WFR-CHIMERE MODELLING AS A TOOL OF OZONE RISK ASSESSMENT TO EUROPEAN

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Keywords: Forests, Ozone, risk assessment, AOT40, PODY

Abstract:

Tropospheric ozone (O3) produces harmful effects to forests and crops, leading to a reduction of carbon assimilation that, consequently, influences land sink and crop yield production.

To assess the potential negative O3 impacts to vegetation, the European Union uses the Accumulated Ozone over Threshold of 40 ppb (AOT40). This index has been chosen for its simplicity and flexibility in handling different ecosystems as well as for its linear relationships with yield or biomass loss.

However, AOT40 does not give any information on the physiological O3 uptake into the leaves since it does not include any environmental constraints to O3 uptake through stomata. Therefore, an index based on stomatal O3 uptake, which describes the amount of O3 entering into the leaves, would be more appropriate.

We compare different potential O3 risk assessments based on two methodologies (i.e. AOT40 and stomatal O3 uptake) using a framework of mesoscale models that produces hourly meteorological and O3 data at high spatial resolution (12 km) over Europe for the time period 2000-2005.

Results indicate a remarkable spatial and temporal inconsistency between the two indices, suggesting that a new definition of European legislative
standard is needed in the near future. Besides, our risk assessment based on AOT40 shows a good consistency compared to both in-situ data and other model-based datasets. Conversely, risk assessment based on stomatal O3 uptake shows different spatial patterns compared to other model-based datasets. This strong inconsistency can be likely related to a different vegetation cover and its associated parameterizations.
WFR-CHIMERE MODELLING AS A TOOL OF OZONE RISK ASSESSMENT TO EUROPEAN FORESTS

Running head: Potential impact of ozone on European forests.

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ABSTRACT

Tropospheric ozone (O$_3$) produces harmful effects to forests and crops, leading to a reduction of carbon assimilation that, consequently, influences land sink and crop yield production. To assess the potential negative O$_3$ impacts to vegetation, the European Union uses the Accumulated Ozone over Threshold of 40 ppb (AOT40). This index has been chosen for its simplicity and flexibility in handling different ecosystems as well as for its linear relationships with yield or biomass loss.

However, AOT40 does not give any information on the physiological O$_3$ uptake into the leaves since it does not include any environmental constraints to O$_3$ uptake through stomata. Therefore, an index based on stomatal O$_3$ uptake, which describes the amount of O$_3$ entering into the leaves, would be more appropriate.

We compare different potential O$_3$ risk assessments based on two methodologies (i.e. AOT40 and stomatal O$_3$ uptake) using a framework of mesoscale models that produces hourly meteorological and O$_3$ data at high spatial resolution (12 km) over Europe for the time period 2000-2005. Results indicate a remarkable spatial and temporal inconsistency between the two indices, suggesting that a new definition of European legislative standard is needed in the near future.

Besides, our risk assessment based on AOT40 shows a good consistency compared to both in-situ data and other model-based datasets. Conversely, risk assessment based on stomatal O$_3$ uptake shows different spatial patterns compared to other model-based datasets. This strong inconsistency can be likely related to a different vegetation cover and its associated parameterizations.
1. INTRODUCTION

In the last decades, atmospheric pollution increases over large areas of the Globe, as a consequence of the continuous increasing of anthropogenic emissions over industrialized regions (Cooper, 2010; Hartmann et al., 2013; Stevenson et al., 2013). Nevertheless, thanks to emissions reduction policies, air pollution showed a flattened trend over Northern America and Europe since 2000 (Hartmann et al., 2013). However, since air pollutants and precursors can be transported across hundreds and even thousands of kilometers, air pollution is able to cause damages in areas far away to the source of emissions. Therefore a clear understanding of the pollution impacts on ecosystems is needed to reduce the associated potential risk (e.g. Malley et al., 2015).

Among common air pollutants, ground-level ozone ($O_3$) is the most damaging to forests and crops (e.g. Paoletti, 2006; Paoletti & Manning, 2007) and frequently it reaches high concentrations over large regions of the world (Hartmann et al., 2013).

Tropospheric $O_3$ concentrations are primarily determined by emissions of $CH_4$, CO, $NO_x$ and volatile organic compounds, as well as by natural processes like lightning and downward transport from the stratosphere (e.g. Oltmans & Levy II, 1994; Stevenson et al., 2006; Sicard et al., 2009). Sicard et al. (2009) have indicated that the intercontinental transport of anthropogenic precursors from North America and Asia is an important source of $O_3$ within Europe, therefore changes in North American and Asian emissions may influence $O_3$ concentrations in the European upper and lower troposphere (Li et al., 2002; Derwent et al., 2004; Fiore et al., 2011).

Over Southern Europe, and particularly around the Mediterranean basin, because of regional road traffic and industrial emissions combined with hot and sunny climate (Dalstein & Vas, 2005), the surface $O_3$ concentrations are higher than other European regions (Jonson et al., 2001; Vestreng et al., 2009; Sicard et al., 2013) and represent a potential threat to vegetation (e.g. Sanz et al., 2000; Paoletti, 2006; Wittig et al., 2009; Anav et al., 2011). Several studies suggest that tropospheric $O_3$ could cause reductions in crop
yield and forest production ranging from 0% to 30% (Ren et al. 2007; Sitch et al., 2007; Wittig et al., 2009; Anav et al., 2011; Tang et al., 2013). In addition, high O₃ concentrations affect vegetation by decreasing both foliar chlorophyll content into the leaves and photosynthesis, leading to an alteration of carbon allocation in the different pools (e.g. Dalstein & Vas, 2005; Bytnerowicz et al., 2007; Karnosky et al., 2007; Sitch et al., 2007; Wittig et al., 2009; Mills et al., 2011a; Anav et al., 2011; Fares et al., 2013; Feng et al., 2015). Besides, O₃ accelerates leaf senescence (Gielen et al., 2007), changes susceptibility to abiotic and biotic stress factors (Karnosky et al., 2002) and makes sluggish or impaired response of stomata to environmental stimuli (Hoshika et al., 2015).

