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Interactions between the terrestrial biosphere and atmosphere during droughts and heatwaves: impact on surface ozone over Southwestern Europe

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Abstract. At high concentration, tropospheric O_3 deteriorates air quality, inducing adverse effects on human and ecosystem health. Meteorological conditions are key to understand the variability of O_3 concentration, especially during extreme weather events. They modify the photochemistry activity and the vegetation state. An important source of uncertainties and inaccuracy in simulating surface O_3 during droughts and heatwaves is the lack of interactions between the biosphere and the troposphere. Based on the biogenic emission model MEGANv2.1 and the chemistry-transport model CHIMERE v2020r1, the first objective of this study is to assess the sensitivity of biogenic emissions, O_3 dry deposition and surface O_3 to biomass decrease and soil dryness effect (using several configurations) during the extremely dry summer 2012. Secondly, this research aims at quantifying the variation of observed (EEA's air quality database, 2000-2016) and simulated (CHIMERE, 2012-2014) surface O_3 during summer heatwaves and agricultural droughts that have been identified using the Percentile Limit Anomalies (PLA) method. Our sensitivity analysis shows that soil dryness is a key factor during drought events, decreasing considerably the C_5H_8 emissions and O_3 dry deposition velocity. This effect has a larger impact than the biomass decrease. However, the resulting effect on surface O_3 remains limited.

Based on a cluster approach using the PLA indicator, we show that observed O_3 concentration is on average significantly higher during heatwaves (+18$\mu g/m^3$ in daily maximum) and droughts (+9$\mu g/m^3$) compared to normal conditions. Despite a difference of several $\mu g/m^3$, CHIMERE correctly simulates the variations of O_3 concentration between the clusters of extreme events. The overall increase of surface O_3 during both heatwaves and droughts would be explained by O_3 precursor emission enhancement (in agreement with $HCHO$ satellite observations), O_3 dry deposition decrease and favourable weather conditions. However, we simulated a decrease of C_5H_8 emissions (in agreement with $HCHO$ observations) during droughts not accompanied by a heatwave, resulting in a non-significant difference of surface O_3 compared to normal conditions (from both observations and simulations).

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Finally, we stress that considerable uncertainties characterize our simulated surface-troposphere interactions. Multi-year flux measurements would contribute to better assess the model performance. Nevertheless, we emphasize the need for a more dynamical representation of interactions between vegetation, hydrology, meteorology and atmospheric chemistry in models in order to improve the simulation of summer O_3.

1 Introduction

Tropospheric ozone (O_3) plays a critical role in maintaining the oxidative capacity of the troposphere. However, as a high oxidant, it also deteriorates air quality at high concentrations, inducing adverse effects on human and ecosystem health. Both short and long-term O_3 exposure significantly increases the risk of morbidity and mortality from cardiovascular or respiratory causes (e.g. Jerrett et al., 2009; Nuvolone et al., 2018). It also causes visible damage on plant epidermis as well as photosynthesis inhibition, seriously threatening vegetation growth (Anand et al., 2021) and crop yields (De Andrés et al., 2012). Tai et al. (2014) estimate that O_3 pollution enhanced by global warming (RCP8.5) could lead to a global crop yield reduction by 3.6% in 2050.

Tropospheric O_3 is a secondary air pollutant formed by photochemical reaction chains initiated by the oxidation of volatile organic compounds ($VOCs$) in the presence of nitrogen oxides (NO_x). NO_x/VOC emission ratio determines the chemical regime of O_3 production at local scale in a highly nonlinear relationship (Jacob, 1999). Methane (CH_4) and carbon monoxide (CO) are also O_3 precursors in background conditions.

Meteorological conditions are a key driver for O_3 formation and transport (Mertens et al., 2020). Indeed, O_3 production is favoured by warm and sunny conditions, and O_3 peaks therefore occur mainly during summer. The overall objective of this paper is to quantify the variation of surface O_3 concentrations over the Euro-Mediterranean area during extreme weather events of heatwaves and droughts.

The Euro-Mediterranean region is often affected by both heatwaves (Russo et al., 2015) and droughts (Spinoni et al., 2015). The intensity and frequency of such events have increased over the last decades in the region. According to projections based on climate models, these trends could last and worsen over the 21st century (Perkins-Kirkpatrick and Gibson, 2017; Spinoni et al., 2018).

Heatwaves are often associated with high O_3 concentration. For example, Vautard et al. (2005) show that the persistent heatwave of August 2003 led to high O_3 concentration over the Western Europe for several days (daily maximum above 150$\mu g/m^3$, spatially averaged). Jaén et al. (2021) report hourly values of O_3 exceeding 250$\mu g/m^3$ during a shorter heatwave (28-29 June 2019) at local scale (Barcelona). These values are well above the European Union (EU) recommendation of a 8h-average maximum of 120$\mu g/m^3$.

Porter and Heald (2019) highlighted that despite a strong correlation between surface temperature and O_3 concentration identified in Europe, this relationship is nonlinear and complex as it depends on several mechanisms including precursor emissions, reaction rates and lifetimes in the atmosphere. High temperatures favour emissions of biogenic $VOCs$ ($BVOCs$), which can induce an increased formation of O_3 in the case of a NO_x-limited regime as many areas in the Mediterranean basin (Richards

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et al., 2013). Persistent and intense solar radiation also allows long photochemical episodes (Jaén et al., 2021). Heatwaves in the Mediterranean being often related to a blocking situation of atmospheric systems, stagnant anticyclonic conditions lead to the accumulation of both precursors and O_3 (Otero et al., 2021). Moreover, extreme temperature and high O_3 concentration can cause stomatal closure reducing the dry deposition of O_3, as highlighted by Gong et al. (2020) with the O_3-vegetation feedback.

Droughts also significantly impact atmospheric chemistry through biosphere and atmospheric cycle perturbation (Wang et al., 2017). Drought impact on air pollutants and more specifically O_3 is difficult to investigate for several reasons. It is firstly related to the characterization of droughts. Due to its multiscalar character, drought events can last on timescales ranging from days to months, making their extent difficult to assess (Vicente-Serrano et al., 2010). Moreover, drought definition depends on the type considered (Svoboda and Fuchs, 2016). Meteorological droughts correspond to a rainfall deficit or an excess of evapotranspiration, agricultural ones to soil water shortage for plant growth, and hydrological ones to surface and/or underground flow decrease. Another difficulty is that droughts affect not only the atmosphere but also the land biosphere, and to a larger extent than heatwaves.

The variability of O_3 concentration is generally not well represented in chemistry-transport models (CTMs) compared to observations because of the lack of land-biosphere interactions in many models (Wang et al., 2017). Uncertainties related to BVOC emission modeling could lead to a BVOC-derived O_3 uncertainty estimated at about 50% in Europe (Curci et al., 2010). Another major cause of uncertainties concerns the simulation of meteorological conditions.

Meteorological conditions specific to droughts are low relative humidity, low or absence of precipitation and cloud cover. The latter leads to an increase of downward solar radiation. Temperature can also be higher during droughts than during normal conditions. Wang et al. (2017) found that drought periods in the United States over 1990-2014 were characterized by temperatures up to 2°C hotter and radiation $12.4 W/m^2$ higher. Their analysis of surface measurements shows a mean enhancement in O_3 concentration of 3.5 ppbv (8%), explained by enhanced precursor emissions and photochemistry.

Through soil moisture deficit, droughts can induce considerable adverse effects on the biosphere, leading to a decrease of the overall biomass but also of the BVOC emission activity. Guion et al. (2021) report a decrease of $\sim 10\%$ of Leaf Area Index (LAI) observed by satellite during droughts in Southern Europe. Areas of low altitude and vegetation with short root systems are more sensitive. Based on simulations from the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006, 2012) and using MODIS LAI between 2003 and 2018, Cao et al. (2021) found that vegetation biomass variability in China is a major controlling factor of the interannual variation of BVOC emissions.

Biogenic emission models such as the MEGAN model, consider an emission reduction when drought episodes occur only for isoprene (C_5H_8). Emmerson et al. (2019) simulated a reduction in C_5H_8 emissions of 24-52% over Southeastern Australia due to soil dryness in summer. However, there is a lot of uncertainty on how plant activity reacts to water stress. In the case of C_5H_8, the response to water stress could occur in two phases: a state of increasing emissions due to leaf temperature stimulation during the drought onset, followed by a state of isoprene synthase limitation (Potosnak et al., 2014). For monoterpenes, the response to water stress may be different depending on the species (Bonn et al., 2019). For instance, sabinene emissions (experimentally measured) from the European beech (*Fagus sylvatica*) decrease strongly with decreasing soil water availability

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while trans-β-ocimene emissions (from the same plant species) remain constant.

Dry deposition velocity directly depends on the stomatal conductance, which can be modified under hot and dry conditions (Lin et al., 2020). Stomatal conductance depends on several parameters. As identified by Kavassalis and Murphy (2017), the leaf-to-air vapor pressure deficit that depends on both temperature and relative humidity, is a key variable regulating the stomatal conductance. Extreme hot and dry conditions can cause stomatal closure to reduce transpiration. Moreover, lack of rainfall prevents wet deposition and plant water stress slows dry deposition. However, there are still large uncertainties about the relationship between the soil moisture and the dry deposition (Clifton et al., 2020b).

Considering the difficulty for models to simulate accurately soil moisture (Cheng et al., 2017) and the uncertainties about plant and BVOC species response to water stress (Bonn et al., 2019), the impact of soil moisture is often dismissed in biogenic emissions and dry deposition schemes used in CTMs. Therefore, knowledge about the respective effect on O_3 remains limited. To evaluate the effects of droughts and heatwaves on surface O_3 in Southwestern Europe, the variation of both BVOC emissions and dry deposition as a function of meteorological and hydrological conditions are analyzed in this paper including the soil dryness effect.

Agricultural droughts and heatwaves are identified based on the coupled meteorological and land surface vegetation regional model RegIPSL for the 1979-2016 period (Guion et al., 2021). Summers are less affected by drought events (33% of them) than by heatwaves (97%). Nevertheless, drought periods are generally much longer than heatwaves (22.3 days against 5.8). Those two types of extreme weather events can be highly correlated over the Western Mediterranean (temporal correlation R between 0.4 and 0.5).

Atmospheric chemistry is simulated using the regional CHIMERE CTM (Menut et al., 2021) over three selected summers (2012, 2013 and 2014), including the online calculation of biogenic emissions using the MEGANv2.1 model. Simulations are analyzed in conjunction with surface O_3 observations from the European surface network Air Quality e-Reporting (European Environment Agency, https://www.eea.europa.eu/data-and-maps/data/aqereporting-9, last access: 30 July 2021). Concentrations of formaldehyde ($HCHO$) are used as a proxy for BVOC emissions. $HCHO$ is produced in high yield during the oxidation of hydrocarbons. As an intermediate product of the oxidation of most VOC and being characterized by a short lifetime (about a few hours), $HCHO$ has been used in many studies to infer VOC emissions (e.g. Millet et al., 2006, 2008; Curci et al., 2010). Observations of $HCHO$ total column by the Ozone Monitoring Instrument (OMI) onboard the Aura satellite (Levelt et al., 2006) are used.

The observations and models used for this research are presented in Sect. 2 and 3, respectively. The RegIPSL, CHIMERE and MEGAN models, as well as the different modeling experiments undertaken are detailed. Results (see Sect. 4) are divided into three parts. Firstly, the sensitivity of C_5H_8 emissions, O_3 dry deposition and surface O_3 concentration to drought effects (biomass decrease and soil dryness) is assessed (see Sect. 4.1). Secondly, the variation of observed and simulated O_3, integrating drought and heatwave effects, is analyzed with respect to the variation of troposphere-atmosphere interactions during extreme weather events (see Sect. 4.2). Finally, the number and distribution of days exceeding the EU air quality standard are presented (see Sect. 4.3).

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2 Observations

2.1 European surface network of O_3 concentration

The in-situ measurements of surface O_3 provided by the EEA are used. The AQ e-Reporting project gathers air quality data provided by EEA member countries as well as european collaborating countries. Merging with the statistics of AirBase v8 project (2000-2012), AQ e-Reporting offers a multi-annual time series of air quality measurement data until 2021, categorized by station types and zones across Europe.

Thunis et al. (2013) quantified the various sources of uncertainty for O_3 measurements (e.g. linear calibration and ultraviolet photometry) and estimated a total uncertainty of 15%, regardless of concentration level.

Only measurements from stations classified as background and rural are considered for this study. The number of stations taken into account is 167, 188 and 207 respectively for summer 2012, 2013 and 2014.

2.2 Satellite observations of formaldehyde total column

The $HCHO$ total column observations from the OMI/Aura instrument are used as a quantitative proxy of BVOC emissions. OMI has an equator crossing time at 01:45 pm on the ascending node, about 14.5 revolutions per day and a swath width of 2600km, allowing daily global coverage (Levelt et al., 2006, 2018). The OMHCHOd level 3 product (Chance, 2019) is used for this research. It provides $HCHO$ total column with a spatial resolution of $0.1° \times 0.1°$, based on the backscatter solar radiation in the ultraviolet (306 and 360 nm). Uncertainties on the $HCHO$ retrieval is significant, varying between 30% (pixels with high concentrations) and 100% (with low concentrations), and mainly due to cloud and aerosol scattering along the field of view (González Abad et al., 2015). All satellite retrievals (e.g. OMI, GOME2A) of $HCHO$ products present a systematic low mean bias (20-51%) compared to aircraft observations (Zhu et al., 2016). However, the analysis using $HCHO$ total column from OMI presented in this study is mainly focused on the relative difference during droughts and heatwaves so that no correction factor has been applied.

