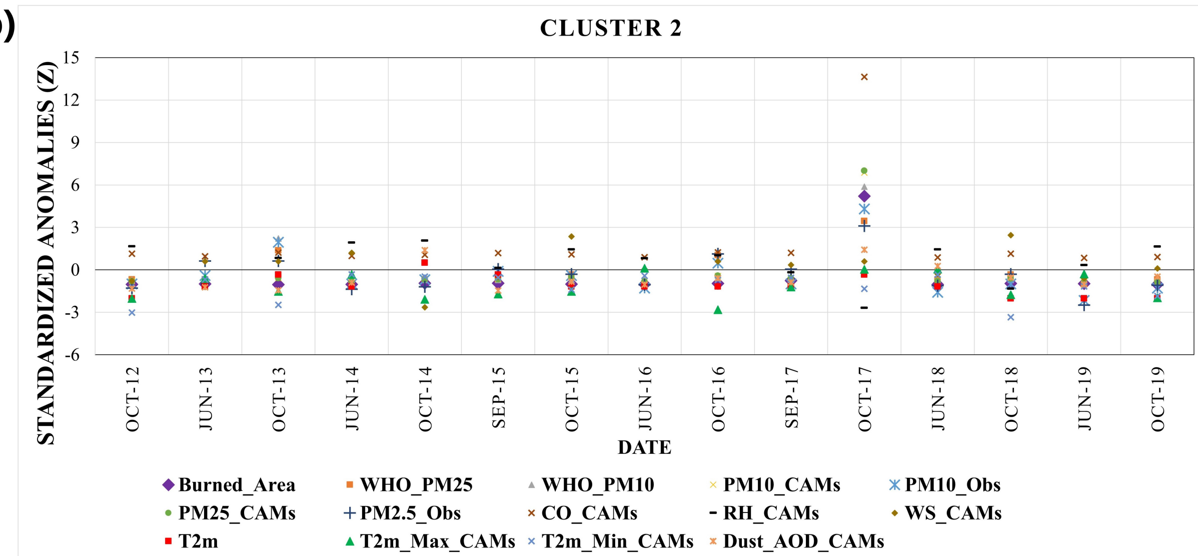


Figure. 3. The intra-annual variability of standardized anomalies (Z-scores) of the variables Burned_Area, PM10_Obs, PM2.5_Obs, CO_CAMs, PM10_CAMs, PM25_CAMs, RH_CAMs, WS_CAMs, T2m, T2m_Max_CAMs, T2m_Min_CAMs and Dust_AOD_CAMs from 2012 to 2019 **(a)** Cluster 1, **(b)** Cluster 2.

The two clusters were submitted to linear regression analysis to better understand the relationship between the health outcomes (DCS; DRS; PNEU; COPD; ASMA) and the pollutant-atmosphere interaction indexes (PAI and API). From 2012 to 2019 during the fire seasons in Portugal (June to October), the average number of casualties due to cardio-respiratory diseases (DCS; DRS; PNEU; COPD; ASMA) was 7.1 (±0.5) deaths per hundred thousand each month (Dth hd^{-1} mh^{-1}). During 2012-2019 death caused by diseases of the circulatory system (DCS) showed an average of 22.49 (±1.0) Dth hd^{-1} mh^{-1}, while diseases of the respiratory system (DRS) was 8.01 (±0.6) Dth hd^{-1} mh^{-1}, pneumonia (PNEU)3.57 (±0.5) Dth hd^{-1} mh^{-1}, chronic obstructive pulmonary disease (COPD) 1.69 (±0.2) Dth hd^{-1} mh^{-1} and Asthma (ASMA) 0.08 (±0.03) Dth hd^{-1} mh^{-1}.

A significantly positive correlation was found in Cluster 1 between DRS, PNEU and COPD, and PAI Index - ($r_{DRS} = 0.49$, *p-value* < 0.01), ($r_{PNEU} = 0.42$, *p-value* < 0.04), ($r_{COPD} = 0.44$, *p-value* < 0.03) - **Figure. 4(a)**, **(b)** and **(c)**, respectively. It means that the combination between Fire-Pollutants-Meteorology components related to extreme atmospheric conditions can be significant and positively related to respiratory mortalities due to DRS, PNEU and COPD. These results were considered statistically significant, *p-value* < 0.05. For ASMA, DCS and PAI Index, the correlation was positive but not significant (*p-value* > 0.05), **Figure. 4(d)** and **(e)**. Nevertheless, linear regression applied as a diagnostic method to detect cause of death patterns during the fire season which indicate that deaths due to ASMA (r_{ASMA} = 0.17, *p-value* > 0.05) and DCS (r_{DCS} = 0.17, *p-value* > 0.05) also tends to increase because of extreme atmospheric conditions related to Fire-Pollutants-Meteorology.

Figure. 4(f) shows a positive correlation between DCS and API index (r_{DRS} = 0.45, *p-value* < 0.05). API index in the PCA was correlated with temperature (T2m; T2m_Min_CAMs; T2m_Max_CAMs) and Dust_AOD_CAMs. The linear regression in Cluster 1 indicates that deaths due to DCS were positively correlated with high temperatures during the fire seasons, especially when minimum and maximum temperature are high, heat stress. The results were considered statistically significant, *p-value* < 0.05. API are also negatively correlated with wind speed and relative humidity. The association between increased temperature (heat stress) and adverse health outcomes such as cardiovascular and respiratory diseases has been extensively studied (Baccini et al. 2008; Lin et al. 2009; Lin et al. 2013; Yang et al. 2012; Vitolo et al., 2019). The combination of wildfire smoke exposure with heat stress due to high temperatures can amplify cardio-respiratory mortality outcomes contributing to increase the total disease burden. API index was also related to dust aerosols. Dust aerosols play an important impact in Europe due to dust storms from the Sahara Desert. Studies show that cardiovascular hospitalization increases after African dust storm episodes (Middleton et al., 2008; Neophytou et al., 2013).

A positive near zero correlation coefficient between API and respiratory deaths outcomes (DRS, PNEU, COPD and ASMA) was found with *p-value* > 0.05, 0.06 < r < 0.22 (see **Figure. S1** in Supporting Information **S1**). The relationship between API and respiratory diseases during Portugal wildfire seasons (2012-2019) were statistically not significant in the Cluster 1, but the correlation was positive for DRS and COPD.