Currently, the European standard (Directive 2008/50/EC) used to protect vegetation against negative impacts of O₃ is the Accumulated Ozone over a Threshold of 40 ppb (AOT40). It is a numerical index that describes the hourly O₃ accumulated exposure of ecosystems (UNECE, 2010). Exposure is generally limited to the period when stomata are open and AOT40 is, for simplicity, calculated for daylight hours (8AM-8PM) during the growing season (April to September). However, one can argue that different exposure periods should be recommended to allow differences in phenological development: for instance, in the Mediterranean region plants are often active during the winter months, with limited conductance during the summer as a result of water limitation (Paoletti, 2006). For this reason, UNECE (2010) recommends accumulation over different daylight hours (e.g. global radiation > 50 W/m²) or time-windows (e.g. 6-month growing season), depending on the plant receptor. Besides, as suggested by Klingberg et al. (2014), AOT40 does not consider the potential influence of climate change on the length of the growing season. This is particularly true in cold–temperate climates, where rising temperatures will extend the length of the growing season (e.g. Piao et al., 2007, 2015) and thus the duration of the period during which plants can uptake O₃.

Recently, the European Union (EU) is moving towards an index based on stomatal O₃ flux (or uptake) (Ashmore et al., 2004), where the functional (or physiologically effective) “dose” of O₃ to which plants
are exposed is defined as Phytotoxic Ozone Dose with a hourly threshold $Y$ (PODY).

Since $O_3$ effects on vegetation depend not only on the atmospheric concentrations but also on $O_3$ uptake through the stomata (e.g. Musselman et al., 2006; Matyssek et al., 2007), the stomatal $O_3$ flux approach provides an estimate of the critical amount of $O_3$ entering the stomata and has the capacity of accounting for environmental conditions that influence the $O_3$ uptake through stomatal aperture (e.g. Emberson et al., 2000). Thus, unlike AOT40, this method integrates multiple climatic stressors, so that its use is relevant in situations where either high $O_3$ concentrations are associated with environmental conditions that are unfavorable to uptake (Paoletti, 2006) or low concentrations are associated with mild and wet conditions (Matyssek et al., 2007).

A key tool within the European air pollution abatement strategy and legislation work is the chemical transport model of the European Monitoring and Evaluation Programme (EMEP, Simpson et al., 2012). The EMEP model is used for studies on risks and damages caused by air pollution and for integrated assessment modeling (Tuovinen, 2009). The EMEP model simulates emission, transport, transformation, removal of pollutants, and stomatal $O_3$ fluxes (Simpson et al., 2012). The first estimates of stomatal $O_3$ flux at the European scale were presented by Emberson et al. (2000), and made use of the EMEP Lagrangian photo-oxidant model. Since these early calculations, the deposition model (DO$_3$SE, for Deposition of Ozone and Stomatal Exchange) and the EMEP photo-oxidant model were improved (Tuovinen et al., 2004; Simpson et al., 2012).

However, to estimate PODY the EMEP model does not take into account the $O_3$ uptake limitation due to scarce soil water content (Tuovinen et al., 2009), a key variable for water-limited ecosystems (dry and semi-dry habitats), covering 41% of Earth's land surface (Reynolds et al., 2007). In addition, due to its coarse resolution (about 50 km), the EMEP model might not accurately reproduce the right pattern of climate variables, especially in regions of complex topography or over specific surface cover (e.g., coastal regions, mountains and sparse vegetated points). Finally, it does not take into account different forest
trees, but only two generic categories (Tuovinen et al., 2009), i.e. deciduous forest with characteristics based largely upon beech and evergreen forest based largely upon Holm oak.

In view of limitations in the AOT40 definition as well as in the EMEP model, currently used within the EU to evaluate the potential O₃ risks for European forests, we built up a multi-model framework able to generate results for use in integrated assessment modeling, and for studies on potential risks caused by O₃ pollution on European forests. This framework relies on a mesoscale model that generates climate forcing used offline to run a Chemistry Transport Model (CTM). The O₃ concentrations computed through the CTM and the climatic variables computed by a regional weather forecast model allowed us identifying the European forest that are mostly subjected to potential O₃ risks. In order to produce risk assessment maps, we performed continuous individual runs with both models at high spatial resolution (12 km) over Europe for the time period 2000-2005.

The main objectives of this study are to (i) provide a revised version of AOT40 and (ii) assess spatial and temporal consistency between risk assessment based on AOT40 and PODY. Besides, we compare our results with EMEP data to evaluate any different picture of potential risk damages.

2. MATERIALS AND METHODS

2.1 The atmospheric module

The Weather Research and Forecasting (WRF) is a limited-area, non-hydrostatic, terrain-following eta-coordinate mesoscale model (Skamarock et al., 2008). In this study, the version 3.6 with Advanced Research WRF (ARW) dynamic core is implemented over Europe. The model domain (Figure S1) covers almost all Europe (except northern Scandinavia and Iceland) and part of North Africa, and it is projected on a Lambert conformal grid, with a horizontal resolution of 12 km and 30 vertical levels extending from the land/sea surface up to 50 hPa (about 20 km). The initial and boundary meteorological conditions are
provided by the European Centre for Medium-range Weather Forecast (ECMWF) analyses with a horizontal resolution of 0.7° every 6 hours (Dee et al., 2011).

One useful capability of WRF is its flexibility in choosing different dynamical and physical schemes; the main options for physical schemes used in this study are listed in Table S1.

2.2 The atmospheric chemistry module

CHIMERE is an Eulerian offline chemistry-transport model developed to simulate gas-phase chemistry, aerosol formation, transport and deposition at regional scale (Bessagnet et al., 2004; Menut et al., 2013). In our configuration CHIMERE (version 2013b) has a spatial resolution of 12 km and uses 8 vertical levels of increasing thickness away from the ground defined in hybrid sigma-pressure coordinates, with the first level at 0.997 sigma-level (about 20-25 m above the ground) and the top of the last level at 500 hPa. The model domain is shown in Figure S1.