Finally, only cloud free observations and pixels with a vertical column density within the range of -0.8e16 and 7.6e16 $molecules/cm^2$ are selected in order to minimize OMI row anomalies, following Zhu et al. (2017). For a suitable comparison with model simulations, $HCHO$ data were regridded on the CTM grid.

2.3 In-situ observations of 2m temperature and C_5H_8 surface concentration

To validate our simulations in the most complete way, two data sets are added to our study but do no represent the main focus of the paper. Firstly, in-situ observations of temperature at 2 meters above the surface (T_{2m}) from the E-OBS dataset (Cornes et al., 2018) are used to validate the simulated temperature by the Weather Research and Forecasting (WRF) model (Skamarock et al., 2008).

Secondly, simulated C_5H_8 concentrations from different experiments undertaken in this study are compared to in-situ observational data gathered by the EBAS infrastructure (https://ebas.nilu.no/).

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3 Models

3.1 WRF-ORCHIDEE

3.1.1 Model description

The regional WRF-ORCHIDEE model (RegIPSL) couples the land surface model ORCHIDEE (ORganising Carbon and Hydrology In Dynamic EcosystEms) (Maignan et al., 2011) and the meteorological model WRF. ORCHIDEE is composed of three modules; SECHIBA that simulates the water and energy cycle, STOMATE that resolves the processes of the carbon cycle allowing therefore an interactive phenology and finally, LPJ that computes the competition between the Plant Functional Types (PFTs). However, LPJ module is not used for this research.

RegIPSL model has been used following the recommendation of the Med-CORDEX (Coordinated Regional Climate Downscaling Experiment over the Mediterranean domain) initiative (Ruti et al., 2016). On a Lambert-conformal projection, the domain covers the Euro-Mediterranean with a spatial resolution of 20km. There are 46 vertical levels, with over half below 500hPa. The lateral boundaries of the atmosphere were nudged towards the ERA-Interim reanalysis.

The performed Med-CORDEX simulations over the 1979-2016 period have been evaluated against surface observations of temperature and precipitation, demonstrating that it is suitable for research on droughts and heatwaves (Guion et al., 2021).

3.1.2 Indicators of droughts and heatwaves

Based on simulations performed by the coupled WRF-ORCHIDEE regional model, we identified heatwaves and agricultural droughts over the Euro-Mediterranean using indicators of extreme weather events, as described in Guion et al. (2021) (data access: https://thredds-x.ipsl.fr/thredds/catalog/HyMeX/medcordex/data/Droughts_Heatwaves_1979_2016/catalog.html).

Following the approach of Lhotka and Kyselý (2015), we computed the Percentile Limit Anomalies of 2m above surface temperature (PLA_{T2m}) for heatwave detection and of soil dryness (PLA_{SD}) for agricultural drought detection. The soil dryness is computed as the complement of the soil wetness index, which is described in ORCHIDEE as the ratio between the soil moisture and the accesible water content.

For each cell of longitude (i) and latitude (j) of the domain, the monthly distribution of the 2m temperature/soil dryness is normalized and the 75^{th} percentile is computed ($p = 0.75$). The daily $PLA_{T2m/SD}$ indicator is equal to the daily deviation ($dX_{i,j,d}$) between the reference variable ($X_{i,j,d}$) and the corresponding monthly percentile ($X^{p}_{i,j,m}$) (Equation 1). A heatwave/drought is identified when the daily $PLA_{T2m/SD}$ is positive for three consecutive days.

$$dX_{i,j,d} = X_{i,j,d} - X^{p}_{i,j,m} \quad (1)$$

The chosen percentile for the detection of extreme weather events is generally higher, between 80 and 95 (e.g. Stéfanon et al., 2012). We chose the percentile 75 in this study for a larger population. As a result, events detected here cover not only extreme events, but also periods that are significantly drier and warmer than normal conditions.

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3.1.3 Soil water stress for plants

The Soil Water Stress (SWS) calculated by RegIPSL based on liquid water content in the soil is used to describe the water stress for plant transpiration and photosynthesis (De Rosnay et al., 2002). It is an index varying between 0 (plants fully stressed) and 1 (not stressed) that takes into account the water transfer between the soil layers along root profiles (11 layers in the version of ORCHIDEE used here). For each PFT (p), soil class (s) and soil layer (v), it is computed as follows:

$$SWS_{p,s} = \frac{SM_{p,s,v} - SMw_{p,s,v}}{SMnostress_{p,s,v} - SMw_{p,s,v}} \times nroot_{p,s,v} \quad (2)$$

where SM is the soil moisture (liquid phase), SMw the level at wilting point, $SMnostress$ the level at which SWS reaches 1 and $nroot$ the normalized root length fraction in each soil layer (between 0 and 1). There are three soil classes to avoid competition for water uptake: bare soil, soil with short root systems and soil with long ones.

3.2 MEGAN and the soil moisture factor

Biogenic emissions of VOCs and NO were computed using the MEGAN model v2.04 (Guenther et al., 2006), including several updates from the version 2.1 (Guenther et al., 2012). It is used on-line in the CHIMERE model at hourly time step. Emission fluxes are calculated based on emission factors at canopy standard conditions that are defined as $303K$ for the air temperature (T), 1500 $\mu mol/m^2.s$ for the photosynthetic photon flux density ($PPFD$) at the top of the canopy, 5 m^2/m^2 for the LAI and a canopy composed of 80% mature, 10% growing and 10% old foliage. Environmental conditions are taken into account using activity factors (γ) that represent deviations from the canopy standard conditions, so that the emission rate (ER [$\mu g.m^{-2}.h^{-1}$]) for a given species i is calculated in each model grid cell as:

$$ER_i = EF_i \cdot \gamma_{LAI} \cdot \gamma_{PPFD,i} \cdot \gamma_{T,i} \cdot \gamma_{Age,i} \cdot \gamma_{SM,i} \cdot \gamma_{CO2,i} \cdot \rho_i \quad (3)$$

where EF is the emission factor [$\mu g.m^{-2}.h^{-1}$] provided by the MEGAN model and ρ the production/loss term within canopy that is assumed to be equal to one. γ_{LAI} is the activity factor based on LAI observations from the MODIS MOD15A2H product (Myneni et al., 2015) improved by Yuan et al. (2011) (http://globalchange.bnu.edu.cn/research/lai). This improvement is undertaken with a two-step integrated method: (1) the Modified Temporal Spatial Filter is used to fill the gaps and replace low quality data by consistent data; (2) the TIMESAT Salvitzky and Golay filter is applied to smooth the final product. The temporal resolution is 8 days.

$\gamma_{PPFD,i}$, $\gamma_{T,i}$ and $\gamma_{Age,i}$ are the activity factors accounting for light, temperature and leaf age respectively. They are calculated in MEGAN using meteorological variables from the WRF model. The activity factors accounting for the soil moisture (γ_{SM})

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and CO_2 concentration (γ_{CO2}) are applied on C_5H_8 species only. γ_{CO2} is fixed (CO_2 concentration at 395 ppm) and γ_{SM} depends on the deviation between the soil wetness and a fixed wilting point, computed as follows:

$$\begin{cases} \gamma_{SM} = 1 & \text{if } \theta > \theta_1 \\ \gamma_{SM} = \frac{(\theta - \theta_w)}{\Delta\theta_1} & \text{if } \theta_w < \theta < \theta_1 \\ \gamma_{SM} = 0 & \text{if } \theta < \theta_w \end{cases} \tag{4}$$

where θ is the soil wetness (volumetric water content in m^3/m^3), θ_w the wilting point (level from which plants can not extract water from the soil, in m^3/m^3), $\Delta\theta_1$ equal to 0.04 (empirical parameter) and θ_1 the sum of θ_w and $\Delta\theta_1$. θ values (third layer, 60cm depth) are provided by the WRF-Noah land surface model (Ek et al., 2003; Greve et al., 2013). θ_w values are computed over the domain (Supplementary Information: Fig. S1) using tabulated values from Chen and Dudhia (2001) that are soil type specific and parametrized on Noah soil wetness values. However, emission response to drought is very sensitive to the wilting point (Müller et al., 2008). Wang et al. (2021b) computed a relative difference of biogenic emissions varying between 50 and 90% depending on the wilting point values.

Based on a comprehensive data set of in-situ measurements of C_5H_8 emissions, Bonn et al. (2019) computed an activity factor of soil moisture in function of the soil water availability. The fitted function is relatively in good agreement with the γ_{SM} algorithm in MEGAN.

As an alternative, Jiang et al. (2018) propose to use an activity factor of soil moisture that integrates the soil water stress for plant photosynthesis. Below a critical value (fixed here at 0.5), the activity factor using SWS from RegIPSL decreases lineary as follows:

$$\begin{cases} \gamma_{SWS} = 2\,SWS & \text{if } SWS \leq 0.5 \\ \gamma_{SWS} = 1 & \text{if } SWS > 0.5 \end{cases} \tag{5}$$

The different soil wetness functions presented here will be subject to dedicated simulations for comparison.

3.3 CHIMERE

3.3.1 General description and configuration

The CHIMERE v2020r1 model (Menut et al., 2021) Eulerian 3-dimensional regional CTM computes gas-phase chemistry, aerosols formation, transport and deposition. It can be guided by pre-calculated meteorology (offline simulations) or used in an online simulation with the WRF regional meteorological model (Skamarock et al., 2008), including the direct and indirect effects of aerosols in option. In this study, we chose to use the online version of CHIMERE but without any aerosol effects in order to reduce the calculation time and to isolate possible feedbacks.

Here, the reduced scheme MELCHIOR2 (Derognat, 2003) is used for gas phase chemistry (44 species, almost 120 reactions).

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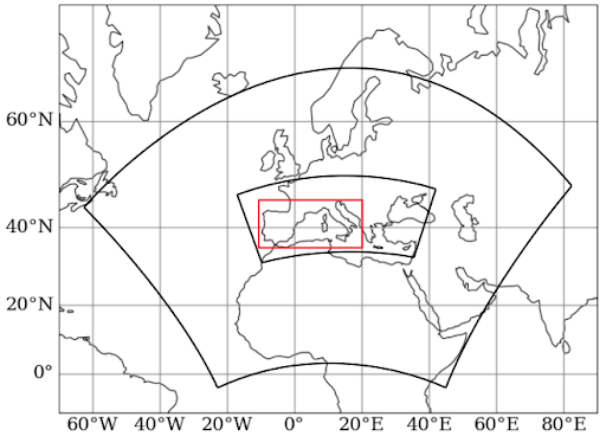

Figure 1. Domains used for the CHIMERE simulations: EUMED60 at 60km resolution is the large domain, the smaller nested domain is Med-CORDEX at 20km resolution. The Southwestern Europe region studied here is shown in red.

The aerosol module includes aerosol microphysics, secondary aerosol formation mechanisms, aerosol thermodynamics and deposition, as detailed in Couvidat et al. (2018); Menut et al. (2021). The photolysis rates are calculated online using the Fast-JX module version 7.0b (Bian and Prather, 2002) which accounts for the radiative impact of aerosols. Horizontal and vertical advection are based on the scheme of Van Leer (1977). The physical and chemical time steps are respectively 30 and 10 minutes. The simulations have been performed for June-July-August (with a 5 days spin-up) of years 2012, 2013, 2014, over two nested simulation domains (Fig. 1), and on 15 vertical layers, from 998hPa up to 300hPa. The large domain covers Northern Africa and Europe at 60km horizontal resolution (EUMED60, 164x120 cells) and the small domain is close to that of Med-CORDEX although smaller; it covers the Western Mediterranean region at 20km horizontal resolution (Med-CORDEX, 222x93 cells). The area studied in more detail in the following is the Southwestern Europe (35-46°N, 10°W-20°E).

Boundary conditions for the larger domain are provided by a climatology from the global CTM LMDZ4-INCA3 (Hauglustaine et al., 2014) for trace gases and non-dust aerosols, and from the GOCART model (Chin et al., 2002) for dust.

The biogenic emissions are calculated online by the MEGAN model (see Sect. 3.2). Sea salts and dimethyl sulfide marine emissions are calculated online using the scheme of Monahan (1986) and Liss and Mervilat (1986), respectively. Mineral dust emissions are also calculated on-line (Marticorena and Bergametti, 1995; Alfaro and Gomes, 2001). Consistently with the WRF model, the land cover classification used for the calculation of biogenic emissions and dust emissions is from the United States Geological Survey (USGS) land cover database (http://www.usgs.gov). The biomass burning emissions from the APIFLAME v2.0 model (Turquety et al., 2020) are used. The anthropogenic surface emissions are derived from the EMEP database at $0.1° \times 0.1°$ resolution (https://www.ceip.at/webdab-emission-database) for the year 2014. The inventory was preprocessed by the emiSURF program in order to calculate hourly emissions fluxes on the CHIMERE grid and the period of simulation (https://www.lmd.polytechnique.fr/chimere/formation_2021/emisurf.pdf).