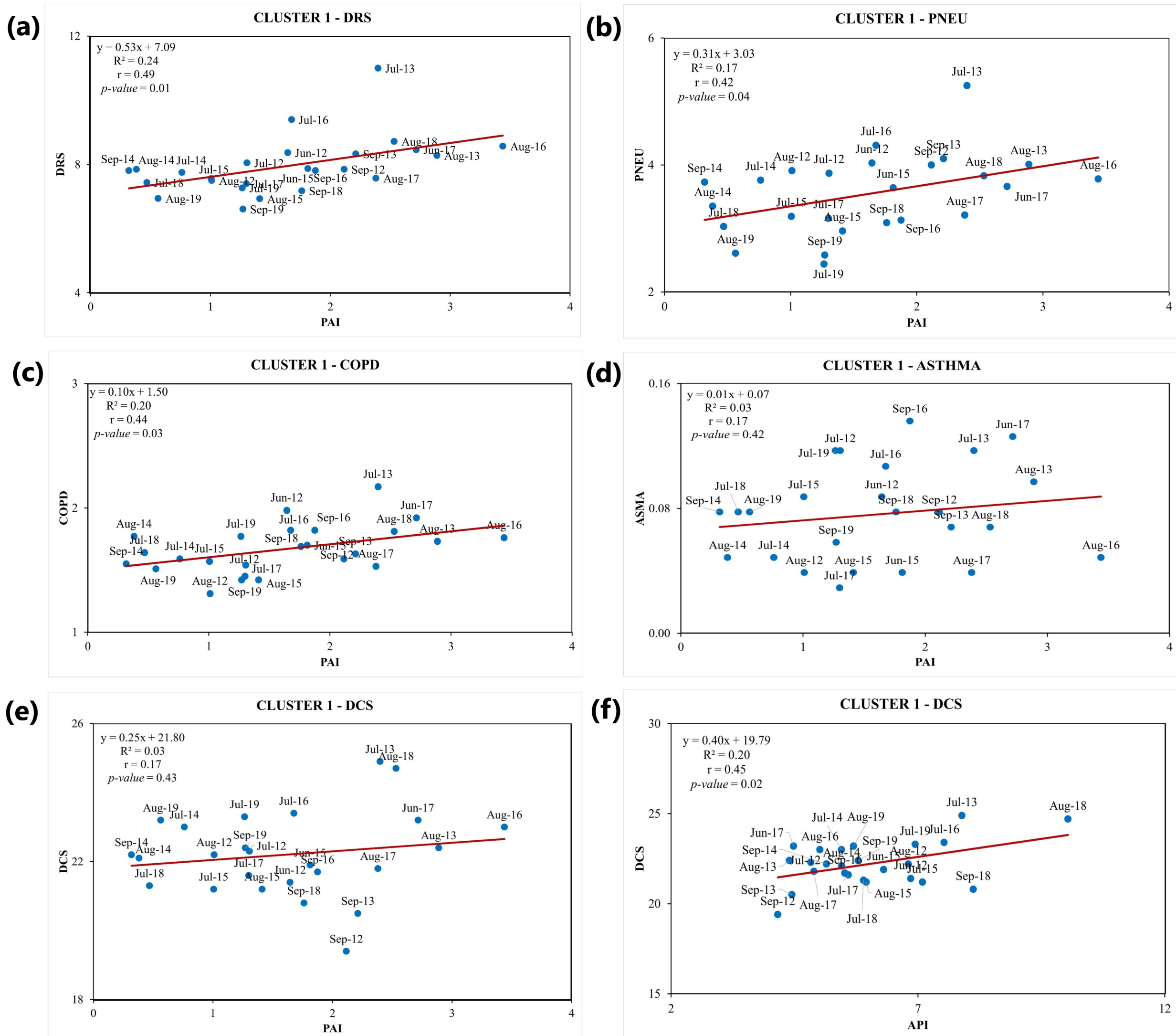


Figure. 4. Linear regression analysis between health outcomes (DRS, PNEU, COPD, ASMA, DCS) and pollutant-atmospheric-interaction indexes (PAI and API) for Cluster 1: **(a)** DRS x PAI; **(b)** PNEU x PAI; **(c)** COPD x PAI; **(d)** ASMA x PAI; **(e)** DCS x PAI; and **(f)** DCS x API.

Cluster 2, related to the months with cleaner air and stable atmospheric conditions near the surface, present a weak correlation between PAI-API indexes and health outcomes, as shown in **Figure. 5**. These results suggest that the Fire-Pollutants-Meteorology during these months was statistically not significant (p-$value > 0.05$) to increase the number of deaths related to DRS, PNEU, COPD, ASMA and DCS. Correlation coefficient varied between -0.24 < r <

0.18, *p-value* > 0.05. As explained earlier, the October 2017 wildfire episode was classified as an exception. This short but extreme weather event was responsible for pushing the line of the linear regression causing a shift. Without October 2017, Cluster 2 tends to have a negative or near zero correlation between PAI-API indexes and health outcomes. These results are very important to show how difficult it is for linear models to deal with outliers but also to show how difficult it is to represent extreme weather events. In the vicinity of climate change the extreme weather events, such as the October 2017 wildfire episode in Europe will become more frequent.

Figure. S2 in Supporting Information **S2** shows that correlation coefficient between $-0.37 < r < 0.11$ API and cardiorespiratory deaths outcomes (DRS, PNEU, COPD, ASMA and DRS) with *p-value* > 0.05). The relationship between API and respiratory diseases during Portugal wildfire seasons (2012-2019) were statistically not significant in the Cluster 2, related to cleaner and more stable meteorological conditions.