The gas-phase chemical mechanism used by CHIMERE is MELCHIOR2 (Lattuati, 1997), which consists of a simplified version (40 chemical species, 120 reactions) of the full chemical mechanism MELCHIOR; this latter describes more than 300 reactions of 80 species. Photochemical rates are calculated as a function of height using the Tropospheric Ultraviolet and Visible (TUV) radiation model (Madronich et al., 1998). External forcing such as meteorological fields, primary pollutant emissions, and chemical boundary conditions are required to calculate the atmospheric concentrations of tens of gas-phase and aerosol species. Meteorological fields required by CHIMERE, such as wind, temperature, cloud liquid water content, surface heat fluxes, cloud cover, and precipitation, are directly provided by the WRF simulations. Besides, to accurately reproduce the gas-phase chemistry, emissions must be provided every hour for the specific species of the chemical mechanism. For studies over Europe, the EMEP inventory (Vestreng, 2003) is usually used for emissions of NOx, CO, SO2, PM2.5 and PM10. Since EMEP data are given as annual emissions, the annual fields are transformed in hourly fluxes, as described by Menut et al. (2012).
Biogenic emissions of 6 species (isoprene, α-pinene, β-pinene, limonene, ocimene, and NO) are calculated through the MEGAN model (Guenther *et al.*, 2006). Boundary conditions are provided as a monthly climatology of the LMDz-INCA global chemistry-transport model (Hauglustaine *et al.*, 2004; Folberth *et al.*, 2006) for gaseous species and the GOCART model (Ginoux *et al.*, 2001) for aerosol species. More details regarding the parameterizations of the above mentioned processes are described in Menut *et al.* (2013).

### 2.3 Modelling AOT40 and stomatal ozone flux across Europe

The chemistry transport model provides O$_3$ concentrations at 20-25m height, so we can consider these values as given at the top of a forest canopy, providing a reasonable estimate of O$_3$ concentrations at the sunlit upper canopy leaves (UNECE, 2010). Therefore, we can use these O$_3$ concentrations for determining AOT40 and PODY, without using any deposition model (Wesely *et al.*, 1989; Seinfeld & Pandis, 2006). This assumption is valid only in case of forest trees and using a "big-leaf" approach (Friend, 2001; Dai *et al.*, 2004), while for grassland or cropland a deposition model must be used to scale the O$_3$ concentration at the canopy height (typically < 2m) taking into account the surface-layer turbulence (Seinfeld & Pandis, 2006).

The AOT40 is defined as the sum of the exceedances above 40 ppb computed over the growing season (i.e. from April to September) and for the daylight hours (UNECE, 2010). The daylight hours can be defined in different ways: i) as the time when the solar zenith angle is equal to or less than 89° (Simpson *et al.*, 2012), ii) as the clear-sky global radiation exceeding 50 W m$^{-2}$ (UNECE, 2010), iii) as a flat time-window from 8 AM to 8 PM (UNECE, 2010; Klinberg *et al.*, 2014). We consider the flat time-window methodology physiologically unsubstantiated, since plants do not sharply open (close) their stomata at 8AM (8PM). Moreover, during summer mainly in the semi-arid environments, such as the Mediterranean region, radiation and temperature before 8AM are strong enough to promote carbon assimilation by plants.
(e.g. Fares et al., 2013). This means that stomata are open during the first hours of the morning while they are closed during the warmer hours of the day, suggesting that during these hours O$_3$ is not absorbed by leaves and thus it does not produce any damage. In addition, plants in the Mediterranean region are often active during winter months (Paoletti, 2006), but with limited conductance values during summer as a result of water limitation. This suggests that computing AOT40 only over the growing season (i.e. April-September) would not take into account the possible O$_3$ damages at the beginning or end of the growing season for deciduous trees or year round for evergreen trees. We have estimated over different eddy covariance sites (Reichstein et al., 2005) of evergreen vegetation that computing the GPP only over the period April-September leads to a reduction of GPP of about 30-40%, while over some deciduous broadleaves forests we found a reduction <8% (data not shown). Similarly, excluding all the months outside this time range would lead to an underestimation of AOT40 up to 50% for conifer trees, while in case of deciduous trees the underestimation is much smaller (0-5%, as discussed later).

Given the above limitations, we compute AOT40 year-round for all forests and when the stomatal conductance ($g_{sto}$) is higher than 0. Since deciduous broadleaves forests do not have any remarkable activity during winter, we believe that using the whole year for the AOT40 computation does not considerably affect its magnitude. However, as soon as the environmental conditions become favourable and the growing season starts, deciduous trees start to uptake O$_3$. Thus, compared to the original UNECE (2010) formulation, our proposed revision is more flexible since it allows to account for the effect of inter-annual variations and climate change, namely for deciduous forests it can take into account whether or not the green-up and dormancy dates are delayed due to changing (or varying) climate (e.g. Menzel et al., 2006; Schwartz et al., 2006; Piao et al., 2007; Wolkovich et al., 2012; Fu et al., 2014; Piao et al., 2015; Wang et al., 2015). Similarly, an AOT40 computed when stomata are open is more adequate than an index computed only between 8AM and 8PM or for hours with a solar radiation exceeding 50 W m$^{-2}$. However, one could argue that rarely before 8AM O$_3$ concentrations exceed 40 ppb, hence our proposed
change may not significantly influence the magnitude of AOT40. Nevertheless, this approach is more plausible from a physiological point of view as well as more flexible since it allows to account for extreme events (e.g. heat waves), impact of wildfires that may significantly increase the O\textsubscript{3} concentrations (Turquety et al., 2014), or anomalous years. In addition, this method might be applied at global scale, or in highly polluted regions where O\textsubscript{3} can be systematically above 40 ppb all the day.

Given the above considerations, our revised AOT40 is defined as the accumulated amount of O\textsubscript{3} over the threshold value of 40 ppb, i.e.

\[
AOT40 = \int_{t=01-Jan}^{31-Dec} \max([O_3] - 40, 0) dt; g_{sto} > 0
\]

\[
0; g_{sto} = 0
\]

(1)

where [O\textsubscript{3}] is the hourly O\textsubscript{3} concentration (ppb), \(dt\) is the time step (1 hour) and \(g_{sto}\) is the stomatal conductance computed according to Equation 2. The function "maximum" ensures that only values exceeding 40 ppb are taken into account.

However, even if we modified the original formulation of AOT40 taking into account the stomatal conductance, there are still other limitations when using this index: in fact, it does not provide any information on the physiological O\textsubscript{3} uptake by leaves and it does not include any environmental constraint to O\textsubscript{3} uptake. It is well known that soil moisture and hence water stress play a pivotal role in the O\textsubscript{3} uptake by plants (Paoletti & Manning, 2007). As a consequence, AOT40 may be inadequate for the quantification of O\textsubscript{3} impacts on vegetation, particularly in water-limited regions (e.g. Paoletti, 2006; Paoletti & Manning, 2007; Matyssek et al., 2007).