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3.3.2 Dry deposition scheme

Dry deposition in CHIMERE is resolved online using a canopy resistance approach, based on the scheme of the EMEP model (Wesely, 1989; Emberson et al., 2000; Simpson et al., 2003, 2012). Canopy resistance is calculated from stomatal conductance (g_{sto}) which increases proportionally with LAI, and the bulk non-stomatal conductance. g_{sto} varies between a maximum (g_{max}) and minimum (g_{min}) daytime stomatal conductance [mmol $O_3/m^2.s$] depending on a series of meteorological conditions represented by factors of relative conductance (f) (Eq. 6). The parameters used to calculate these factors vary with land cover (lc) and the seasonality (Emberson et al., 2000). The land cover classes used in the dry deposition scheme being different from the one used in CHIMERE (USGS), a matrix of conversion is applied.

$$g_{sto} = g_{max,lc} \cdot f_{phen,lc} \cdot f_{light,lc} \cdot max(g_{min,lc}, (f_{temp,lc} \cdot f_{VPD,lc} \cdot f_{SWS,lc})) \quad (6)$$

where f_X are factors of relative conductance determined by the leaf/needle age ($X = phen$), irradiance ($X = light$), temperature ($X = temp$), leaf-to-air vapour pressure deficit ($X = VPD$) and the soil moisture ($X = SWS$). Environmental variables required for f_X factors are from the WRF model except for f_{SWS} that is based on SWS from RegIPSL. f_{SWS} for which the calculation is the same as γ_{SWS}, follows the parametrization suggested by Simpson et al. (2012) for the stomatal conductance.

3.3.3 Experiments

The years 2012-2013-2014 were selected for the large diversity of hydro-climatic conditions and the availability of O_3 observations over the study area. During summer 2012, Southwestern Europe was affected by severe droughts (PLA_{SD} > 0.1 of soil dryness) and heatwaves (PLA_{T2m} > 1.5°C) (SI: Fig. S2). The summer 2013 was wet in the Iberian Peninsula, while the summer 2014 was wet in Italy and the Balkans. A few heatwaves occurred in summer 2013 over Southern France, Northern Italy and Northern Spain. Summer 2014 was colder (PLA_{T2m} of -4.0°C) for the entire study area. The analysis method we propose here is divided into two steps.

Firstly, a focus is made on drought effects (biomass decrease and soil dryness) that are generally poorly represented in modeling experiments, over summer 2012. Several experiments are conducted on the Med-CORDEX domain in order to analyze the sensitivity of simulated biogenic emissions, dry deposition and surface O_3 to drought effects. Table 1 summarizes the CHIMERE simulations conducted. The "Reference" simulations include all the emissions as defined in Sect. 3.3.1 but without accounting for the soil moisture factor in the emission scheme of MEGAN and deposition one of CHIMERE. A simulation with no biogenic emissions is also performed ("NoBio-emiss") to estimate their relative contribution to O_3 concentration.

The LAI used in the emission scheme of MEGAN is year dependant. To evaluate the effect of biomass decrease by droughts, a simulation with LAI corresponding to a wet summer was used ("HighLAI-emiss"). Summer 2012 which was affected by an important biomass decrease over most of the study area (SI: Fig. S3), has been simulated with the LAI of the wet summer of 2014 (higher than the 2012-2014 mean).

The LAI database used in the dry deposition scheme in CHIMERE does not vary with the year, so that the biomass decrease

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Table 1. Name, description and aim of the simulations launched with the CHIMERE model on the nested Med-CORDEX domain for summers 2012, 2013 and 2014.

Simulation name	Description	Aim
Reference (R.)	CHIMERE reference (v2020r1)	Default for dry and hot periods
	On biogenic emissions	
NoBio-emiss	R. without biogenic emissions	Contribution of biogenic emissions to O_3
HighLAI-emiss	R. with wet summer LAI	Effect of biomass decrease
γ_{SM}-emiss	R. with γ_{SM} factor from Noah	Effect of soil dryness
γ_{SWS}-emiss	R. with γ_{SWS} factor from ORCHIDEE	Effect of soil dryness
γ_{SWSfit}-emiss	R. with γ_{SWSfit} factor from ORCHIDEE	Effect of soil dryness
	On gas dry deposition	
LAIdecr-dep	R. with prescribed LAI reduction	Effect of biomass decrease
LAIdecr/f_{SWS}-dep	R. with prescribed LAI reduction and f_{SWS} factor from ORCHIDEE	Effect of biomass decrease and soil dryness

during the summer 2012 is not reflected. In order to evaluate the importance of this effect, a simulation ("LAIdecr-dep") with a LAI decrease of 5% for forests and 20% for grass and crops (close to the variations observed in 2012 (SI: Fig. S3)) has been conducted.

The effect of soil dryness for the BVOC emissions is evaluated using three different approaches:

- using γ_{SM} from WRF-Noah in MEGAN ("γ_{SM}-emiss" experiment),
- using γ_{SWS} from RegIPSL (WRF-ORCHIDEE) in MEGAN ("γ_{SWS}-emiss" experiment),
- using γ_{SWSfit} from RegIPSL (WRF-ORCHIDEE) in MEGAN ("γ_{SWSfit}-emiss" experiment). The fitted function of Bonn et al. (2019) is applied on the SWS.

The soil dryness effect on the dry deposition is evaluated using the f_{SWS} from ORCHIDEE in CHIMERE ("f_{SWS}-dep" experiment).

Secondly, based on a simulation configuration integrating all drought and heatwave effects ("γ_{SWSfit}-emiss & LAIdecr/f_{SWS}-dep" experiment, hereafter called "all-emiss-dep" experiment), the variation of C_5H_8 emissions, O_3 dry deposition velocity and surface O_3 concentration is analyzed over summers 2012, 2013 and 2014. In order to compare isolated and combined extreme events with normal conditions, we used a cluster approach based on the PLA_{T2m} and PLA_{SD} indicators.

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3.3.4 Validation of surface O_3, NO_2 and 2m temperature

Simulated surface O_3 and NO_2 concentrations by CHIMERE were compared to the EEA observations while simulated temperature at 2m above the surface by WRF was compared to the E-OBS observations (Cornes et al., 2018). Table 2 presents the mean validation scores of the "Reference" simulations over Southwestern Europe for summer 2012. Validation scores of O_3 and NO_2 are very similar for summers 2013 and 2014.

The simulated surface O_3 is slightly higher than the observations for hourly and daily mean values (bias "observation - model" of -1.93 and -0.13$\mu g/m^3$) and lower for the daily maximum values (+10.94$\mu g/m^3$). The simulated diurnal cycle is lower than the observed one: CHIMERE overestimates the daily minimum and underestimates the maximum. The temporal correlation (R pearson coefficient) varies between a 0.53 in daily mean and 0.57 in hourly mean. The spatial distribution of bias and correlation scores of daily maximum values for each summer is shown in SI (Fig. S4). The highest bias ($>$ +25$\mu g/m^3$) and lowest correlations (R of $\approx$ 0.2) are obtained near large urban areas (e.g. Madrid, Milano) which may be less well represented in the model even if the stations are classified as rural background. There are substantial uncertainties related to the NO_x/VOC emissions in (peri-)urban areas.

These O_3 scores show an overall agreement with those calculated by Gaubert et al. (2014) and Menut et al. (2021) over Europe. The averaged daily maximum bias and correlation coefficient computed are slightly higher and lower, respectively, in this study mainly because the area is limited to Southwestern Europe. Due to the multiple sources of O_3 precursors and favourable temperature and light conditions, the Mediterranean is identified as a region of important uncertainty for modeling O_3 concentration (Richards et al., 2013).

Validation scores are also computed for the surface NO_2 using the EEA observations, with a different distribution of stations than for ozone. In agreement with the results of Menut et al. (2021), CHIMERE underestimates NO_2 concentrations (+4.95$\mu g/m^3$ in daily mean and +8.57$\mu g/m^3$ in daily maximum) compared to EEA observations. Lamsal et al. (2008) emphasize that the chemiluminescence analyzer, the measurement technique primarily found in air quality stations, is subject to significant interference from other reactive species containing oxidized nitrogen (e.g. PAN, HNO_3). It may partly explain the constant underestimation compared to observations.

The mean temporal correlation coefficients do not exceed 0.4. These low validation scores may also be linked to the emission inventory used as well as meteorological conditions (boundary layer height representation in particular). As a major O_3 precursor, part of the uncertainty of this study is directly related to NO_2 emissions and concentrations.

Finally, as a critical variable for simulating diurnal and seasonal O_3 concentrations, temperatures at 2m above the surface (T_{2m}) have been compared to the E-OBS observations. Averaged over the Southwestern Europe, the bias is close to 0°C while the RMSE is significant (8.19°C in daily mean and 12.63°C in daily maximum). The spatial distribution of the bias presents large variations (SI: Fig. S5). The daily maximum T_{2m} is overestimated in the Southern Mediterranean (up to 5°C) compared to observations while it is underestimated in the norhtern part (up to 5°C). Nevertheless, the averaged scores of temporal correlation are high (around 0.75).

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Table 2. Comparisons between observed and simulated surface O_3 concentration, NO_2 concentration and 2m temperature, averaged over Southwestern Europe during summer 2012, for the "Reference" CHIMERE simulation.

	Observations	Model	Bias (obs. - mod.)	RMSE	Pearson correl. (R)
O_3	$[\mu g/m^3]$	$[\mu g/m^3]$	$[\mu g/m^3]$	$[\mu g/m^3]$	
Hourly	83.21	85.14	-1.93	20.45	0.57
Daily mean	83.93	84.06	-0.13	14.31	0.53
Daily max	116.32	105.39	10.94	21.49	0.54
NO_2	$[\mu g/m^3]$	$[\mu g/m^3]$	$[\mu g/m^3]$	$[\mu g/m^3]$	
Hourly	7.65	3.70	3.95	5.57	0.25
Daily mean	7.55	2.60	4.95	5.24	0.40
Daily max	14.65	6.08	8.57	9.87	0.37
T_{2m}	$[°C]$	$[°C]$	$[°C]$	$[°C]$	
Daily mean	22.25	22.25	-0.01	8.19	0.76
Daily max	23.13	28.34	0.56	12.63	0.75

Even if such validation scores are close to those found in the scientific literature (e.g. Panthou et al., 2018), the temperature uncertainties significantly contribute to those of the O_3 simulated by CHIMERE.

4 Results

4.1 Sensitivity to soil dryness and biomass decrease effects

The sensitivity analysis to soil dryness and biomass decrease effects is focused on summer 2012 as droughts mainly occurred during this year (over the 2012-2014 period). Most of Southwestern Europe was affected by a biomass decrease in 2012 (compared to the wet summer 2014), except Eastern Spain. The LAI difference between summer 2012 and 2014 can reach -30% over central Italy (Appenines region) corresponding to a mean decrease of 0.5 m^2/m^2 (SI: Fig. S3).
C_5H_8 is the main contributor to the total mass of $BVOCs$ emitted (70%), followed by the model species APINEN (13%, including e.g. α-pinene, sabinene).

4.1.1 C_5H_8 emissions

Figure 2 (lower panel) shows the daily mean C_5H_8 emissions in Southwestern Europe for summer 2012 simulated by the MEGAN model ("Reference" simulation). The spatial distribution of C_5H_8 over the Mediterranean is heterogeneous, ranging from areas of zero or low emissions ($\sim 1 \times 10^{-3} g.m^{-2}.h^{-1}$) to high emitting areas ($\sim 1.5 \times 10^{-3} g.m^{-2}.h^{-1}$) such as in the Balkans, Apennins, Sierra Morena, Sardinia and Central Portugal. Similar spatial distributions were found for APINEN model

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species. Differences in spatial patterns among species depend on the variation of emission factors over the land cover classes. The effect of biomass decrease and soil dryness on C_5H_8 emissions during the dry summer 2012 is also shown in Figure 2. Biomass decrease averaged over June, July and August is characterized by negative differences of emissions over most of the southern part of the region, reaching -20% in Northern Spain and -25% in Northern Italy.

The impact of soil dryness is assessed with the "γ_{SM}-emiss", "γ_{SWS}-emiss" and "γ_{SWSfit}-emiss" MEGAN simulations. The quantified impact can vary considerably between them. In the "γ_{SM}-emiss" experience, C_5H_8 emissions stay constant as long as the soil moisture is above the wilting point (θ_w). Once this point is reached (water is missing), it is assumed that plant can not synthesize C_5H_8 anymore and the emissions decrease steeply. θ_w is constant and depends only on the soil type. For instance, silt soil is characterized by lower θ_w than clay soil, so that soil dryness is more quickly reached for the latter. Due to the high spatial variability of soil type, the difference induced by "γ_{SM}-emiss" is also characterized by large variability. Within a same region (e.g. Central Italy), the difference sharply varies between -50% and 0% while the region is all over affected by an agricultural drought according to the PLA_{SD} indicator (SI: Fig. S2).

For the alternative approach based on the dynamical SWS function, the relative difference of C_5H_8 emissions ("Reference" .vs. "γ_{SWS}-emiss" experiment) is more spatially homogeneous than "γ_{SM}-emiss", and the overall reduction is larger. The strongest stress values are located in plains and for plants with short-root systems, in agreement with the sensitivity analysis performed by Vicente-Serrano (2007) about drought effects on vegetation. Semi-arid regions (e.g. Andalusia) are strongly affected as the water budget is almost permanently in deficit (high solar radiation and low/no precipitation). Being adapted to recurrent droughts, some plant species (e.g. *Arundo donax* in a Moroccan ecotype) can reduce the isoprene synthesis as the result of pressure selection to preserve their viability (Haworth et al., 2017). However, the emission reduction could be overestimated as irrigation is largely used in many semi-arid areas (e.g. García-Vila et al., 2008). A specific analysis should be undertaken to cover the diversity of answers to drought stress in both natural and human influenced systems within such regions.

The third experiment "γ_{SWSfit}-emiss" presents the same areas of C_5H_8 decrease as "γ_{SWS}-emiss" but in a lower extent. C_5H_8 decreases when SWS values are the lowest. Semi-arid regions are also more affected (-50%) than others (e.g. -20% in Central Italy).