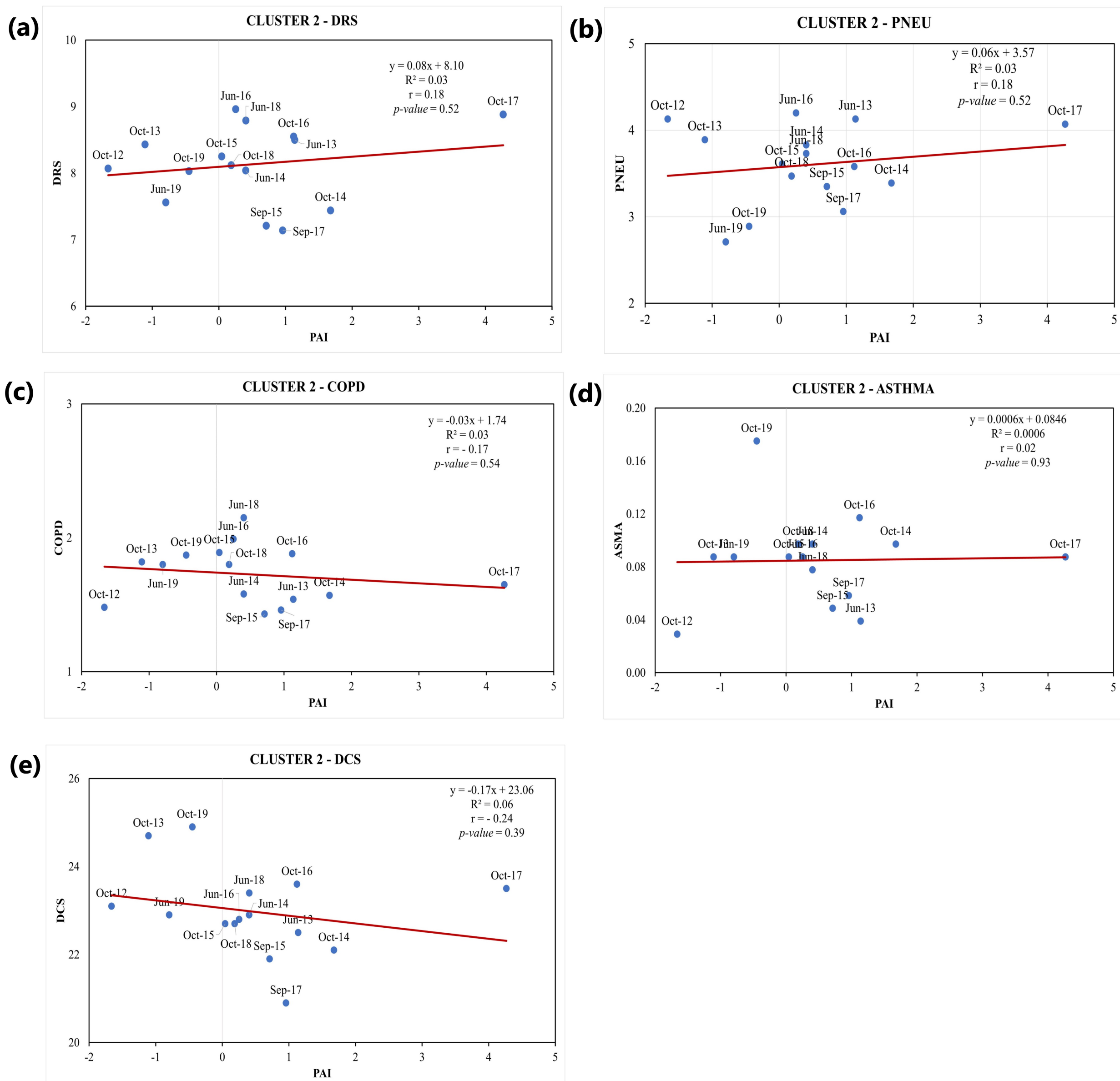


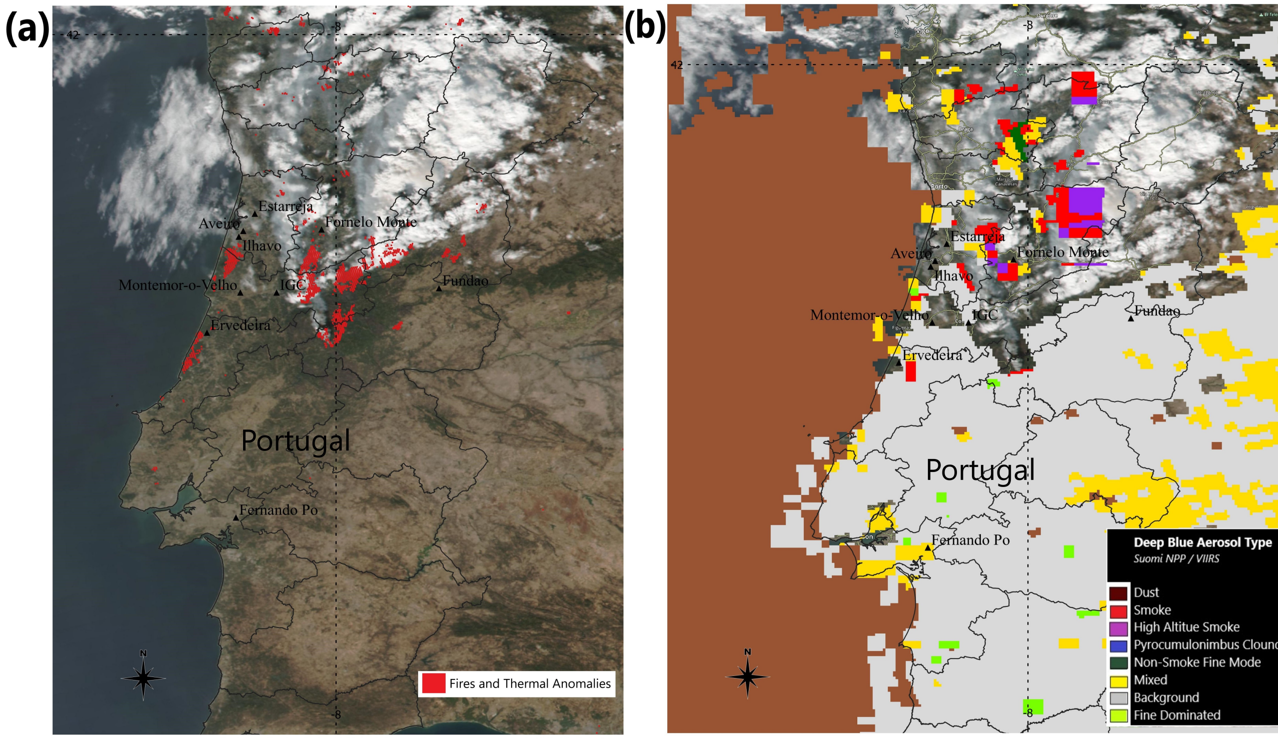
Figure. 5. Linear regression analysis between health outcomes (DRS, PNEU, COPD, ASMA, DCS) and pollutant-atmospheric-interaction indexes (PAI and API) for Cluster 2: **(a)** DRS x PAI; **(b)** PNEU x PAI; **(c)** COPD x PAI; **(d)** ASMA x PAI and **(e)** DCS x PAI.