Conversely, the stomatal flux-based approach integrates the effects of multiple climatic factors, vegetation characteristics and local and phenological inputs (Paoletti & Manning, 2007). The stomatal flux-based
model, or DO3SE model, is based on the Jarvis’ (1976) algorithm for calculation of stomatal conductance. This algorithm describes species-specific effects of soil water availability, vapour pressure deficit, air temperature, and solar radiation on stomatal functioning. The leaf-level stomatal conductance (g\text{sto}, in mmol O\text{3} m\text{2} s\text{1}) was estimated using this multiplicative model and the parameters suggested in UNECE (2010) for different forest types (Table 1):

\[
g_{\text{sto}} = g_{\text{max}} \times f_{\text{light}} \times \max(f_{\text{min}}, f_{\text{temp}} \times f_{\text{VPD}} \times f_{\text{SWC}}) \tag{2}
\]

where \(g_{\text{max}}\) is the maximum stomatal conductance of a plant species to O\text{3}, and \(f_{\text{min}}\) is the minimum stomatal conductance expressed as a fraction of \(g_{\text{max}}\). The other functions are limiting factors of \(g_{\text{max}}\) and are scaled from 0 to 1. \(f_{\text{light}}, f_{\text{temp}}, f_{\text{VPD}},\) and \(f_{\text{SWC}}\) are the variation in \(g_{\text{max}}\) with photosynthetic photon flux density (PPFD, \(\text{µmol photons m}^{-2} \text{s}^{-1}\)), surface air temperature (\(T, ^\circ\text{C}\)), vapour pressure deficit (VPD, kPa) estimated through the surface air humidity, and volumetric soil water content (SWC, m\text{3} m\text{3}), respectively.

The \(f_{\text{light}}, f_{\text{temp}}\) and \(f_{\text{VPD}}\) functions are expressed by the following formulations (Emberson et al., 2000; UNECE, 2010):

\[
f_{\text{light}} = 1 - e^{-(\text{light} \times \text{PPFD})} \tag{3}
\]

\[
f_{\text{temp}} = \left[\frac{T - T_{\text{min}}}{T_{\text{opt}} - T_{\text{min}}}\right] \times \left[\frac{T_{\text{max}} - T}{T_{\text{max}} - T_{\text{opt}}}\right]^{(\frac{T_{\text{max}} - T_{\text{opt}}}{T_{\text{opt}} - T_{\text{min}}})} \tag{4}
\]

\[
f_{\text{VPD}} = \min\left\{1, \max\left[f_{\text{min}}, \frac{(1 - f_{\text{min}}) \times (\text{VPD}_{\text{min}} - \text{VPD})}{\text{VPD}_{\text{min}} - \text{VPD}_{\text{max}}} + f_{\text{min}}\right]\right\} \tag{5}
\]
where light\(a\) is a species-specific light response constant, PPFD is hourly photosynthetic photon flux density, \(T_{\text{opt}}\), \(T_{\text{min}}\), and \(T_{\text{max}}\) represent the optimum, minimum, and maximum temperature for stomatal conductance, respectively, VPD\(_{\text{min}}\) and VPD\(_{\text{max}}\) are minimum and maximum Vapour Pressure Deficit for stomatal conductance, respectively. The values of parameters specific to different vegetation types were derived from UNECE (2010) and are listed in Table 1.

Unlike other risk assessments used as “worst case” scenario (e.g. Simpson et al., 2007; Tuovinen et al., 2009), we included the \(f_{\text{SWC}}\) function in the \(g_{\text{sto}}\) computation because is well known that soil moisture is critical for stomata opening/closure. The soil-water limitation function is defined as:

\[
f_{\text{SWC}} = \min\left[1, \max\left(f_{\text{min}}, \frac{SWC - WP}{FC - WP}\right)\right]
\] (6)

where WP and FC are the soil water content at wilting point and at field capacity, respectively; these two parameters are constant and depend on the soil type (Table S2).

Once the stomatal conductance is computed, the flux of \(O_3\) entering the leaves is accumulated over a species-specific phenological time window and expressed as PODY, where \(Y\) represents a detoxification threshold below which it is assumed that any \(O_3\) molecule absorbed by the plant will be detoxified (Mills et al., 2011a). Since the \(Y\) recommended threshold values are still under discussion within the scientific community (Assis et al., 2015), we test two different thresholds: 1 nmolO\(_3\) m\(^{-2}\) s\(^{-1}\) as recommended by UNECE (2010) and 0 nmolO\(_3\) m\(^{-2}\) s\(^{-1}\) as suggested by the fact that the detoxification processes are dynamic and cannot be modeled by a constant threshold value (Musselmann et al., 2006).

Unlike the European standards (UNECE, 2010) and similarly to our AOT40 definition, we computed the stomatal \(O_3\) uptake year round rather than over the growing season only. Thus, the stomatal \(O_3\) uptake (mmol m\(^{-2}\)) with a \(Y\) threshold of 0 (i.e. POD0) is expressed as:
where $g_{sto}$ represents hourly values of stomatal conductance and $[O_3]$ is the hourly O$_3$ concentrations (ppb). Similarly, the stomatal O$_3$ uptake with a Y threshold of 1 nmol m$^{-2}$ s$^{-1}$ (i.e. POD1) is expressed as:

$$POD1(t) = \max(POD0(t) - 1, 0)$$

It should be noted that the four meteorological variables in Equation 2 (i.e. air temperature, air humidity, solar radiation and soil moisture) are simulated by a mesoscale model (section 2.1), while O$_3$ concentrations are modelled through a chemistry transport model (section 2.2). In addition also the composition of the soil (Figure S1) and the dominant vegetation data (Figure S2) are required to estimate the stomatal conductance. The spatial distribution of trees is based on the European Forest Institute (EFI) data (Tröltzsch et al., 2009; Brus et al., 2011); however, because of lack of specific parameterizations for all the different tree species, we converted these original data to the DO$_3$SE categories (Table 1) using the European Environmental Agency (EEA) biogeographic regions (Figure S1).