Three areas of high emissions are analyzed more specifically (boxes in Fig. 2): the Balkans, the Pô Valley and Central Spain. Their temporal evolution of C_5H_8 emissions during the summer 2012 for the different experiments are shown in Figure 3. Droughts and heatwaves occured over the three regions. Droughts do not negate the dependence on light and/or temperature, and emission peaks are driven by heatwaves with higher 2m temperature and solar radiation (SI: Fig. S6). However, the peak values are reduced if LAI decrease and soil dryness are accounted for. Averaged over the three regions, the biomass decrease induces a 3% decrease of the total summer amount of C_5H_8 emitted. The soil dryness parameter induces difference of -12% for "γ_{SM}-emiss", -39% for"γ_{SWS}-emiss" and -13% for "γ_{SWSfit}-emiss". Based on experimental measurements over three summers (2012, 2013 and 2014) in a Mediterranean environment (Observatoire de Haute Provence, *Quercus pubescens* plant species), Saunier et al. (2017) identified a 35% emission decrease due to severe droughts. Demetillo et al. (2019) measured in California a C_5H_8 concentration reduction of 50% during severe droughts (2014 and 2015). These values are close to and even

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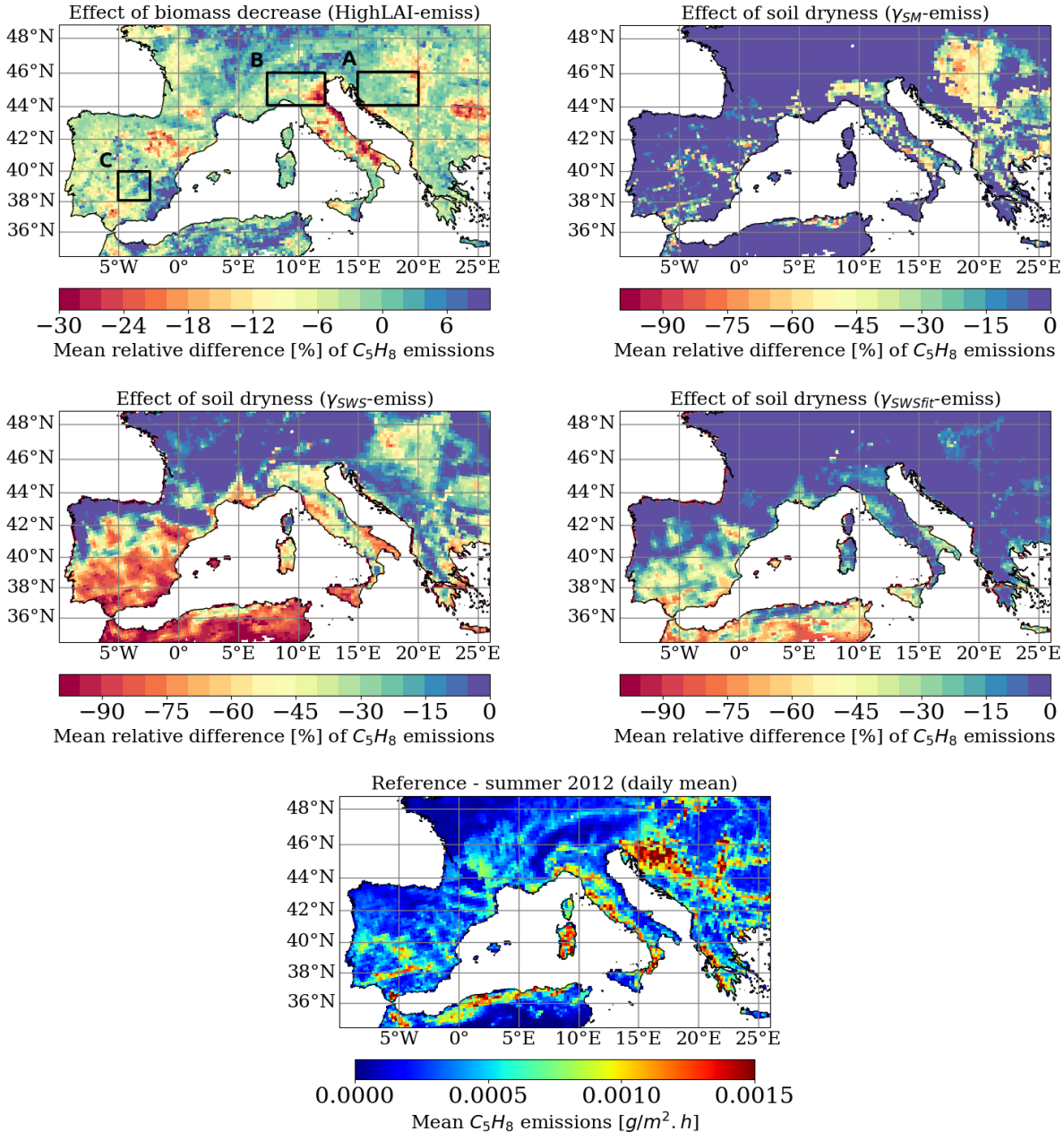

Figure 2. Summer mean (JJA) of daily mean isoprene emissions [$g.m^{-2}.h^{-1}$] for 2012 from the "Reference" simulation (lower panel). Mean relative difference [%] of C_5H_8 emissions due to biomass decrease and soil dryness. The relative difference is computed between the "Reference" and "HighLAI-emiss" simulation to quantify the biomass decrease effect (upper left panel), and "γ_{SM}-emiss"/"γ_{SWS}-emiss"/"γ_{SWSfit}-emiss" to quantify the soil dryness effect. For diagnostic purposes, areas of interest are designated within the dashed rectangles: here named as the "Balkans" (A), "Pô Valley" (B) and "Central Spain" (C).

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in the range of the simulated experiments. "γ_{SWS}-emiss" experiment is considered here as the lower limit of the reduction range of C_5H_8 emissions due to dry conditions, SWS being included in the calculation of the emissions rate as soon as soil water stress for plant stress is below 0.5.

Finally, the different simulated experiments from the MEGAN-CHIMERE model have been compared to observations of surface C_5H_8 concentrations from EBAS data set (Fig. 4). The Ersa station has an almost complete time series for summer 2012. Based on the PLA indicator, Northern Corsica was affected by two heatwaves but no drought. However, we detected soil dryness conditions close to the drought limit (mean PLA_{SD} of -0.09). The temporal correlation between the simulated and observed C_5H_8 concentration is similar for each experiment (R coefficient around 0.68). Averaged over the summer, the lowest mean bias with the observations is obtained with "γ_{SWSfit}-emiss" (-28 pmol/mol) and the largest one with "γ_{SM}-emiss" (-151 pmol/mol). Nevertheless, over July and August, "γ_{SWS}-emiss" experiment presents the lowest mean bias (+56 pmol/mol). The "Reference" and "γ_{SM}-emiss" experiments have equal values as the soil wetness from WRF-Noah is above the local wilting point. However, this comparison between observations and simulations is made at a single station. It would be valuable to carry out the same exercise at several locations (including areas representative of semi-arid environment) and over several summers.

4.1.2 O_3 dry depositon velocity

Figure 5 (upper left panel) presents daily mean dry deposition velocity values that range between 1.0 and 1.8cm/s within our areas of interest. The mean effect of biomass decrease (relative difference between the "Reference" and the "LAIdecr-dep" experiment, upper right panel) over summer 2012 does not exceed -8%, and is lower over the forested areas (as it was prescribed in our experiment).

The cumulative effects of biomass decrease and soil dryness is represented with the "LAIdecr-dep/f_{SWS}-dep" experiment (Fig. 5, lower pannel). The soil dryness effect is much larger than that of biomass decrease. Values range from 0 to -35%. Central Italy and the Iberian Peninsula are the most affected regions.

The temporal evolution of O_3 dry deposition velocity for the three areas of interest is shown in Figure 6. The biomass decrease effect is larger (e.g. -9% over the Pô Valley) during the first half of summer. It is close to -1% at the end of summer. This is explained by the LAI decrease prescribed from mid-July in the dry deposition scheme. In addition to that, the dry deposition velocity is characterized by a decreasing trend over summer imposed by the fixed phenology factor (f_{phen}).

Regarding the soil dryness, its effect (larger than the biomass decrease) is almost constant during summer (e.g. -12% over the Pô Valley). Based on measurements over Central Italy, Lin et al. (2020) computed a relative difference of about -50% between August 2004 (wet summer, ∼0.8cm/s) and August 2003 (dry summer, ∼0.4cm/s). However, summer 2003 was characterized by considerable heatwaves in Italy (PLA_{T2m} positive) which might intensify the decrease. We emphasize that dry deposition velocity generally decreases during heatwaves (Fig. 6) as the near surface temperature is above the optimal temperature of stomatal conductance. In conclusion, both effects related to droughts and heatwaves (the most intense) lead to a reduction in O_3 deposition.

Identifying soil dryness as a major driver of inter-annual variability of O_3 deposition velocity, Lin et al. (2019) emphasize that error in modeling the deposition may considerably rely on the ability of models to simulate accurately the precipita-

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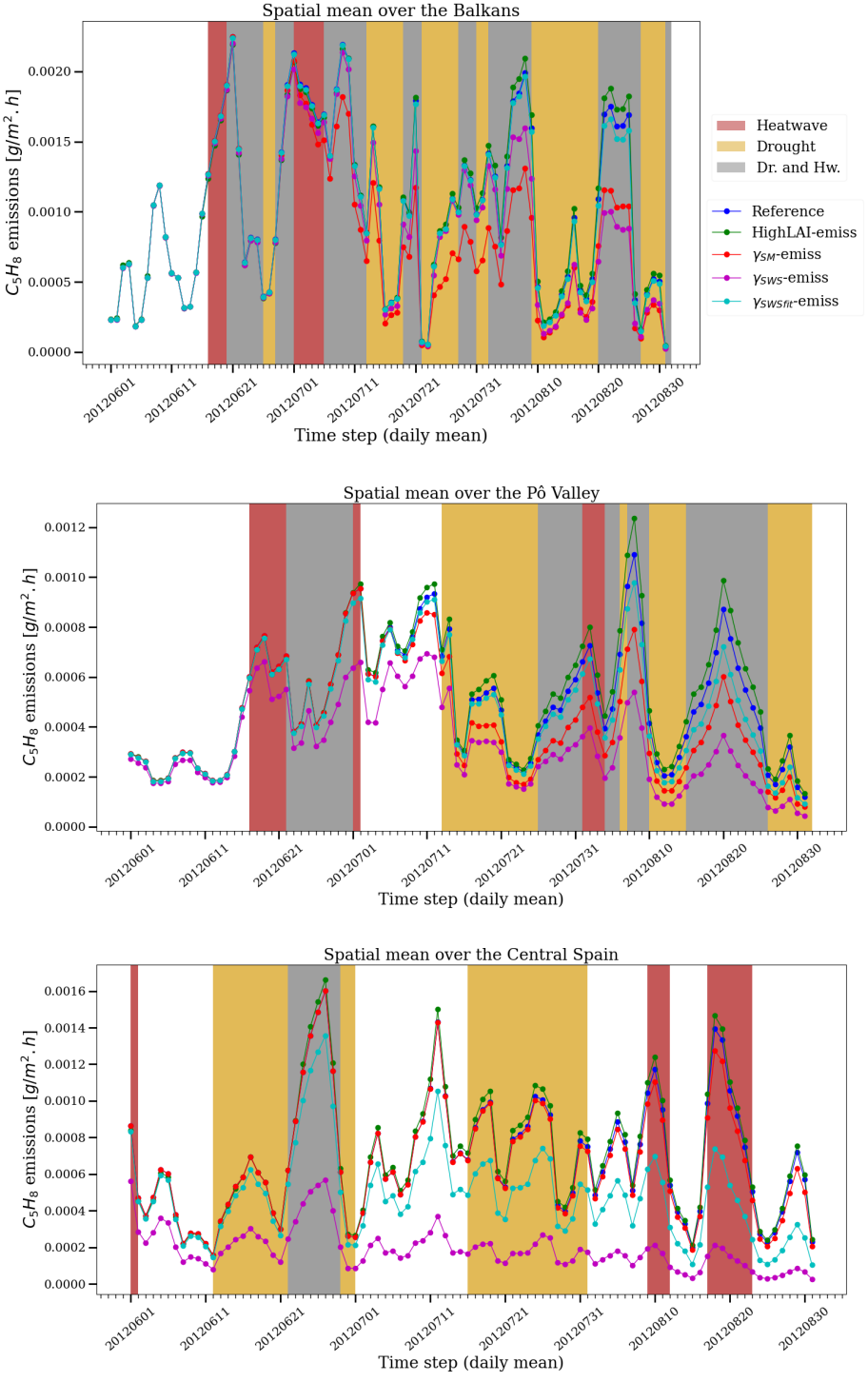

Figure 3. Daily mean C_5H_8 emissions [$g.m^{-2}.h^{-1}$] during the summer 2012, spatially averaged over the Balkans (upper panel), Pô Valley (middle panel) and Central Spain (lower panel) for the different MEGAN experiments. The colored bands highlight periods of droughts and heatwaves.