3.3 PM10, PM2.5 and BC forecasting during 2017 wildfire episode using HYSPLIT model

Figure. 6(a) shows the October 15th-16th, 2017, wildfire episode and associated smoke loading over Portugal. Black triangles refer to the locations of the background APA stations used in this work. MODIS fire and thermal anomalies from Terra (MOD14) and Aqua (MYD14) detection algorithm (Giglio et al., 2016) shows spatial distribution of active fire and thermal anomalies measured by satellite observations and smoke plumes from fires for the episode. **Figure. 6 (b)** presents the spatial distributions of VIIRS Deep Blue Aerosol Type for October 15th, 2017. VIIRS data includes retrieved AOD and Ångström exponent, as well as dust, urban/industrial (non-smoke fine mode), smoke, high-altitude smoke, mixed, and background aerosols (Hsu et al., 2016). VIIRS products for the fire episode showed heavy smoke plumes at low and high-altitude during the fire event on October 15th, 2017. This fire outbreak (October 15th – 16th, 2017) was declared the largest wildfire in recorded history in Portugal. This episode happened during one Sahara dust event in the same days, which also contributed to unhealthful air quality in some regions of Portugal. **Figure. 6 (c)** shows an aerosol index layer from the Suomi NPP/Ozone Mapping and Profiler Suite (OMPS) for October 15th. It is possible to identify high amounts of aerosol related to smoke from wildfire in the atmosphere near Fornelo Monte, Portugal. The Aerosol Index reaches the range of five over some regions in Portugal, **Figure. 6 (c),** which indicates heavy concentrations of aerosols that could impact human health.

Daily concentrations of PM10 and PM2.5 are presented in **Figure. S3 (a)** and **(b)** for the years of 2011-2020. The black line with triangles shows the daily PM10 and PM2.5 for the year 2017, which largely increased during the October 15th-16th of 2017 fire outbreak. For these days, the PM10 and PM2.5 daily concentrations were above WHO 2005 guidelines (WHO, 2005) as well as above the new WHO 2021 guidelines (WHO, 2021). **Figure. S3 (a)** and **(b)** in Supporting Information **S3** also showed that common PM10 and PM2.5 concentrations overpass WHO guidelines during fire seasons. Considering that background stations in this study were relatively far from the wildfire's occurrences, it reveals the impact of the regional transport of pollutants far beyond the source emissions.

Regarding PM10 concentration in atmosphere, Tarín-Carrasco et al., (2021) found a significant positive correlation between burned area and PM10 for some regions of Portugal, as well as a significant association between PM10 concentrations and mortality, these being apparently related to large wildfires in some of the regions occurring during the fire season (June–July–August–September) from 2001 to 2016.


(a)
(b)
Estarreja
Aveiro
Ilhavo
Fornelo Monte
Montemor-o-Velho
IGC
Fundao
Ervedeira
Portugal
Fernando Po
Fires and Thermal Anomalies
Deep Blue Aerosol Type
Suomi NPP / VIIRS
Dust
Smoke
High Altitue Smoke
Pyrocumulonimbus Clound
Non-Smoke Fine Mode
Mixed
Background
Fine Dominated

(c)

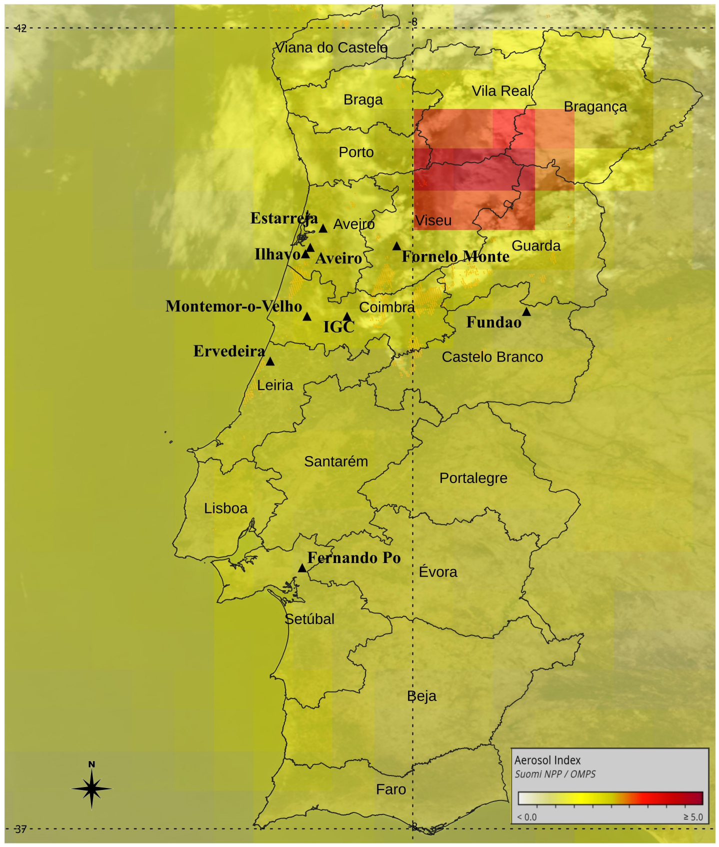
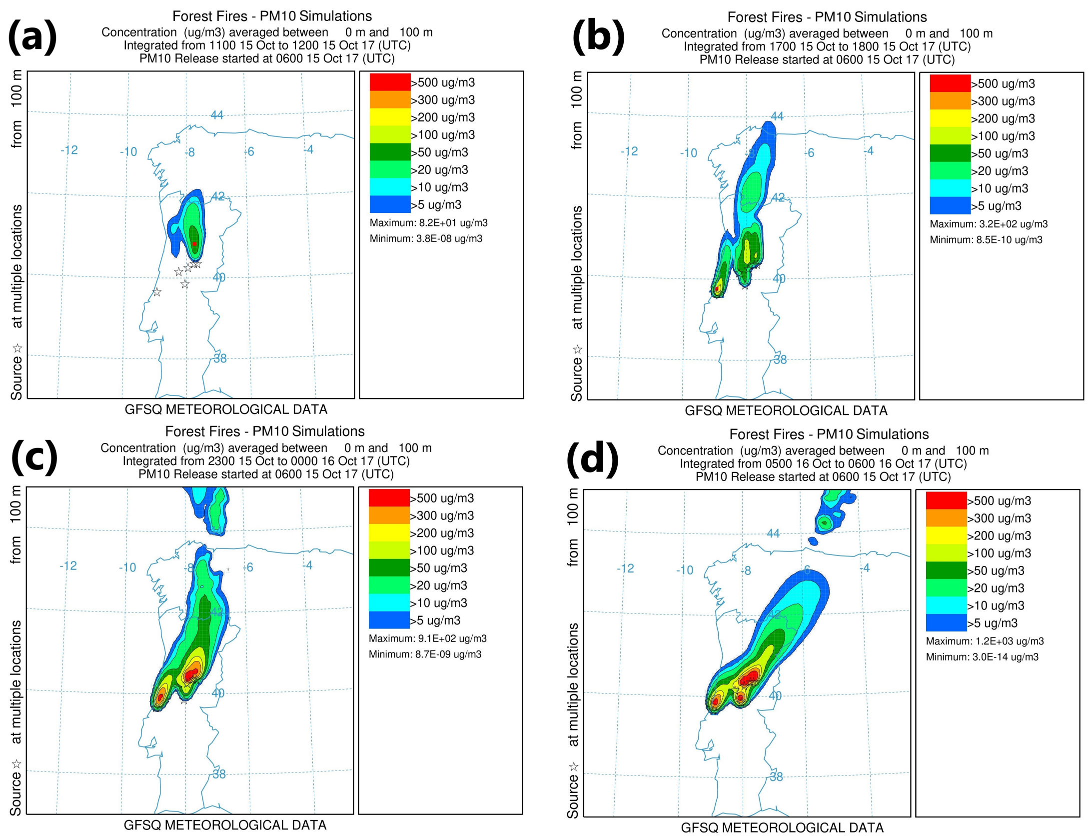