### 2.4 Observation data

To assess the performances of CHIMERE in reproducing the ground-level O$_3$ concentrations over different years, we compared model outputs with in-situ observations from several background monitoring sites from EMEP ([http://www.emep.int](http://www.emep.int)) and AIRBASE ([http://acm.eionet.europa.eu/databases/airbase/](http://acm.eionet.europa.eu/databases/airbase/)) networks. More details on the station type classifications and the different measurement techniques are available through the AIRBASE and EMEP websites.
To avoid pitfalls in the model validation, among the many available measurement sites we selected the stations with a full daily data capture over the years. We also use a statistical model of gross primary production (GPP) based upon flux tower local observations, satellite fraction of absorbed photosynthetically active radiation (fAPAR) and climate fields (Jung et al., 2009; Beer et al., 2010; Jung et al., 2011) to assess the spatial consistency among our stomatal model and GPP. This model uses a Model Tree Ensemble (MTE) which is a machine learning system where the target variable (i.e. GPP) is predicted by a set of multiple linear regressions from explanatory variable (Jung et al., 2009; Beer et al., 2010; Jung et al., 2011).

3. RESULTS

Since the magnitude of AOT40 and PODY depends on climate and O\textsubscript{3} bias simulated by WRF and CHIMERE, in the next section we briefly show how CHIMERE reproduces the O\textsubscript{3} concentrations, while the validation of the meteorological forcing is discussed in the Supporting Information. In particular, the evaluation of forcing variables is needed to assess whether any abnormal value in PODY or AOT40 can be related to poor performances of models in reproducing key variables used to evaluate the risk or is mainly due to the general parameterization or vegetation map used for our large regional domains.

3.1. Performances of atmospheric chemistry model

Figure 1 displays the spatialized bias between CHIMERE results and observations. Looking at the figure, several considerations can be drawn. First, consistent with Terrenoire et al. (2015) CHIMERE systematically overestimates the surface ozone concentration. Second, there is a clear south-north gradient, with Mediterranean sites showing larger biases than continental stations. The highest annual mean concentrations are located below 45° of latitude where the strongest photolysis over Europe occurs: this partially explains the larger bias, namely the higher is the O\textsubscript{3} concentration, the larger may be the
bias, as confirmed by the mean normalized bias (MNB, not shown). Third, the bias does not significantly change during years, although from year-to-year we used a different number of stations. Four, surprisingly during the 2003 CHIMERE displays the lower bias, despite it was a very anomalous year (summer heat wave).

To assess how CHIMERE reproduces the temporal variability of daily surface O$_3$ concentrations, Figure 2 shows the spatialized correlation coefficient between CHIMERE outputs and in-situ observations. Consistently with Szopa et al. (2009), the highest correlations are found over central Europe where most of the sites show a correlation above 0.6, whereas worse results are obtained in the Mediterranean area.

To provide a synthetic view of CHIMERE performances in reproducing the daily O$_3$ variability, we compared observed and modeled concentrations averaged over all the available stations during a given year (Figure 3). The daily temporal variability of O$_3$ concentrations is well simulated, although CHIMERE tends systematically to overestimate the mean daily concentrations during the study period (bias ~11 ppb on average). Terrenoire et al. (2015) claimed that the overestimation of O$_3$ by CHIMERE is likely related to the NO$_2$ underestimation and the overestimated O$_3$ concentrations of the lateral boundary conditions used to force the model. However, we hypothesize that this overestimation is an artifact and might be related to the different heights between the lowest model layer and measurement data. Specifically, for the validation we used the O$_3$ concentrations at the lowest model layer (about 20-25m), while the sensors may be placed at different heights (typically 2 meters). Therefore, to provide a proper validation of the model, we should use the O$_3$ dry deposition computed by the model to scale down the concentration to the reference height using a logarithmic profile. However, since CHIMERE has already been extensively validated (e.g. de Meij et al., 2009; Menut et al., 2013; Martin et al., 2014), we compare the simulated O$_3$ concentrations from the first vertical level of CHIMERE with in-situ data.
Despite the observed differences in the mean values, it is noteworthy that extreme events, like 2003 heat wave, are well captured by the model, adding reliability to our framework to assess different inter-annual variations correctly.

3.2. Risk assessment for European forests

The AOT40 index (Figure 4) shows a clear latitudinal gradient with minimum values ranging between 2000-4000 ppb*h over large parts of UK, Southern Scandinavia and North-Western Europe, and maximum values exceeding 50000 ppb*h over Italy, Greece, South-eastern France and Southern Spain. The higher AOT40, observed in Mediterranean region, depends on the strongest photolysis rates due to the high temperatures typical of this area and on the high O$_3$ precursor emissions, leading to higher O$_3$ concentration in the lower troposphere (Sicard et al., 2013).

Most of the forested areas of UK and Northern Europe, characterized by minimum values of AOT40, are free from exceedances, i.e. they do not exceed the critical limit for forest protection set to 5000 ppb*h (UNECE, 2010), with a low risk to be subject to O$_3$ damages. Conversely, below 50°N all the European forests might be potentially damaged by O$_3$.

Our model results also indicate that the inter-annual variations of single-year AOT40 calculations are not very large (Figure 4): in fact, although the magnitude of AOT40 is slightly different over the 6 analysed years, the spatial pattern is homogeneous among different simulations, i.e. areas with maximum (or minimum) risk are always the same, whereas the Mediterranean region is systematically the most exposed to O$_3$ risk. During the anomalous year 2003, characterized by a strong heat wave in central and Southern Europe, it is clearly visible how AOT40 increases in terms of magnitude over the whole Mediterranean region. Similarly, continental forests are more exposed to O$_3$ risk with respect to other years, as pointed out by the decreasing south-north gradient.
Unlike AOT40, POD0 does not show any latitudinal gradient (Figure 5). We found the minimum values of about 12-14 mmol m\(^{-2}\) over Eastern Europe, while in the Mediterranean region and along the French Atlantic coasts, the maximum values exceed 40 mmol m\(^{-2}\).

Consistent with AOT40 results, the spatial pattern of POD0 is homogeneous among different years, although its magnitude considerably changes during the years. Notably, because of the heat wave that affected the Mediterranean area, in 2003 there is a relevant decrease in POD0 values: this strong reduction is mainly explained by the dryness of soils that leads to a stomatal closure during the high daily temperature maxima. In fact, the heat wave produced a strong water stress on plants that, in order to minimize the water loss, closed their stomata, leading also to a lower amount of O\(_3\) entering the leaves.

We also found that both AOT40 and POD0 indices have higher values along coastlines, because of the both lower deposition velocity of O\(_3\) over sea areas (Garland et al., 1980; Simpson et al., 2007; Ganzeveld et al., 2009; Coleman et al., 2010), and shipping tracks, industrial development, road traffic increment, high insolation and sea/land breeze recirculation (Sicard et al., 2013) leading to high O\(_3\) concentrations.