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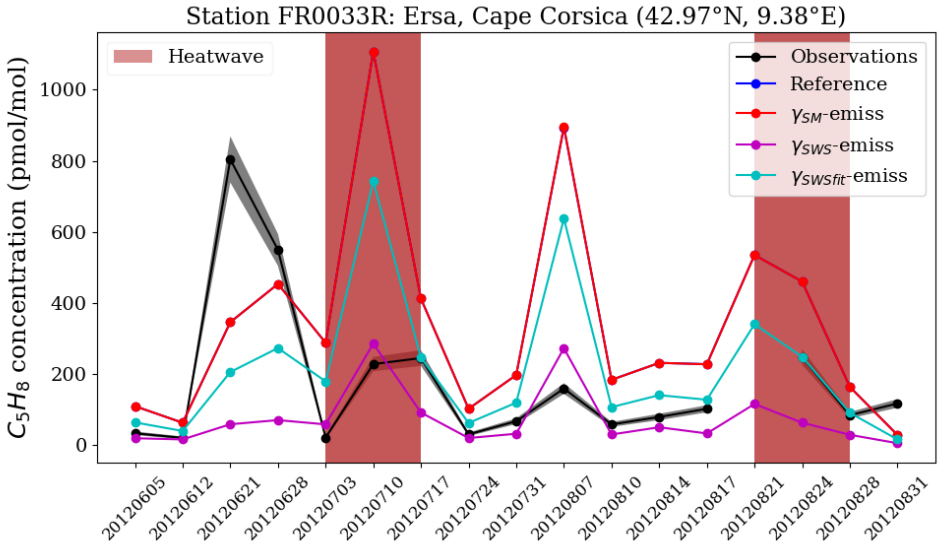

Figure 4. Observed surface C_5H_8 concentration $[pmol/mol]$ during the summer 2012 at the Ersa station (FR0033R, Cape Corsica) from the EBAS data set, compared to the different simulated experiments undertaken by the MEGAN-CHIMERE model. The shaded curve in black represents the precision range of the measurements. The "Reference" and "γ_{SM}-emiss" experiments here have equal values.

tion distribution. They assessed the sensitivity of their deposition velocity scheme (Geophysical Fluid Dynamics Laboratory LM3.0/LM4.0) to two different meteorological forcings and found a factor two over the Northern Europe. We need to emphasize that to the best of our knowledge, there is no study yet in the scientific literature that fully assess the O_3 dry deposition of CHIMERE against observations or its sensitivity to different meteorological forcings.

4.1.3 O_3 surface concentration

Figure 7 (upper left panel) maps the mean surface O_3 concentration simulated by CHIMERE ("Reference" experiment) over the summer 2012. High concentrations (between 100 and 110$\mu g/m^3$) are located in the eastern part of the study area (e.g. the Balkans and Italy). The spatial distribution of mean surface O_3 shows similar patterns as the distribution of PLA_{T2m} (SI: Fig. S2), highlighting the critical role of the temperature. Surface O_3 remains high above the sea due to transport and the absence of dry deposition (e.g. 120$\mu g/m^3$ over the Adriatic sea). The contribution of BVOC emissions on surface O_3 is also presented (upper right panel). BVOC emissions are known to be significant precursors of O_3 production in the Mediterranean region. It varies between 4% (e.g. Iberian Peninsula) and 22% (e.g. Northern Italy) in our study, that is included in the range of values reported in the scientific literature (e.g. Mertens et al., 2020). Finally, our areas of interest are located in rural regions, mostly characterized by a NO_x-limited regime (SI: Fig. S7).

A simulation including drought effects on both C_5H_8 emissions and O_3 dry deposition has been conducted with MEGAN-CHIMERE ("γ_{SWS}-emiss & LAIdecr/f_{SWS}-dep" experiment). The resulting effect on surface O_3 is shown in Figure 7 (lower

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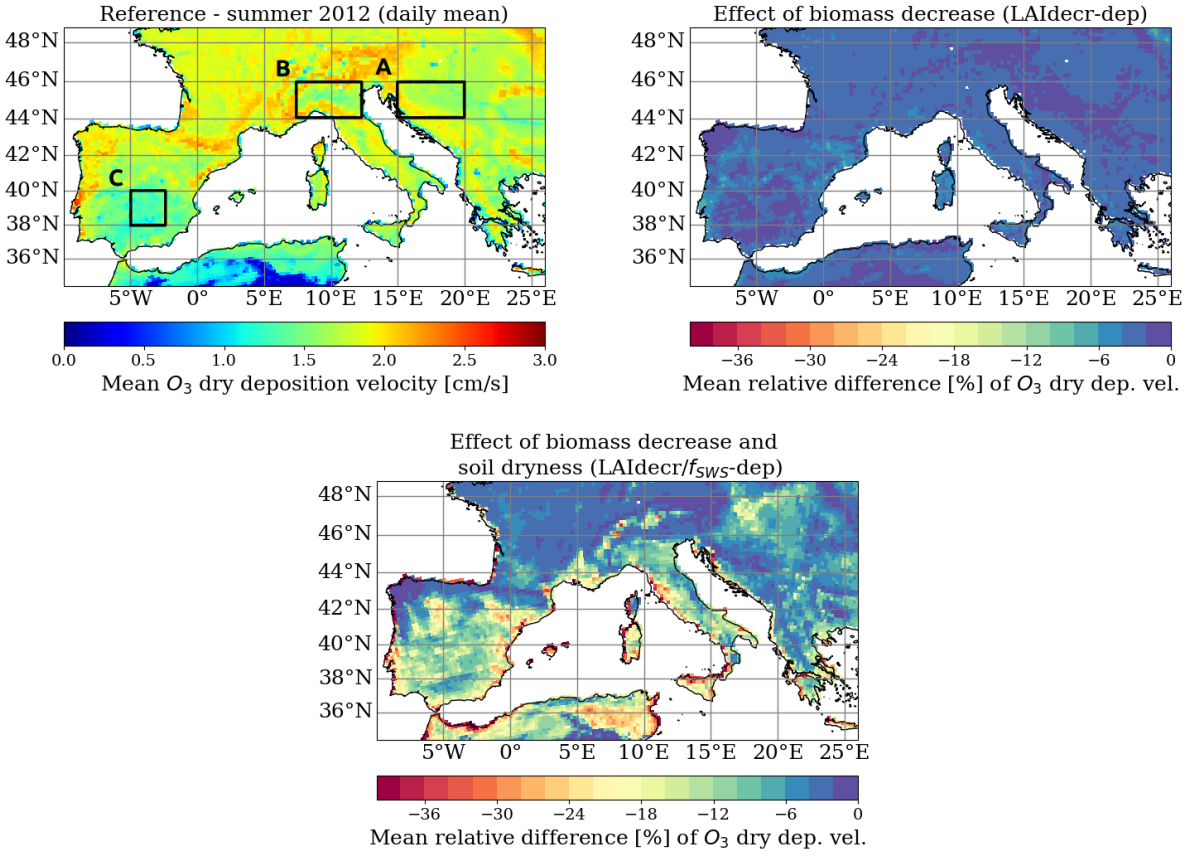

Figure 5. Summer mean (JJA) of daily mean O_3 dry deposition velocity $[cm/s]$ for 2012 from the "Reference" simulation (upper left panel). Mean relative difference [%] of O_3 dry deposition velocity due to biomass decrease and soil dryness. The relative difference is computed between the "Reference" and "LAIdecr-dep" simulation to quantify the biomass decrease effect (upper right panel) and "LAIdecr-dep/f_{SWS}-dep" to quantify combined biomass decrease and soil dryness effect (lower panel).

panels). On average during the summer (lower left), O_3 concentration is slightly higher over the continent (between +0.5% and
435 +3.0%) due to the decrease of O_3 deposition as dominant effect, while it is slightly lower (between -1% and -1.5%) over the sea and ocean due to the lower transport of O_3 precursors, compared to the "Reference" experiment. However, the O_3 increase over the Iberian Peninsula may be overestimated as the LAI reduction we applied to the whole domain (e.g. -20% for grass PFTs) is larger compared to the variation of MODIS LAI in this specific region (SI: Fig. S3).

Drought effects on surface O_3 induced by C_5H_8 emission reduction is not constant. It is largest during combined heatwaves
440 (i.e. when biogenic contribution is high). As a result, drought effects on C_5H_8 emissions can be dominant (compared to the effects on O_3 deposition) during simultaneous droughts and heatwaves, inducing a decrease of O_3 peaks by a few $\mu g/m^3$ both over continent and sea/ocean. The maximum absolute relative difference (Fig. 7, lower right panel) reaches -5% over the Pô

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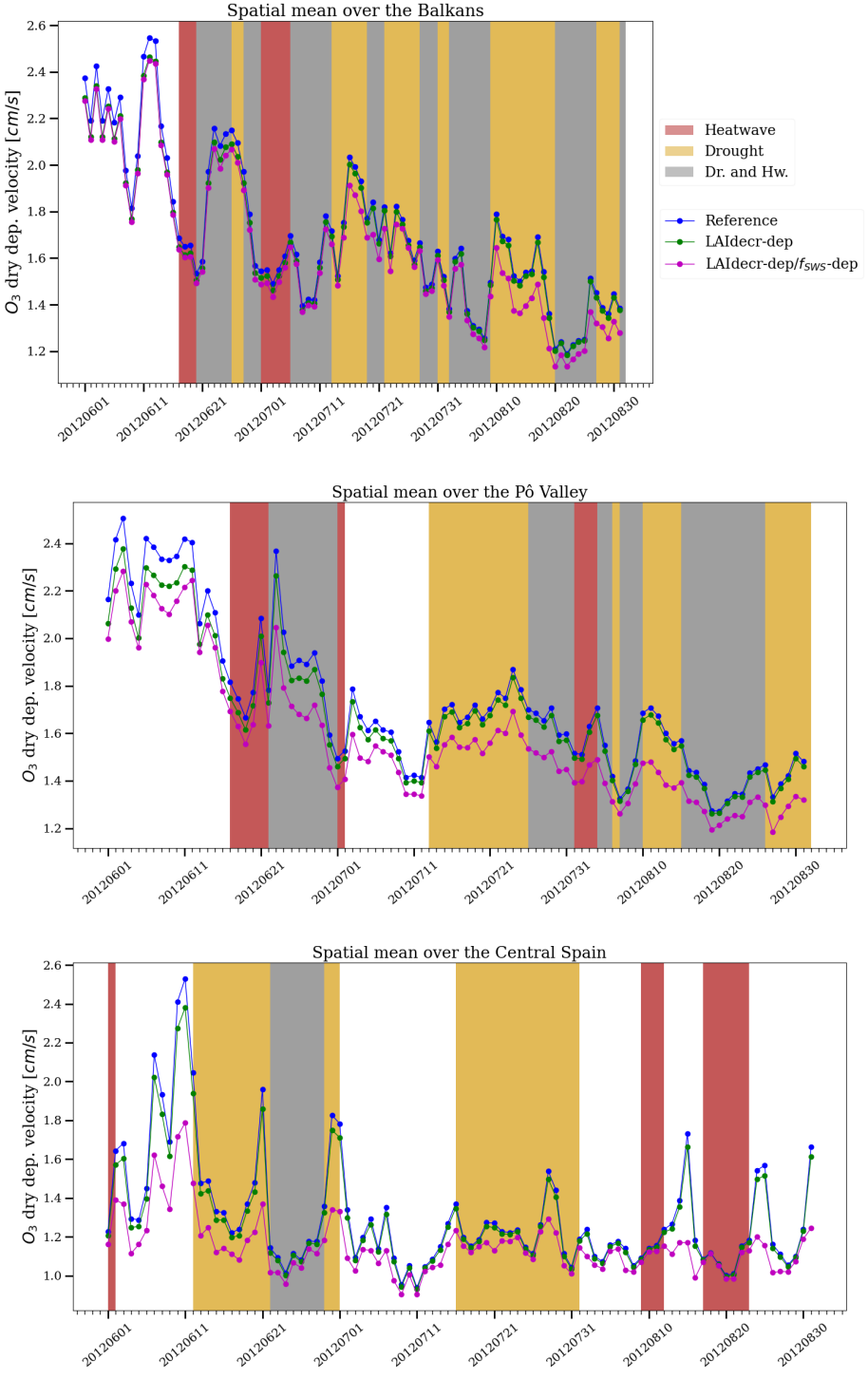

Figure 6. Daily mean O_3 dry deposition velocity [cm/s] during the summer 2012, spatially averaged over the Balkans (upper panel), Pô Valley (middle panel) and Central Spain (lower panel) for the different CHIMERE experiments.