Figure. 6. **(a)** October 15th-16th, 2017**,** MODIS Fire and Thermal Anomalies, **(b)** VIIRS Deep Blue Aerosol Type on October 15th, and **(c)** OMPS Aerosol Index layeron October 15th, black triangles are PM10 and PM2.5 APA background/rural stations locations used in this study. The VIIRS Deep Blue Aerosol Type layer provides information related to the aerosol composition over land and ocean. OMPS Aerosol Index layer indicates the presence of ultraviolet (UV)-absorbing particles in the air (aerosols) such as dust and smoke plumes.

HYSPLIT model was run for October 15-16th 2017 in the forward mode. The HYSPLIT model generated atmospheric fields at 1-h intervals (near-surface pollutant concentrations are time dependent) with hourly fire emission rates starting from identified GFAS fire emission data to produce the 48-h atmospheric dispersion forecast for PM10, PM2.5 and BC, separately. To facilitate an estimation of the relative contribution from wildfires, the pollutant concentration was averaged for the vertical layer immediately above the surface between 0 and 100 m, assuming that there will be uniform and rapid vertical mixing in this lowest turbulent layer.

The forward atmospheric dispersions from the wildfire sources for PM10 and

PM2.5 are presented in **Figure. 7.** At 1200h and 1800h UTC on October 15th and 0000h and 0600h UTC on October 16th, 2017. The dispersion pattern is the same for PM10 and PM2.5 whereas the magnitude alone differs by relative fraction. The general pattern of dispersion from wildfire sources is noted to be towards northwards, under the prevailing wind flow of south/southwest relative to the observation. The pollutant dispersion is similar to a plume expanding in the forward direction following the wind flow. The dispersion pattern was influenced by hurricane Ophelia that produced strong winds all over Portugal (Potes et al., 2018; Turco et al., 2019; Augusto et al., 2020) and dispersed wildfire pollutants and dust from Sahara not only in Portugal but across all western European countries including Spain, France, Belgium, the Netherlands, and the UK.