Comparing the AOT40 and POD0 values it is clear that the two indices highlight very different spatial and inter-annual distributions of risks. AOT40 suggests strong exceedances in Southern Europe with maximum over Italy and Greece, while POD0 suggests that the potentially mostly damaged forests are located over Atlantic regions of France, Spain and Portugal. This behaviour is chiefly evident during the anomalous year 2003: specifically, risk assessment based on AOT40 shows that in 2003 the most sensitive areas to potential O\(_3\) risks are located over the Southern part of the domain and that this index has its maximum value during this particular warm year. Conversely, risk assessment based on POD0 highlights an opposite temporal pattern, namely during 2003 the potential O\(_3\) damages are lower than in other years.

In addition, comparing AOT40 and POD0, as expected we found that POD0 shows higher values typical of relatively well watered environment, like north-west France and Atlantic region, even if associated to lower ozone concentration, while AOT40 shows higher values where the O\(_3\) concentrations are higher.
Given the opposite temporal pattern between AOT40 and POD0 as well as their spatial differences, one can question the reliability of these indices. Thus, in order to bear out the correctness of POD0 in reproducing the right spatial pattern, we compare the stomatal conductance (i.e. the driving variable of POD0) with an observation-based GPP dataset for three years (2001, 2003 and 2005). Before describing the differences in the spatial patterns between stomatal conductance and GPP, two considerations are needed. First, the GPP can be regarded as a proxy of stomatal conductance: in fact, in general the higher the stomatal conductance, the higher the photosynthesis. Second, our estimated stomatal conductance and the observational-based GPP are two independent datasets.

Comparing observation-based GPP with our stomatal conductance model (Figure 6), we found both datasets peak over Southern UK, Ireland and Atlantic coasts of France. In addition, the stomatal model well reproduces the spatial variability of GPP over Italy and Atlantic coasts of Spain and Portugal. Thus, the good consistency between the spatial patterns obtained by the stomatal model and the observation-based GPP suggests that results obtained by our framework represent the real condition in a satisfactory way.

3.3. Comparison with EMEP model and station data

To further investigate the performances of our framework, we compared AOT40 and POD1 with EMEP (v2013) model outputs (Simpson et al., 2012). Figure 7 shows the annual mean O₃ concentration as modelled by CHIMERE (upper panels) and EMEP (lower panels) over different years. The spatial pattern is consistent among models: in fact, both CHIMERE and EMEP simulate higher O₃ concentrations over the Mediterranean region, although CHIMERE predicts higher concentrations than EMEP, and lower concentrations in the northern part of the domain. Given the higher spatial resolution, the O₃ changes due to the orography are more evident in CHIMERE. It also should be noted that the mean annual concentration does not drastically change among years.
Figure 8 shows the AOT40 as simulated by CHIMERE (upper panels) and EMEP (lower panels) over three different years; besides, AOT40 values computed using local site measurements of O₃ are also displayed. Here, to be consistent with EMEP, we computed the AOT40 according to the original formulation (UNECE, 2010), namely between 8AM-8PM for the time period 1⁰ April–30⁰ September. In addition, it should be noted that the EMEP AOT40 is computed at a default height of 20m for the forests (Simpson et al., 2007), thus the two indices are highly consistent.

Compared to in-situ data, our AOT40 is slightly higher, but the spatial variation with latitude is well reproduced. The overestimation of AOT40 could arise from the overall positive bias of CHIMERE in reproducing the surface O₃ concentration (Figure 3).

Similarly, the AOT40 spatial pattern is consistent between our results and EMEP outputs, reflecting the similarity between the simulated surface O₃ concentrations. Although the annual mean O₃ concentration simulated by CHIMERE is higher than EMEP (Figure 7), it is remarkable how this latter shows a higher AOT40 than CHIMERE (Figure 8). However, since AOT40 resembles more the O₃ maxima than O₃ mean and it is accumulated during the growing season only, while Figure 7 shows the mean annual value, this result is not surprising.

Figure 9 shows the comparison between POD1 levels as simulated by our multi-model framework and EMEP over three different years. Unlike the AOT40, because of the lack of meteorological data needed to compute the stomatal conductance, we were unable to provide a detailed validation of POD1 against local measurements. A clear difference both in terms of magnitude and spatial distribution between the two models is evident, with EMEP predicting maximum risks over the whole Mediterranean region, while our results suggest that the most sensitive regions to potential O₃ risks are located over the Atlantic coasts of France.

When comparing the POD1 computed using the soil water limitation with EMEP, we found a large difference in magnitude (greater than 10 mmol m⁻² over the peak areas). Thus, one can argue that such a
difference might be explained by the soil water limitation function that is not taken into account into EMEP, leading to a low water-stress for vegetation and hence to higher canopy conductance. To disentangle the large difference in POD1 magnitude, we compute the POD1 neglecting the function relative to soil water content (middle panel). This leads to a substantial change in magnitude mainly over Southern Spain and Mediterranean region, where our POD1 results increased significantly. Nevertheless, a substantial difference between our results and the EMEP model remains over the whole study area. Given the similarity between \( O_3 \) data and since large biases in temperature do not cause large changes in POD1 (not shown), we believe one likely explanation for such a large mismatch is given by the different vegetation maps used to derive the stomatal conductance and the relative parameterizations used for each species.

4. DISCUSSION

Forests have several relevant functions for economy, nature conservation, environmental protection, and carbon sinks (Sicard et al., 2013). Thus, to protect forests against \( O_3 \) pollution, appropriate standards and realistic thresholds, representative of actual field conditions, are needed. Currently, the AOT40 index is used as standard for the protection of European vegetation, but there is now scientific consensus regarding the use of PODY as new standard. However, a validation of the threshold \( Y \) under field conditions is still missing. In fact one of the open issues to be answered in the near future is the lack (or scarce amount) of epidemiological studies looking at relationship between both indexes and damages in the field conditions; indeed previous comparison studies between AOT40 and PODY are based on observation in controlled experimental conditions (De Marco et al., 2010). Our results suggest that the AOT40 exceeds the critical levels recommended by UNECE (2010) for the forests protection (i.e. 5000 ppb*h) over large parts of Europe (Figure 4). In contrast, PODY-based critical levels have been established only for a few species, thus we cannot clearly discriminate all the
areas subjected to potential O\textsubscript{3} risks. However, for POD1 Mills et al. (2011b) identified a critical level of 4 mmol m\textsuperscript{-2} for birch and beech, and a value of 8 mmol m\textsuperscript{-2} for Norway spruce. Considering these thresholds, our results suggest that most of the European forests are potentially exposed to O\textsubscript{3} risks, while the damages seem to be less relevant in Eastern Europe and some areas over South-Eastern Spain (Figure 9, upper panels).