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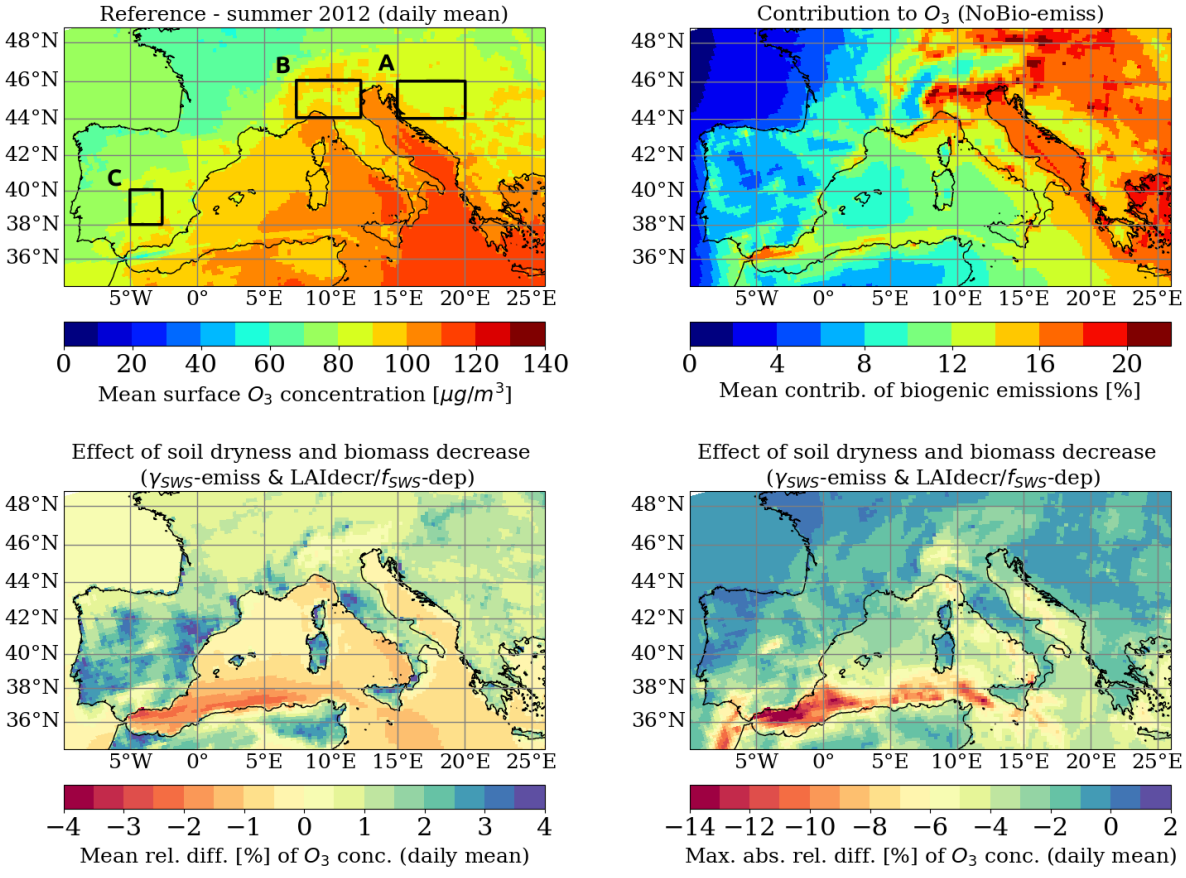

Figure 7. Summer mean (JJA) of daily mean O_3 surface concentration [$\mu g/m^3$] for 2012 from the "Reference" simulation (upper left panel). Mean contribution of biogenic emissions to O_3 surface concentration based on the "NoBio-emiss" experiment (upper right panel). Mean (lower left panel) and maximum absolute (lower right) relative difference [%] of O_3 surface concentration due to biomass decrease and soil dryness. The relative difference is computed between the "Reference" and "γ_{SWS}-emiss & LAIdecr-dep/f_{SWS}-dep" simulation from CHIMERE.

Valley and -14% along the Strait of Gibraltar. O_3 formation over the latter is favoured by large NO_x shipping emissions. Conducting a similar modeling experiment based on the γ_{SM} from MEGANv3, Jiang et al. (2018) simulated for the dry summer 2010 a maximum absolute relative difference of surface O_3 in August by -4%. O_3 reduction (-10% on average) due to severe droughts was also measured in California over the period 2002-2015 (Demetillo et al., 2019). This was identified as being related to a steep decrease of C_5H_8 concentrations.

For each area of interest, the temporal evolution of surface O_3 based on the different CHIMERE experiments and the EEA observations (AQ e-Reporting) is presented (Fig. 8 for the Pô Valley and Fig. S8 for the Balkans and Central Spain in SI). Since the resulting effects of biomass decrease and soil dryness on surface O_3 are less than the bias between model and observations

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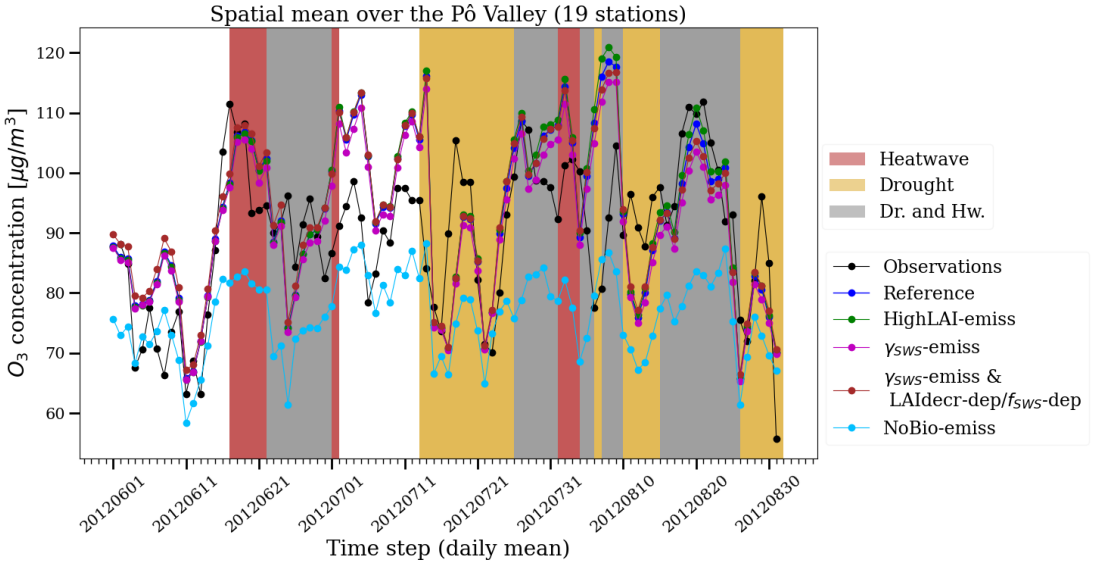

Figure 8. Daily mean O_3 surface concentration [$\mu g/m^3$] during summer 2012, spatially averaged over the Pô Valley from the EEA observations and the different CHIMERE experiments.

in our study (see Sect. 3.3.4), no simulation can be designated with certainty as the best fit.

4.2 Statistical variation during droughts and heatwaves

In this result section, a statistical analysis is performed using observations and simulations of the summers 2012, 2013 and 2014. The "all-emiss-dep" experiment has been chosen because it includes drought and heatwave effects in the most comprehensive way. Moreover, the C_5H_8 emission approach ("γ_{SWSfit}-emiss") has shown good performance compared to observations (Fig. 4), remaining rather conservative, not in the lower limit of the C_5H_8 reduction range.
Clusters of droughts and heatwaves (isolated or combined) are constructed based on the PLA_{T2m} and PLA_{SD} indicators and are used to represent the spatial distribution of C_5H_8 emissions, O_3 stomatal conductance and O_3 surface concentration during such events.

4.2.1 C_5H_8 emissions

Figure 9 shows the distribution of C_5H_8 emission rates over the Southwestern Mediterranean for clusters of extreme weather events. On average, the daily maximum C_5H_8 emission is significantly higher (t-test, $p < 0.01$) during isolated and combined heatwaves than normal conditions (no heatwave nor drought) with a mean value of 0.16×10^{-2} against $0.12 \times 10^{-2} g.m^{-2}.h^{-1}$. However, emissions during droughts have the same mean value as in normal conditions. During isolated droughts (clus-

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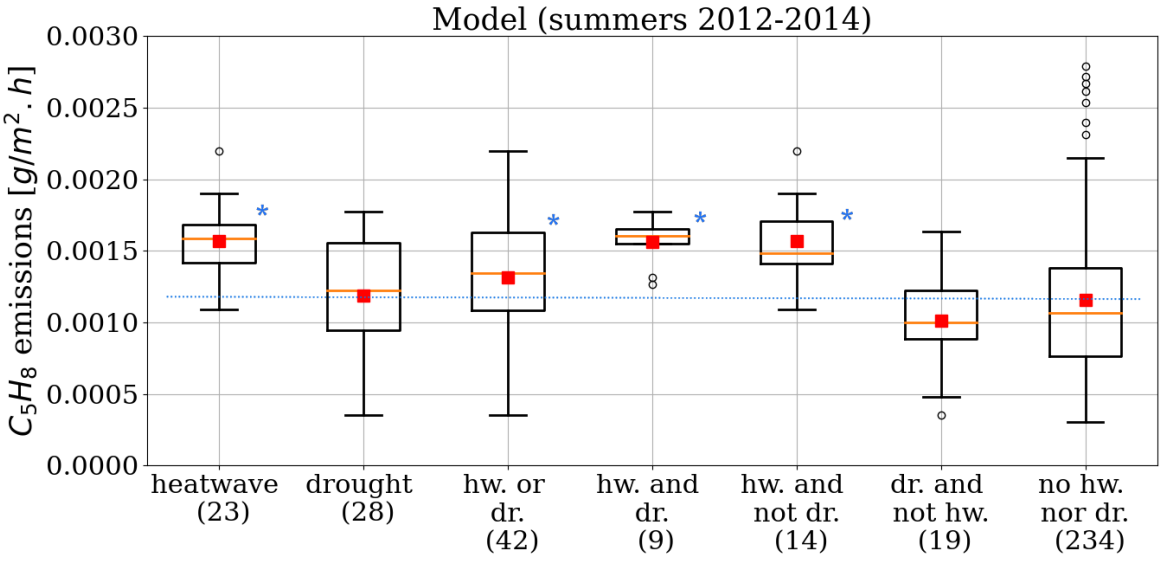

Figure 9. Daily maximum C_5H_8 emission rate [$g.m^{-2}.h^{-1}$] simulated by the MEGAN model ("γ_{SWSfit}-emiss" experiment) over South-western Europe, clustered by identified extreme weather events (PLA_{T2m} and PLA_{SD} indicators from RegIPSL). The number of days is indicated in parentheses. The analyzed period is June-July-August 2012, 2013 and 2014, covering a total of 276 days. The red squares show the mean of the distribution and the black circles are the outliers. The blue dotted line indicates the mean value of the normal conditions ("no hw. nor dr." cluster) and the blue stars if the mean value of the considered cluster is significantly different (t-test, at least p < 0.1) from the normal conditions. The box covers the InterQuartile Range (IQR) between Q1 (25th percentile) and Q3 (75th percentile). The lower whisker is limited to a statistical minimum (Q1-1.5*IQR) and the upper one to a statistical maximum (Q3+1.5*IQR).

ter "drought and not heatwave"), the mean daily maximum C_5H_8 emission rate is lower than normal conditions ($-0.02 \times 10^{-2} g.m^{-2}.h^{-1}$, non-significant difference). These results are in general agreement with the observed $HCHO$ total column by satellite instrument, used as proxy of BVOC emissions variation (see Sect. 4.2.2).

Weather conditions are considerably different between droughts combined to heatwaves and those which are not (SI: Fig. S9). The combined (resp. isolated) droughts are characterized by a 2m temperature of 23.9°C (resp. 22.6°C), a shortwave radiation of 342.2W/m^2 (resp. 296.0W/m^2) and a cloud cover of 1.8% (resp. 3.1%). Those weather variables are used for the computation of the activity factors γ_P and γ_T, thus directly affecting emissions. During isolated droughts, γ_{LAI} (0.51) and γ_{SWS} (0.89) are smaller than for normal conditions (0.48 and 0.76 respectively, significant difference for both). Nevertheless, this negative signal is not large enough for significant variation of the emission rates (compared to normal conditions).

Gathering worldwide data from experimental measurements of biogenic emissions under different climate drivers, the scientific review presented by Feng et al. (2019) (based on 74 articles) estimated to +53% the emission change of C_5H_8 during warm conditions and -15% during dry conditions. Those variations are close to what we simulated (+35% and -13% respectively).

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4.2.2 $HCHO$ total column

Based on the "Reference" and "NoBio-emiss" CHIMERE simulations, we computed that biogenic emissions contribute between 60 and 80% of the $HCHO$ concentration over Southwestern Europe. Variations of $HCHO$ concentration may therefore be used as indicator of BVOC emission variations during droughts and heatwaves. This allows us to use satellite observations of $HCHO$, which is particularly interesting due to the lack of in-situ data. Observations of the $HCHO$ total column from the OMI instrument are used.

Table 3 presents the average difference of $HCHO$ ($\Delta HCHO$) during extreme events compared to normal conditions (no heatwave nor drought). Over summers 2005 to 2016, $HCHO$ is significantly higher during heatwaves for the Balkans, Pô Valley and Central Spain (+15% on average). ΔHCHO is also positive during droughts but to a lesser extent (+3% on average, non-significant for Central Spain only). However, isolated droughts induce a significant decrease for the Balkans (-7%), Pô Valley (-6%) and Central Spain (-6%). Those results are consistent with the variation of C_5H_8 emissions simulated by MEGAN when a soil moisture parameter is taken into account (see Sect. 4.2.1). Wang et al. (2021a) also report a significant $HCHO$ column decrease (up to -30%) induced by a prolonged drought in a forested area in China.

The observed and simulated $HCHO$ total columns have been compared for the "γ_{SWSfit}-emiss" experiment over summer 2012. Time series showing $HCHO$ evolution for the three areas of interest are shown in SI (Fig. S10). Simulated $HCHO$ columns are generally higher than those from observations, especially for the Balkans. As mentioned in Sect. 2.2, uncertainty on the observations is large (between 30 and 100%) with large spatial variability (about a factor of two larger than in CHIMERE). HCHO products from OMI also present a systematic low mean bias (20-51%) (Zhu et al., 2016). However, the differences could be attributed to wrong specifications of land cover, and thus of the EF (e.g. Curci et al., 2010). Temperate tree PFTs are characterized by high C_5H_8 emission rates (respectively 10 000 and 600$\mu g.m^{-2}.h^{-1}$ for broadleaf and needleleaf types), compared to grassland (800$\mu g.m^{-2}.h^{-1}$) or cropland (1$\mu g.m^{-2}.h^{-1}$) (Guenther et al., 2012). After aggregation of the USGS land cover classes, the vegetation type assumed in CHIMERE in the Balkans is 57% of forest cover, 9% grassland and 33% cropland. Using the MODIS MCD12 product (Friedl et al., 2010), we find a different distribution, with 30% of forest cover, 25% grassland and 31% cropland. Finally, $HCHO$ is a product of oxidation of $VOCs$ that have not been fully evaluated, making more difficult to pinpoint the main sources of error on $HCHO$.