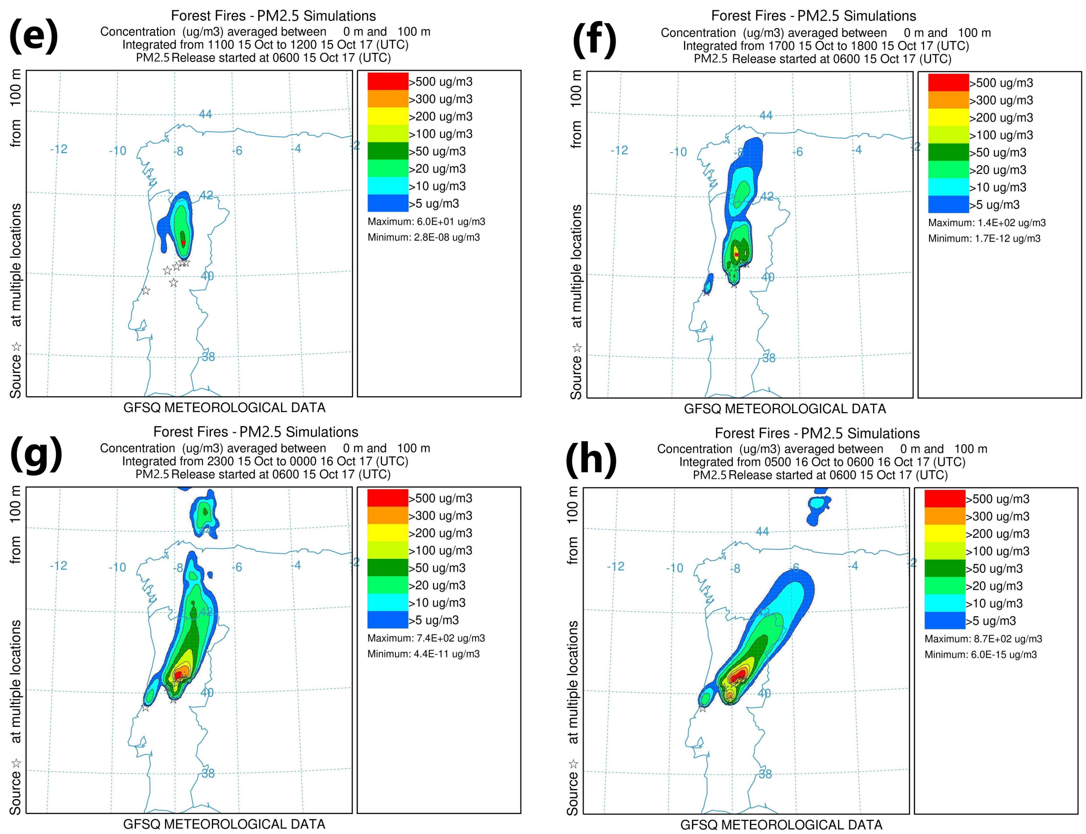


Figure. 7. HYSPLIT-simulation averaged between 0 and 100 m levels at 1200 (UTC), 1800 (UTC) on 15 Oct and at 0000 (UTC), 0600 (UTC) on 16 Oct 17, 2017, sourced from the six identified wildfire region by GFAS **(a)**, **(b)**, **(c)**, **(d)** PM10 concentration (µg/m^3); **(e)**, **(f)**, **(g)**, **(h)** PM2.5 concentration (µg/m^3).

Figure. 8 shows another important pollutant simulated with the HYSPLIT model for the October 15-16th fire outbreak, Black Carbon (BC). BC is an important component of ambient fine particulate matter (PM2.5) emitted by incomplete combustion engines (especially diesel), residential burning of wood and coal, power stations using heavy oil or coal, field burning of agricultural wastes, as well as forest and vegetation fires (Janssen, 2012; Malico et al., 2017). **Figure. 8** Shows that the BC dispersion pattern is similar to PM10 and PM2.5. Regarding BC concentration simulated by HYSPLIT, BC reached values above 20 g/m^3 over Coimbra, Viseu, Guarda, Vila Real and Bragança district, in continental Portugal. **Figure. 8** is in line with **Figure. 6(c)** that shows higher aerosol index in the same area.

Several epidemiological studies have shown the potential impact on health outcome (all-cause and cardiovascular mortality, and cardiopulmonary hospital admissions) related to long and short-term exposure to BC (Bell et al., 2009;

Janssen et al., 2012; Hua et al., 2014; Atkinson et al., 2015; Wang et al., 2021). The World Health Organization Regional Office for Europe found that effect estimates from both short and long-term studies are much higher for BC compared to PM10 and PM2.5 when the particulate measures are expressed per g/m^3 (Janssen et al., 2012). Based on observational data, it has been estimated that BC comprises approximately 5–10% of average urban PM2.5 mass in the US (EPA, 2012).

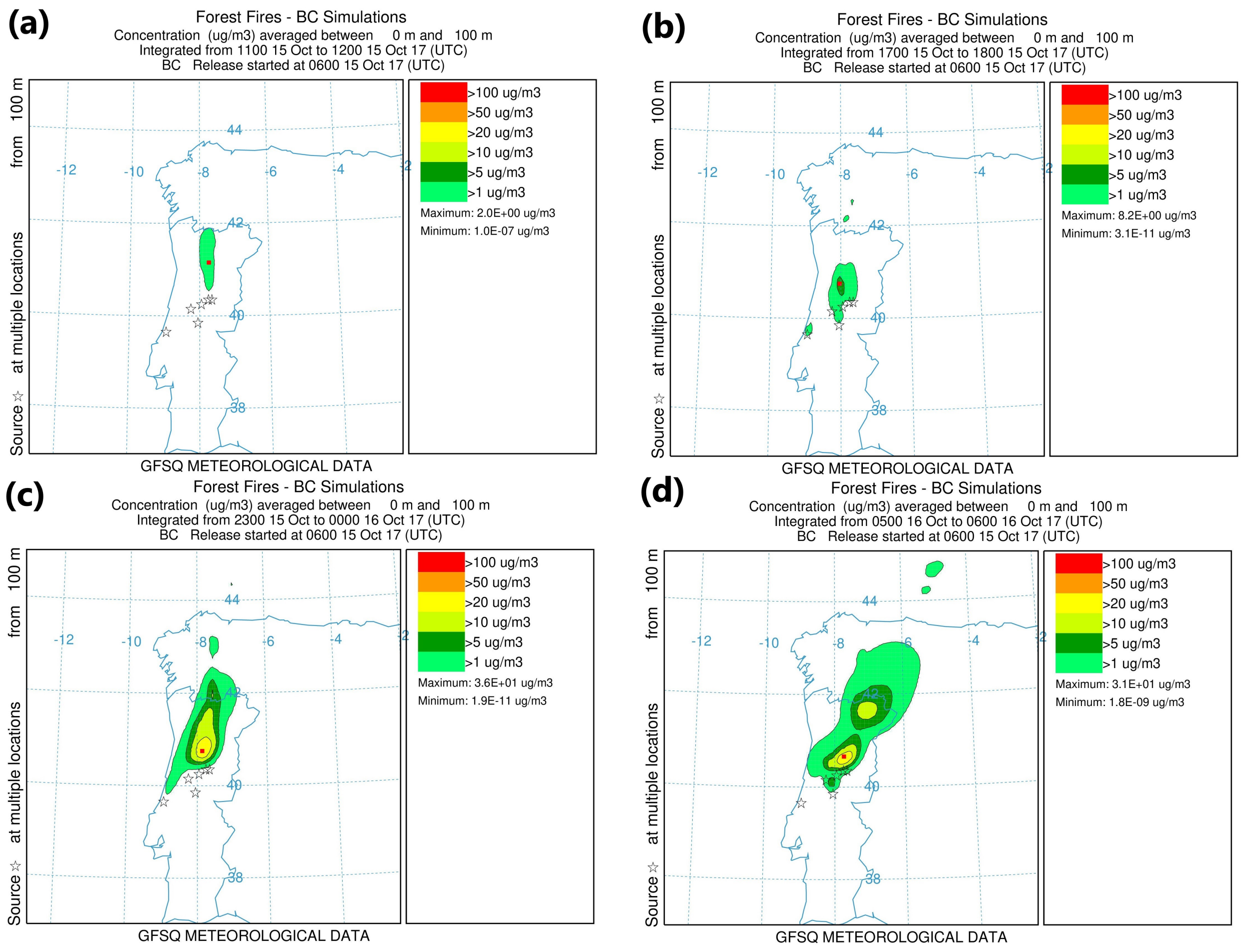
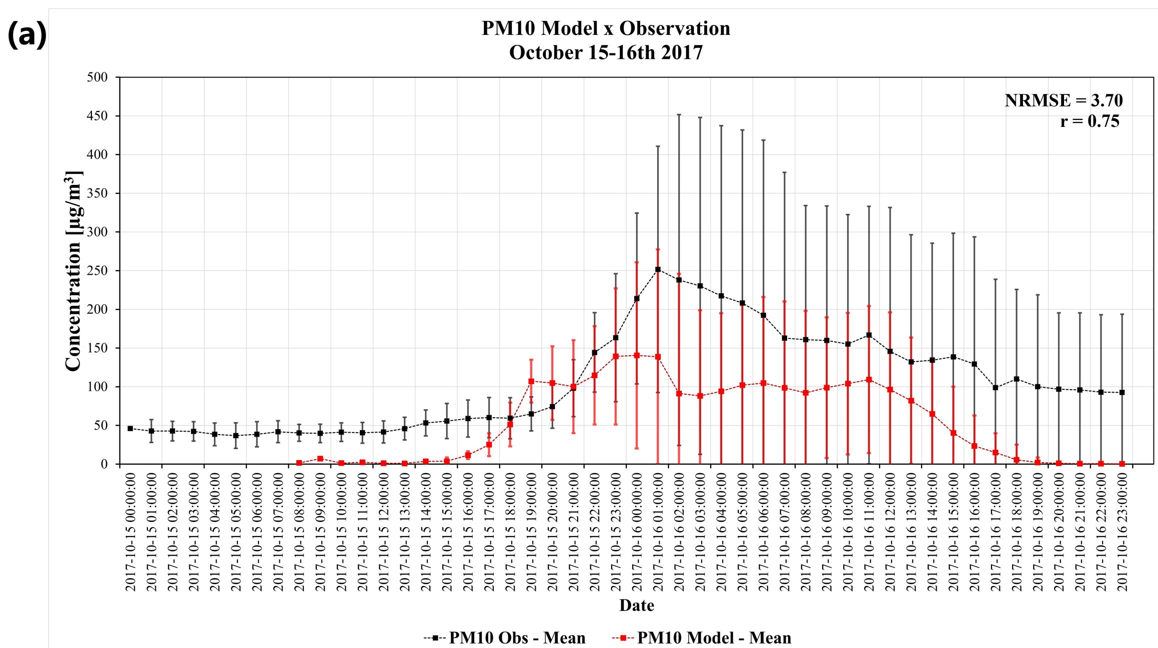