When assessing the O\textsubscript{3} risks by comparing AOT40 and POD0 (Figure 4 and 5), we found remarkable inconsistencies in both temporal and spatial pattern. Specifically, the use of the exposure-based AOT40 drastically changes O\textsubscript{3} risk assessments for vegetation relative to the flux-based PODY (Figure 4 and 5), while no relevant spatial and temporal differences occur when using a threshold-based POD1 metric rather than a no-threshold POD0 metric (Figure 5 and 9). Since both AOT40 and PODY metrics aim to do the same thing, namely point out the risk of ozone-damage to vegetation, the clear differences between them are not easily reconciled (Simpson et al., 2007). Thus a clear standard for the near future must be adopted to protect vegetation against O\textsubscript{3}.

Threshold-based indices assume that plants have adapted to low, pre-industrial, naturally occurring O\textsubscript{3} concentrations, and postulate that O\textsubscript{3} in the mesophyll is detoxified without inducing injury below a defined threshold (Mills et al., 2011a). Therefore, one of the disadvantages of PODY indices is that the thresholds lead to a significant sensitivity to uncertainties in the input flux calculations (Tuovinen et al., 2007). In contrast, it can be argued that any O\textsubscript{3} molecule entering a leaf has the potential to induce a metabolic reaction in the plant (Musselmann et al., 2006); thus, the use thresholds implies that part of O\textsubscript{3} molecules entering into the plant is not phytotoxic thanks to plant detoxification processes activated by the oxidative stress. However, these processes are not yet completely quantified and, consequently, modelled as function of diurnal and seasonal cycles.

Previous modelling approaches to estimate O\textsubscript{3} risk to vegetation (e.g. Tuovinen et al., 2007; Mills et al., 2011b; de Andrés et al., 2012) also showed significant differences in the spatial distribution of AOT40
and PODY, while a spatial and temporal comparison with different Y thresholds was not yet undertaken. Some studies have shown AOT40 levels well above the threshold for forest protection in South-eastern France, in rural alpine areas characterised by a Mediterranean climate, and in the Western Mediterranean basin (Dalstein & Vas, 2005; Sicard et al., 2011, 2013). Also PODY levels are usually higher in Southern Europe than in Nordic countries, although the increase is lower than that for AOT40 (Mills et al., 2011b).

In Southern Europe it is well known that the amount of water available in the soil is the main limiting factor to O₃ uptake (Paoletti, 2006; González-Fernández et al., 2013). However, since large-scale data of soil water content are difficult to retrieve, in most of previous risk assessments the water stress limitation was often neglected (Mills et al., 2011b). This approach represents a simplified flux-based “worst-case” risk assessment method for use in large-scale and integrated assessment modeling (Simpson et al., 2007; Tuovinen; Mills et al., 2011b). Our results indicate that taking into account the soil water function the risk based on POD1 considerably decreases.

The main limitation of AOT40 index is that it does not consider any environmental stress to vegetation, and the risk is based on the air O₃ concentration only. Conversely, limitations in the PODY risk assessment are due to the forest cover data and the parameterization defined for all tree species. Specifically, one of the main parameter controlling the stomatal conductance to which is associated a large uncertainty is the maximum stomatal conductance (gₘₐₓ). Körner (1995) reported that gₘₐₓ does not differ among major forest types, while Reich et al. (1999) suggest that there is no significant difference in gₘₐₓ between evergreen and deciduous trees, although long-lived leaves may have a lower gₘₐₓ. However, Mediterranean trees are known to be less O₃-sensitive than Northern European provenances even without water limitation (e.g. Rinallo & Gellini 1989; Bauer et al., 1997; Paludan-Muller et al., 1999). This suggests that there is still an open discussion on the real meaning of maximum stomatal conductance. The maximum stomatal conductance (gₘₐₓ) is based on the average above the 90th percentile (González-Fernández et al., 2010) or the 98th percentile (Hoshika et al., 2012) of gsto measurements under optimum
environmental conditions for stomatal opening. Variations in the $g_{\text{max}}$ values are therefore due to the different site-specific optimal conditions. However, we believe that $g_{\text{max}}$ for a given species should be assessed by elimination of all environmental constraints, including the soil matrix, which is obtained directly placing the plant in a hydroponic system or in a well-watered environment under optimal conditions of temperature, moisture, light and nutrients. In this case, we should have the real species-specific $g_{\text{max}}$ and then the different limiting functions allow computing the final stomatal conductance.

One of the aims of this work was to propose a revised version of the original formulations of AOT40 and PODY. Indeed, with the original formulation the AOT40 is computed over growing season only (i.e. 1st April-30th September) for both deciduous and evergreen trees, while the PODY uses a latitudinal and altitudinal model to estimate the beginning of growing season (Klingberg et al., 2014). In contrast, we suggested computing the indices year-round, even for deciduous trees, using the stomatal conductance to assess if the environmental conditions are favorable to start the growing season. In fact, this approach allows to consider changes in green-up or dormancy dates related to a changing climate (Menzel et al., 2006; Schwartz et al., 2006; Piao et al., 2007; Wolkovich et al., 2012; Fu et al., 2014; Piao et al., 2015; Wang et al., 2015). In case of AOT40, we found that the percentage of variation between the two methods is small over deciduous forests (<5%), but the possible O$_3$ risk increases up to 50% for some evergreen forests (Figure 10). Conversely, due to the non-linearity between O$_3$ concentration and stomatal conductance, the percentage of change between the PODY computed year-round or during the growing season only (i.e. 1st April-30th September) is quite large even for deciduous forests (~20%). Also the temperature may explain this large difference: in particular, the percentage of change is larger in the southern bound of our domain, where the temperature is typically higher than in the continental Europe.