The comparison between CHIMERE and OMI of $HCHO$ total column variations during drought and heatwave days relies on very few cases. It is therefore difficult to support conclusions with a sufficient level of certainty. Nevertheless, CHIMERE simulations are more sensitive to temperature than OMI observations (+52.2% and +27.7% during heatwaves respectively, averaged over the three areas of interests). Since the summer 2012 was affected by long agricultural droughts, the inclusion of a soil dryness parameter in CHIMERE ("γ_{SWSfit}-emiss" experiment) reduces the simulated $HCHO$ peaks (SI: Fig. S10). However, the mean $\Delta HCHO$ during heatwaves and droughts in CHIMERE remain very similar with the "γ_{SWSfit}-emiss" experiment compared to the "Reference" one (decrease of -2% and -1% respectively). The temporal correlation does not vary significantly either (e.g. R coefficient around 0.5 for the Balkans).

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Table 3. Variation of HCHO total atmospheric column [$molecules/cm^2$] (ΔHCHO) due to heatwaves, droughts and isolated droughts in comparison to normal conditions (no heatwaves nor droughts) for summers (JJA) between 2005-2016 (measurements at 1pm). Summer 2012 is compared with CHIMERE simulations. Results are computed for each pixels and averaged over areas of interests: the Balkans, Central Italy and Central Spain. Stars mean that the difference with normal conditions is statistically significant (t-test, p < 0.1).

	OMI (2005-16)	OMI (2012)	CHIMERE (2012)	
			Reference	γ_{SWSfit}-emiss
Balkans (norm. cdt.)	5.74e15 $mol./cm^2$	4.02e15 $mol./cm^2$	7.64e15 $mol./cm^2$	7.62e15 $mol./cm^2$
Δ with heatwaves	+ 16.9 %*	+ 64.1 %*	+ 93.7 %*	+ 92.2 %*
Δ with droughts	+ 6.0 %*	+ 54.3 %*	+ 63.1 %*	+ 61.9 %*
Δ with isolated droughts	- 6.9 %*	+ 31.6 %*	+ 10.0 %*	+ 9.4 %*
Pô Valley (norm. cdt.)	5.86e15 $mol./cm^2$	5.33e15 $mol./cm^2$	7.59e15 $mol./cm^2$	7.52e15 $mol./cm^2$
Δ with heatwaves	+ 16.0 %*	+ 16.8 %*	+ 35.6 %*	+ 33.9 %*
Δ with droughts	+ 2.3 %*	+ 11.3 %*	+ 23.9 %*	+ 22.7 %*
Δ with isolated droughts	- 6.0 %*	- 0.9 %	- 2.6 %*	- 2.8 %*
Central Spain (norm. cdt.)	5.22e15 $mol./cm^2$	4.83e15 $mol./cm^2$	6.13e15 $mol./cm^2$	5.76e15 $mol./cm^2$
Δ with heatwaves	+ 12.2 %*	+ 2.2 %	+ 28.2 %*	+ 24.9 %*
Δ with droughts	+ 0.4 %	- 6.7 %*	+ 8.9 %*	+ 7.8 %*
Δ with isolated droughts	- 6.1 %*	- 10.8 %*	- 2.7 %*	- 3.0 %*

4.2.3 O_3 stomatal conductance

Surface weather conditions are critical for the stomatal conductance and therefore influence the dry deposition velocity. Figure 10 shows the maximum daily stomatal conductance of O_3 ("LAIdecr/f_{SWS}-dep" CHIMERE experiment) clustered by simulated extreme weather events and averaged over the Western Mediterranean. The same analysis has been performed on the dry deposition velocity and signals induced by extreme weather events are similar.

Droughts and heatwaves (isolated or combined) induce a significant decrease of the O_3 stomatal conductance, quantified at -25% for heatwaves and -35% for droughts compared to normal conditions. The activity factors mainly affected by droughts and heatwaves are f_{temp}, f_{VPD} and f_{SWS}.

The variation of f_{temp} during heatwaves depends on the magnitude of the events and so on their location, since the percentile of PLA indicator is defined for each grid cell. Over the Pô Valley and Balkans for instance, most heatwaves are characterized by temperatures close to optimal values of stomatal conductance that is fixed around 24°C (SI: Fig. S6). However, for those occurring in Central Spain (between 30° and 32°C), f_{temp} decreases by 7% compared to normal conditions. The temperature

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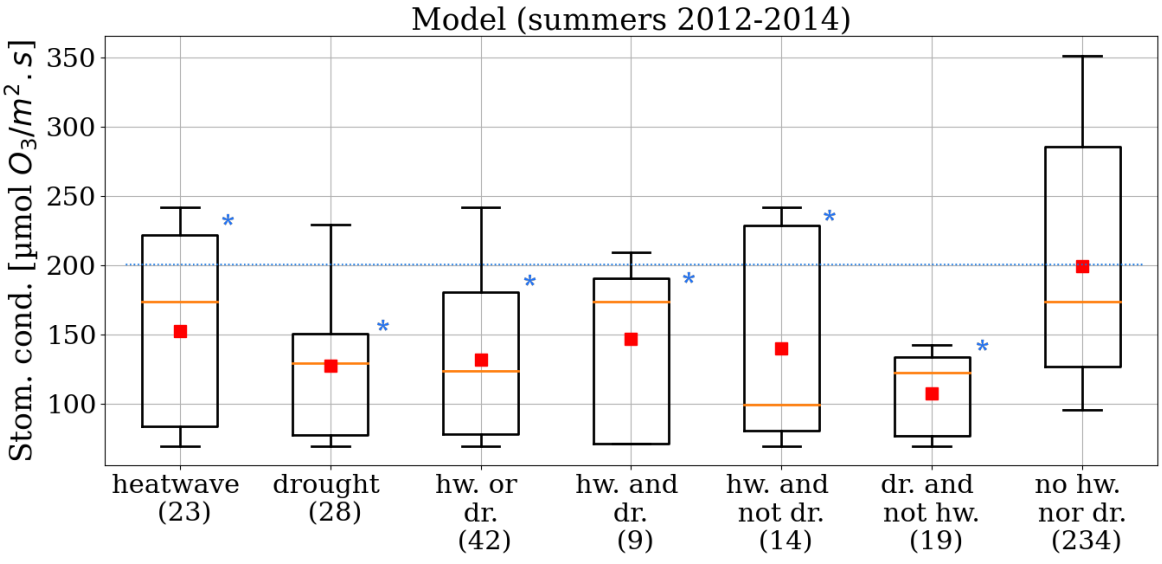

Figure 10. Same as Figure 9 with the simulated O_3 stomatal conductance [$\mu molO_3.m^{-2}.s^{-1}$] by the CHIMERE model ("LAIdecr/f_{SWS}-dep" experiment).

limit before complete stomatal closure is set at 40°C. Therefore exceptional heatwaves occurring in Southern Spain for instance could lead quickly to an accumulation of O_3 at the surface. f_{VPD} that depends both on temperature and relative humidity; significantly decreases during droughts and heatwaves (e.g. -6% averaged over the Southwestern Europe and -6% over the Pô Valley). Finally, f_{SWS} is the factor dominating the signal of variation of stomatal conductance. At the Southwestern Europe scale, this factor is the lowest during isolated droughts with a mean decrease of -35%.

4.2.4 O_3 surface concentration

Figure 11 shows the distribution of the observed (summers 2000-2016 and 2012-2014) and simulated (summers 2012-2014) daily maximum surface O_3 concentrations over Southwestern Europe for each cluster of extreme events. Observed O_3 (upper panel) is significantly (t-test, p < 0.01) higher during heatwaves (+18$\mu g/m^3$) and droughts (+9$\mu g/m^3$) than during normal conditions. Considering all droughts over the United States of America, Wang et al. (2017) also computed a mean increase in surface O_3 (+17% compared to the average). During isolated droughts, daily maximum O_3 (+4$\mu g/m^3$) is larger but the difference is non-significant.

The distribution of the simulated surface O_3 ("all-emiss-dep" experiment) by extreme events over the period 2012-2014 (lower panel) presents similar signals but of lower magnitude: +9$\mu g/m^3$ during heatwaves, +3$\mu g/m^3$ during droughts and non-significant difference during isolated droughts compared to normal conditions. Based on the results discussed above, the difference between the "heatwave" and "isolated drought" cluster could be explained by the different conditions of biogenic

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emissions, dry deposition, temperature and light. However, observations over the same period (middle panel) present a significant increase of the daily maximum O_3 during isolated droughts (+9$\mu g/m^3$), unlike what we simulated. The C_5H_8 emission reduction effect during such extreme event could be counterbalanced in a larger extent by the O_3 dry deposition decrease. It could also be explained by an underestimated impact of the enhanced photochemistry in the simulations, as we simulated favourable weather conditions during both combined and isolated droughts.

In summary, the variation of canopy-troposphere interactions (biogenic emissions and dry deposition) simulated by the MEGAN and CHIMERE models during droughts and heatwaves presents a consistent signal with respect to ozone observations (except for the isolated droughts over summers 2012-2014). Meteorological conditions are critical for the O_3 budget especially during summer droughts and heatwaves. In addition to uncertainties in the modeling of precursor emissions and O_3 deposition (as mentioned above), differences between observations and simulations may also rely on meteorological uncertainties, such as the diurnal temperature cycle (see Sect. 3.3.4) or the planetary boundary layer height.

4.3 Threshold level exceedance of O_3

In the European Union, the air quality standard for O_3 exposure is set at a daily maximum concentration of 120 $\mu g/m^3$ (8h-average) (EEA, 2020). Figure 12 shows the number of days above this threshold based on the AQ e-Reporting observations and the CHIMERE "all-emiss-dep" simulations for summers 2012, 2013 and 2014. Summers 2012 and 2013 present a large number of days exceeding the standard concentration. Even if exceedances occur in similar regions for observations and simulations, the number of days is generally larger in the observations. This is due to the overall underestimated daily maximum in CHIMERE compared to observations (see Sect. 3.3.4). For instance, in the Pô Valley, the most affected region in Southwestern Europe, around 60 exceeding days were observed and 50 were simulated over summer 2012 (significantly more than half of the summer). This region is known for its highly polluted air (O_3 and other pollutants) due to high anthropogenic emissions and unfavourable topographic and meteorological conditions for pollutants dispersion (e.g. Bigi et al., 2012). Other regions affected by O_3 peaks can be highlighted: Southeastern France, central Spain and central Italy for both summers 2012 and 2013.

Table 4 presents the distribution characteristics of stations with at least one day above the EU standard during summer period. On average over 2000-2016, this concerns 54% of EEA stations over the Western Mediterranean. The average number of exceedance days per station is 27 (almost a third of the summer period). Summer 2012 is above the 2000-2016 average, with 61% of stations affected and 28 days on average per station. For the same year, co-located values from CHIMERE simulations ("all-emiss-dep" experiment) are lower: 57% of stations and 18 days on average.

Over all summers between 2000 and 2016, 34% of the exceeding days occurred during heatwaves (with a mean exceeding value of 24$\mu g/m^3$) and 27% during droughts (+18$\mu g/m^3$). The number of days decreases by 13% if we consider only the isolated droughts (14%, +15$\mu g/m^3$). Summer 2012 was affected by exceptional droughts and heatwaves, resulting in high O_3 pollution. Around 80% of days above the EU threshold occurred during heatwaves or droughts for both observations and simulations.