Figure. 8. HYSPLIT-simulation averaged between 0 and 100 m levelsat 1200 (UTC), 1800 (UTC) on 15 Oct and at 0000 (UTC), 0600 (UTC) on 16 Oct 17, 2017, sourced from the six identified wildfire region by GFAS **(a)**, **(b)**, **(c)**, **(d)** BC concentration (µg/m^3).

Comparison with PM10 and PM2.5 observations across Portugal is complicated by the sparse PM10 and PM2.5 background stations network and the fact that the anthropogenic contribution is not modeled by HYSPLIT system. Despite that, the model presented a good performance and promising results. **Figure. 9(a)** compares measured PM10 concentrations with HYSPLIT results over the 9 background monitoring stations at Portugal obtained from the APA dataset for each hour during October 15-16th 2017 fire episode. The model tends to underestimate the concentrations nonetheless it captures the gross features of the hourly relative variation in the PM10 levels near surface, with increasing concentrations from 1500 UTC on October 15th to 1000 UTC on October 16th, a sharp decrease toward the end of the period from 1400 UTC on October 16th, 2017, **Figure. 9(a)**. The PM10 concentration at the background stations were way above WHO guidelines (2005 as well as 2021 guidelines).

Since the anthropogenic emissions were not included in the HYSPLIT simulation, the Normalized Root Mean Square Error (NRMSE) was used to facilitate the comparison between model and observations, as well as Pearson' correlation coefficient (r). The statistical indexes for PM10 indicate that at the surface level, the model reproduced the behavior of observed PM10 concentrations at the background stations during October 15-16th fire episodes, which is indicated by the low NRMSE and r = 0.75, **Figure. 9(a)**.



(b)

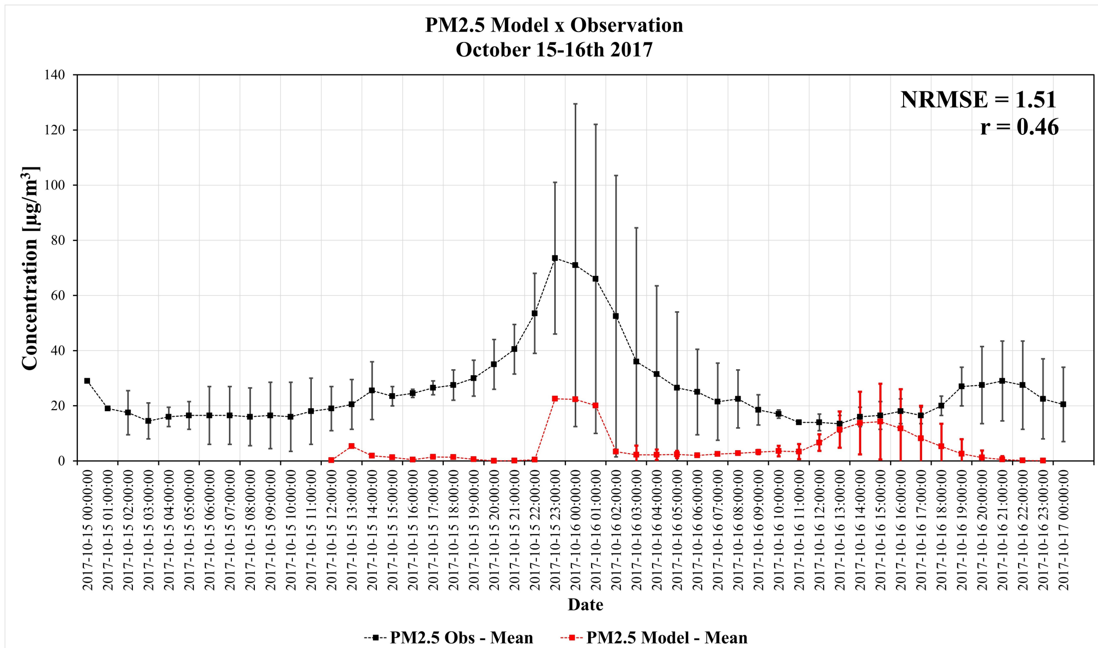


Figure. 9. **(a)** PM10 and **(b)** PM2.5 concentrations for October 15–16th, 2017 measured by APA background stations and modeled by Hysplit. Averaged PM10 and PM2.5 concentration measured by background APA stations (black) and HYSPLIT performance (red) are given.

In the case of PM2.5, only two background stations measured hourly PM2.5 concentration during the October 15-16th 2017 fire episode, the stations of Fundão and Estarreja. In this regard, **Fig. 9(b)** presents the comparison of hourly averaged PM2.5 concentration in these two stations. Modeled PM2.5 concentration is lower than observations but reproduced the behavior of the variations at the background station (NRMSE = 1.51 and r = 0.46) during the October 15-16th, 2017. The background station captured a PM2.5 concentration peak between 2200 UTC on October 15th and 0200 UTC on October 16th, 2017. This PM2.5 concentration peak was also captured by HYSPLIT indicating that the model captures the gross features of the hourly relative variation near the surface.

Model simulation showed concentration of PM10 over Coimbra, Viseu, Guarda, Vila Real and Bragança district varied between 0 and 1200 (g/m^3). For PM2.5 varied between 0 and 872 (g/m^3) while for BC the variation was between 0 and 31 (g/m^3). The background APA station marked a PM10 peak of 1000 (g/m^3) at Ervideira, Aveiro 214 (g/m^3), Estarreja 200 (g/m^3), Fundão 296 (g/m^3) between 0000 (UTC) and 0900 (UTC) on October 16th, 2017. The North of Portugal was the most affected region by the fires and their socio-environmental impacts. According to Augusto et al. (2020) the population was exposed on average to an additional load of PM10 during seven smoky days (three with dust) of October 2017 wildfire episode. The results suggested that PM10 had a significant effect in the cardiorespiratory mortalities during the month of October 2017.

Individually comparing model results with observation stations, the model performance may decrease depending on the location of the monitoring station, distance from the fire emission source and atmospheric conditions. Besides all uncertainty caused by not including anthropogenic emissions due to model limitations, HYSPLIT uncertainty can increase due to smoke from smaller fires not captured by satellites. These fires can also contribute to worsen air quality near fire areas. Another uncertainty point is that fire hotspots are captured by satellites only during the overpass (typically before the afternoon), so fires that ignite after the overpass are not included in the HYSPLIT simulations.

4. Concluding remarks

We found consistent evidence of association among Fire-Pollutants-Meteorology components and cardio-respiratory mortality in Portugal during wildfire seasons (June-October), specifically DCS, DRS, PNEU, COPD and ASMA. We observed increases in cause-specific mortality in months with extreme atmospheric conditions (low relative humidity, high temperature, high pollutant concentrations and, as related, high wildfire activities). With climate change, extreme weather events and uncontrolled wildfires tend to become more frequent, consequently, morbidity and mortality tend to increase if mitigation actions are not taken. Months inside the wildfire season with stable atmospheric conditions and cleaner air present lower cardio-respiratory mortality rates. Understanding the health impacts of Fire-Pollutants-Meteorology together can help society and the decision makers to be better prepared for extreme weather events and ensure that health care services are equipped to mitigate the health impacts in the population in the aftermath of a wildfire season.

The HYSPLIT model was used with GFAS fire emission to forecast PM10, PM2.5 and BC during the October 15-16th, 2017, wildfire outbreak in Portugal. Model results fit the behavior of the observations and presented a positive correlation of $r_{PM10} > 0.7$ and $r_{PM2.5} > 0.4$. Results are mostly consistent with general expectations based on the characteristics and behavior of fire events. Future work is warranted to test the HYSPLIT forecast on more wildfire cases with different fire sizes, emission strengths, seasons, and locations, in order to see whether the model forecast approach is suitable for an operational usage. With more computational power, numerical forecast could also be performed using more complex chemistry transport models such as the Weather Research and Forecast with Chemistry (WRF-Chem) model. Wildfire exposure assessment would benefit from forecasting smoke during wildfire season, and it can be a useful tool for public health policymaking.

Declaration of competing interest

The authors declare no competing interests.

Data Availability Statement

Data sets for this study are publicly available online. surface observational air pollution data obtained from the Online Database on Air Quality (QualAr) of the Portuguese Environment Agency (Agência Portuguesa do Ambiente (APA), at https://qualar.apambiente.pt). Mortality data for Portugal was provided by the National Institute of Statistics (INE) (INE, https://www.ine.pt/). Burned Area data were obtained from the Portuguese Institute for Conservation of Nature and Forests (https://www.icnf.pt/) and Portuguese Institute of the Sea and Atmosphere (https://www.ipma.pt/pt/index.html). The ECMWF data is available through the Copernicus Atmosphere Monitoring Service (CAMS, https://ads.atmosphere.copernicus.eu) on behalf of the European Union including CAMS-Reanalysis. Thermal Anomalies/Fire from MODIS, VIIRS, Suomi-NPP data are available at NASA Worldview (https://worldview.earthdata.nasa.gov/).

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Author contributions

Ediclê de Souza Fernandes Duarte: Conceptualization, Investigation, Methodology, Formal analysis, Wrote the manuscript. Vanda Salgueiro: Conceptualization, Investigation, Methodology, Writing - review & editing. Maria João Costa: Conceptualization, Investigation, Methodology, Writing - review & editing. Paulo Sérgio Lucio: Conceptualization, Investigation, Methodology, Writing - review & editing. Daniele Bortoli: Investigation, Writing review & editing. Miguel Potes: Investigation, Writing - review & editing. Rui Salgado: Investigation, Writing - review & editing.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi...

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