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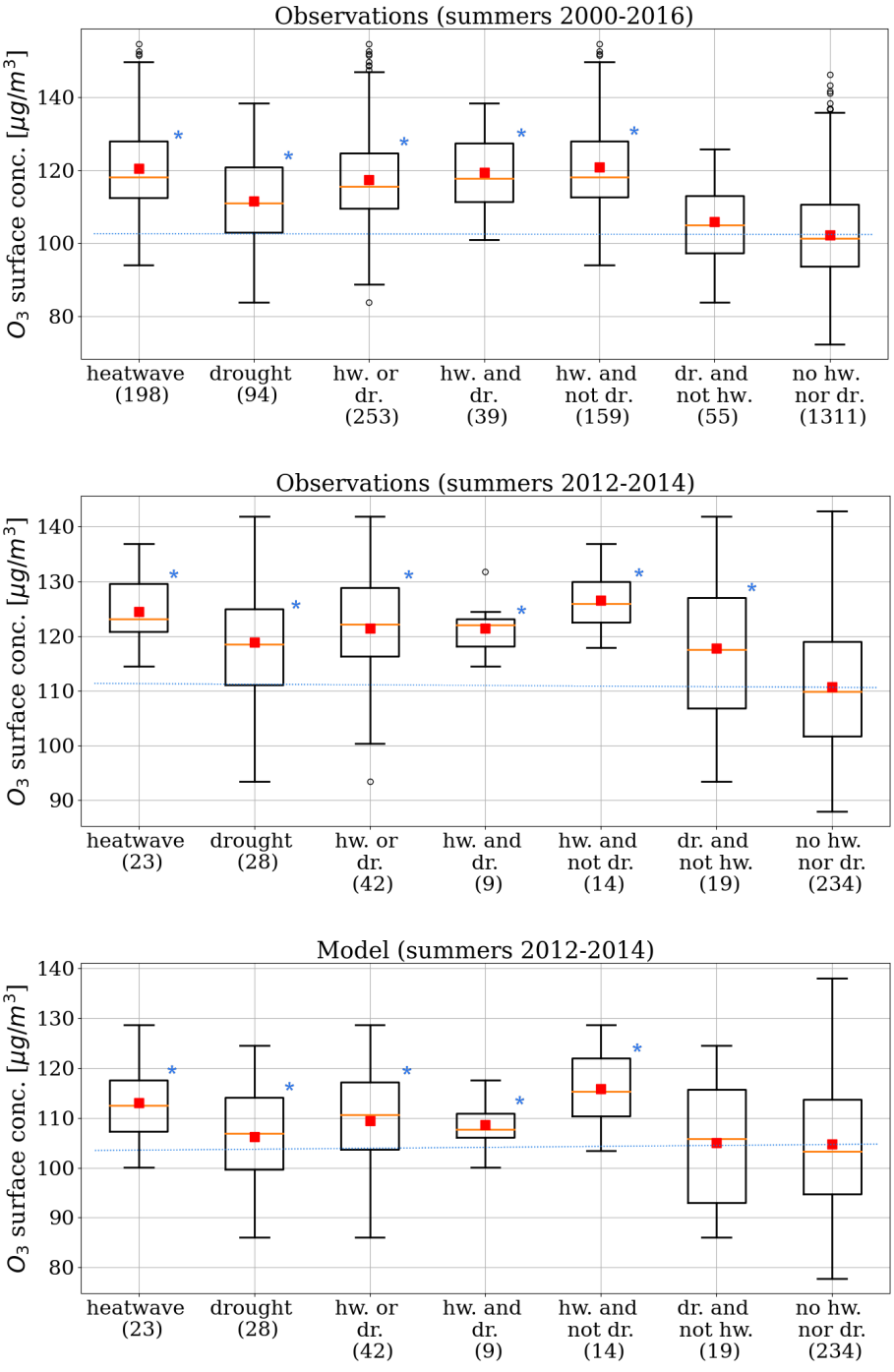

Figure 11. Same as Figure 9 with the observed surface O_3 concentration [$\mu g/m^3$] over the summers 2000-2016 (upper panel) and 2012-2014 (middle panel), and with the simulated surface O_3 ("all-emiss-dep" experiment) over the summers 2012-2014 (lower panel).

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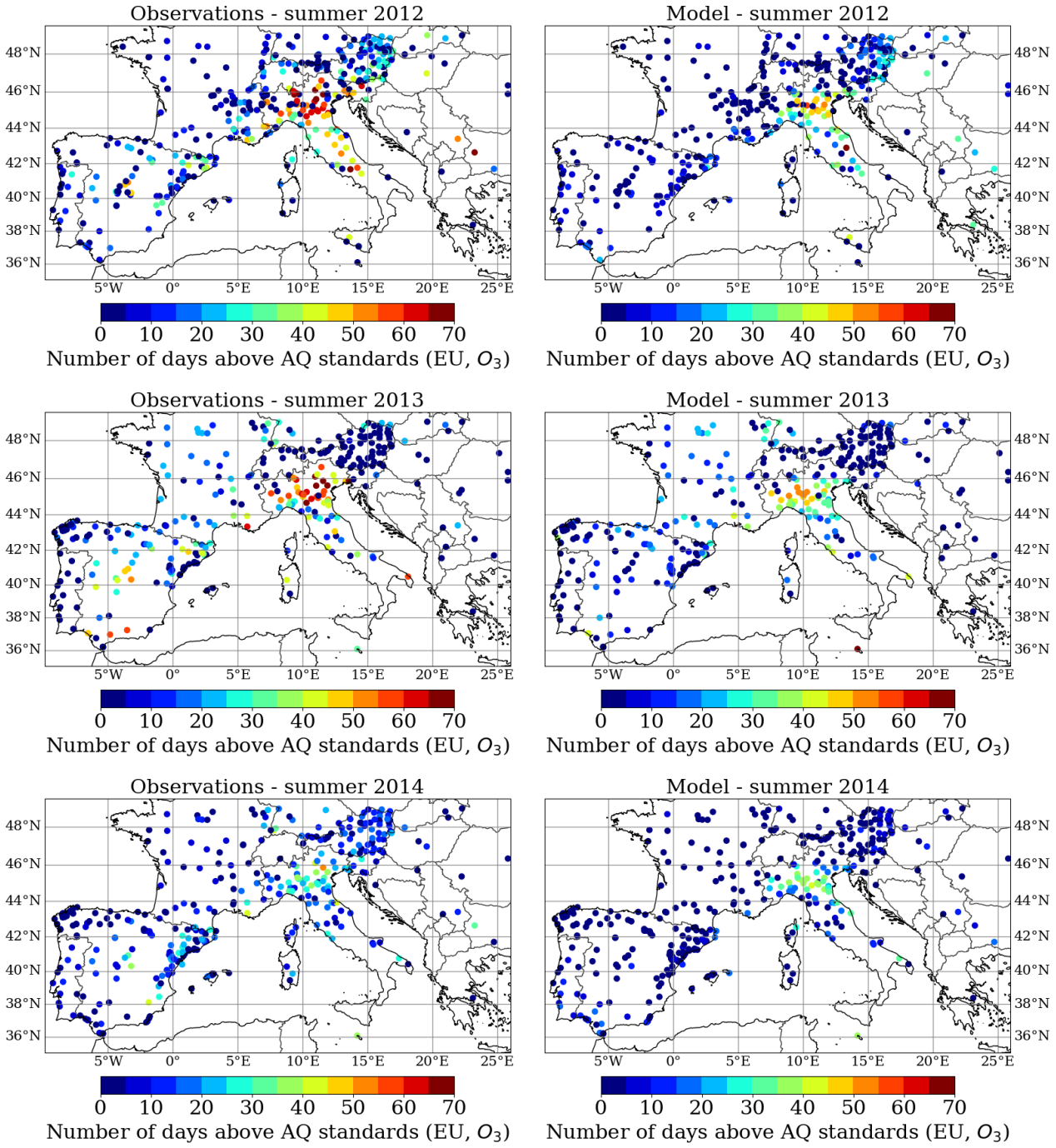

Figure 12. Number of days above the air quality threshold value from the European Union for surface O_3 (maximum daily 8 hour mean of 120 $\mu g/m^3$) over the summer 2012, 2013 and 2014 (JJA, 92 days in total). EEA observations (left column) are compared to CHIMERE ("all-emiss-dep") simulations (right column).

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Table 4. Average percentage of stations with at least one exceedance day (first row) regarding EU standard during summer (JJA). Considering only those stations, the second row shows the average number of exceeding days per station and the lower rows the average distribution of days above the EU standard in function of extreme weather events. The mean exceedance concentration is indicated in parenthesis.

	EEA (2000-2016)	EEA (2012)	CHIMERE (2012)
Average fraction of stations (over all):	54%	61%	57%
Average number of exc. days / station:	27 days	28 days	18 days
Average distribution of exc. days:			
Normal conditions	52% (+17$\mu g/m^3$)	20% (+15$\mu g/m^3$)	21% (+15$\mu g/m^3$)
Heatwaves or droughts	48% (+22$\mu g/m^3$)	80% (+18$\mu g/m^3$)	79% (+17$\mu g/m^3$)
Heatwaves	34% (+24$\mu g/m^3$)	58% (+20$\mu g/m^3$)	65% (+18$\mu g/m^3$)
Isolated heatwaves	21% (+24$\mu g/m^3$)	16% (+21$\mu g/m^3$)	18% (+16$\mu g/m^3$)
Droughts	27% (+18$\mu g/m^3$)	64% (+18$\mu g/m^3$)	61% (+17$\mu g/m^3$)
Isolated droughts	14% (+15$\mu g/m^3$)	22% (+14$\mu g/m^3$)	14% (+15$\mu g/m^3$)

5 Conclusions

The analyses presented in this study were organized around two main objectives. The first one was to assess the sensitivity of biogenic emissions, O_3 dry deposition and surface O_3 to the biomass decrease and soil dryness effect in a CTM model. The extremely dry summer 2012 was chosen and simulations were performed using the MEGAN v2.1 and CHIMERE v2020r1 model. We showed that soil dryness is a key factor during drought events, decreasing considerably the C_5H_8 emissions and O_3 dry deposition velocity. This effect has a larger impact than the biomass decrease. However, the resulting effect on surface O_3 remains limited.

In addition to the soil moisture activity factor used in MEGAN v2.1 that is mainly based on the wilting point (γ_{SM}), we proposed an innovative activity factor based on a soil water stress function (γ_{SWS}) simulated by the land surface and vegetation model ORCHIDEE. The latter induces a larger reduction of C_5H_8 emissions, but with more homogeneous patterns that follow the drought indicator. Furthermore, we adjusted this factor with a function fitted (γ_{SWSfit}) from experimental measurements from Bonn et al. (2019). By comparing the simulated surface concentration of C_5H_8 with observations at the Ersa measurement site (Corsica), γ_{SWSfit} showed promising results. However, such evaluation should be carried out over several sites and several years in order to determine with greater certainty the added value of this approach.

The second objective of this study was to quantify the variation of surface O_3 over the Southwestern Europe during agricultural droughts, combined or not with heatwaves. Agricultural droughts and heatwaves as extreme weather events were identified based on the coupled WRF-ORCHIDEE regional model (RegIPSL) using the Percentile Limit Anomalies (PLA) indicator. During the period 2000-2016, 59% of summer drought days were not accompanied by heatwaves (isolated droughts). For the

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summers 2012-2014, analyzed more specifically in this study, the 2m temperature is on average 5.5% lower during isolated droughts compared to all droughts, the shortwave radiation is 13.5% lower and the cloud fraction is 42% higher. As a result, surface O_3, but also BVOC emissions and O_3 dry deposition velocity, are substantially different if the drought considered is accompanied by a heatwave or not.

Based on a cluster approach using the PLA indicator, we showed that observed surface O_3 (summers 2000-2016) is larger by +18$\mu g/m^3$ in daily maximum during heatwaves and by +9$\mu g/m^3$ during droughts, compared to normal conditions. Despite a difference of several $\mu g/m^3$, CHIMERE correctly simulates the variations of O_3 concentration between the clusters of extreme events. The overall increase of surface O_3 during both heatwaves and droughts would be explained by O_3 precursor emission enhancement (in agreement with $HCHO$ satellite observations), O_3 dry deposition decrease and favourable weather conditions, so that all these mechanisms lead to an accumulation of O_3. However, we simulated a decrease of C_5H_8 emissions (in agreement with $HCHO$ observations) during isolated droughts, resulting in a non-significant difference of surface O_3 compared to normal conditions (from both observations and simulations).

Finally, almost half of summer days (2000-2016 period) exceeding the EU standard of O_3 for air quality in Southwestern Europe occurred during droughts or heatwaves. However, this percentage can increase (up to 80%) for exceptionally dry and hot summers, like in 2012. Only 14% of the exceedance days occurred during isolated droughts.

The implementation of dynamical effects of droughts in the MEGAN - CHIMERE model contributes to a better representation of biosphere-atmosphere interactions. However, comparisons between simulated and observed surface O_3 still show large discrepancies. Important uncertainties appear to be related to BVOC emissions, especially about the land cover classification, to NO_x concentrations for which CHIMERE presents limited performance scores of validation, to O_3 deposition and finally to meteorological conditions (e.g. temperature and PBL height). Such uncertainties need to be addressed to improve the simulation of O_3 during the summer, and especially over the Southwestern Europe.

In addition, some recent knowledge about fundamental processes that have been endorsed for a better representation of surface-atmosphere interactions during extreme weather events are not yet integrated by much, if not all, of the modeling community. Among the many examples are the increased emission of monoterpenes and sesquiterpenes during the development of drought (e.g. Bonn et al., 2019; Peron et al., 2021) or the in-canopy chemistry that is ignored or approximated by the "big leaf" model approach (e.g. Clifton et al., 2020a).

In conclusion, we provide in this paper a detailed analysis of the drought and heatwave effects on modeled biosphere-troposphere interactions controlling surface O_3 concentration. Heatwaves, and droughts in a lower extent, induce a significant increase of the surface O_3. Soil dryness and biomass decrease, as specific effects of droughts, are key factors for the variations of C_5H_8 emissions and O_3 dry deposition over the Southwestern Europe. We emphasize the need for a more dynamical representation of interactions between vegetation, hydrology, meteorology and atmospheric chemistry in models in order to improve the simulation of summer O_3.

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Data availability. The data set of indicators of heatwaves and agricultural droughts ($PLA_{T2m/SD}$) is freely available at https://thredds-x.ipsl.fr/thredds/catalog/HyMeX/medcordex/data/Droughts_Heatwaves_1979_2016/catalog.html. The data from AQ e-Reporting can be found at https://www.eea.europa.eu/data-and-maps/data/aqereporting-9/aq-ereporting-products, from MODIS and OMI instrument at https://lpdaac.usgs.gov/data/, from NCEP at http://www.ncep.noaa.gov and from EBAS at https://ebas.nilu.no/data-access/.

Competing interests. The authors declare no competing financial interest.

Author contributions. AG, ST and JP: Conceptualization. AG, ST, AC, JP, AE and JL: Methodology. AG: Visualization. AG and ST: Writing of the original draft. AG, ST, AC, JP, AE and JL: Review and editing. All authors have read and agreed to the draft version of the manuscript.

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