However, one could argue that our revised algorithms might be not completely correct: for instance, during one given day in winter if all the environmental conditions are favorable, we expect to get a stomatal conductance slightly greater than 0, while we know that the growing season starts when
environmental conditions are favorable for many consecutive days. However, given the relative low percentage of changes (Figure 10), we believe this approximation does not lead to a remarkable change in the magnitude of the indices. Recently, a new approach to assess the influence of climate change on the length of the time period during the year when coniferous and evergreen forest trees are sensitive to O\textsubscript{3} has been proposed (Klinberg et al., 2014); however, to avoid complications associated with day length dependence on budburst, the authors limited their analysis to evergreen tree species. We believe that our methods, even if with its limitations, would be more appropriate, since it allows considering many different tree species. In addition, since we take into account different bioclimates, the parameterizations used for each tree species over different bioclimatic regions would minimize this effect on the canopy conductance, while an overall parameterization for evergreen or deciduous trees would sharpen this problem. We would finally point out that our results and findings have relevant implications, and the following conclusions can be drawn:

- Considering the AOT40, the increase in model resolution does not necessarily lead to better accuracy of risk assessment, as demonstrated by the similarity between our AOT40 and EMEP results. Indeed, a finer resolution allows a better representation of processes over mountainous regions or coastal areas. Conversely, an increase of resolution is expected to lead to a better PODY quantification because of a better representation of different forest trees.

- Generally, there is not a relevant inter-annual variability in the analyzed indices: specifically, during all the years we identified always the same areas as most exposed to potential O\textsubscript{3}-induced injuries. However, a revision of the definition of "length of growing season" is needed to better quantify the risk under a changing climate (Klinberg et al., 2014). For deciduous forests there is
no a dramatic change in the risk, but for evergreen or coniferous forests the risk might be significantly underestimated if considering only the original formulation (i.e. 1st April-30th September).

- AOT40 and PODY clearly show both spatial and temporal inconsistencies; therefore a definition of a clear European standard is needed. While the AOT40 is critical since the risk only depends on atmospheric O$_3$ concentrations, in case of PODY there is large uncertainty depending on the land cover map as well as the parameters used for each vegetation category.

- In light of inconsistencies between AOT40 and PODY, as well as the strong agreement in the spatial pattern between our stomatal conductance model and GPP observation-based data, we believe that using the AOT40 (even with our revisions) just because it has the advantage of being simple is no more justified. An approach based on the amount of O$_3$ entering the leaves would be therefore more appropriate for risk assessments.

- Considering (i) that detoxification processes cannot be represented by a constant threshold value during the day (ii) the well described similarity in the spatial and temporal patterns between POD0 and POD1 (iii) the lack of information on the amount of O$_3$ in the mesophyll that is detoxified without inducing any injury, we suggest using the POD0 for integrated risk assessments.

- As most of numerical models, the methodology presented here to estimate the stomatal conductance is a simplification of the real world. We believe a process-based model with a Ball-Berry parameterization (Ball et al., 1987; Leuning, 1995) would be more appropriate to assess land surface-atmospheric chemistry feedbacks (Anav et al., 2012). Specifically, with this
methodology we are only able to produce risk assessment maps, while it is well known that the ozone-induced damages to vegetation lead to significant changes in vegetation composition and structure that, in turn, affect the planetary boundary layer (Anav et al., 2012). Thus, what is really challenging for the near future, in addition to the definition of a clear standard, is the use of online models that allow accounting for the different feedbacks between land surfaces and atmospheric chemistry and physics.

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Table 1. Species-specific parameterizations used to estimate the stomatal conductance (as in UNECE, 2010).

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FIGURE CAPTIONS

Figure 1. Yearly bias (ppb) obtained comparing the mean daily ozone concentration as simulated by CHIMERE with observations from AIRBASE and EMEP networks.

Figure 2. Yearly correlation coefficient obtained comparing the mean daily ozone concentration as simulated by CHIMERE with observations from AIRBASE and EMEP networks.

Figure 3. Daily variability of observed (gray) and simulated (green) ozone concentrations (in ppb) averaged over all the AIRBASE and EMEP stations. Shaded area represents the standard deviation computed considering all the stations.

Figure 4. Spatial distribution of AOT40 (in ppb*h) over different years, computed according to the revised formulation (Equation 1).

Figure 5. Spatial distribution of POD0 (in mmol m^{-2}) over different years.

Figure 6. Comparison between MTE-GPP (in gC m^{-2} year^{-1}, upper panels) and stomatal conductance (Equation 2 [mmol m^{-2} s^{-1}], lower panels).

Figure 7. Comparison of mean annual ozone concentration as simulated by CHIMERE with ozone concentration from EMEP model (in ppb).

Figure 8. Comparison of AOT40 (in ppb*h) over different years as estimated by our framework (upper panels) and EMEP (lower panels). Squares in the upper panels represent the AOT40 computed using station data.

Figure 9. Comparison of POD1 over different years as estimated by our framework taking into account the water limitation on stomatal conductance (upper panels), without the water stress (central panels), and EMEP (lower panels).

Figure 10. Percentage of change between AOT40 (upper panels) and POD0 (lower panels) computed year-round and only during the growing season.
Figure 1.
Figure 2.
Figure 3.
Figure 4.
Figure 5.
Figure 6.
Figure 7.
Figure 8.
Figure 9.
Figure 10.
Performances of atmospheric model

Figure S3 shows the bias of WRF simulated climate constrains of Equation 2 (i.e. 2-meter air temperature, 2-meter relative humidity, downward shortwave radiation and soil moisture) with respect to ERA-INTERIM reanalysis used as boundary conditions for the WRF simulations. Considering the 2-meter temperature, we found WRF to be colder than its boundary conditions over all the mountain regions; specifically over the Carpathian, Alps, Apennines, Balkans, Pyrenees, Atlas, Pindos and Taurus mountains WRF has a systematic negative bias above 3 °C. However this result is somewhat expected because in regions characterized by complex terrain the downscaling better reflects the temperature changes related to the increasing altitude. In fact, as already discussed by Giorgi & Bates (1989) when a high resolution model boundary layer formulation is used, large biases may not indicate that the model results strongly deviate from reality, but rather that the simulated boundary layer physics is more relevant than the lateral boundary conditions in determining the solution. Indeed, due to the higher spatial resolution, the model produces fields that may be locally more physically realistic than the coarser reference data (Giorgi & Bates, 1989).

Besides, WRF shows a slight positive bias over the Eastern part of the domain, while over all the Atlantic regions of France, UK, Portugal and Scandinavian Peninsula the bias is slightly negative.

The bias in 2-meter relative humidity shows a more homogeneous spatial pattern, with ERA-INTERIM being wetter than WRF almost everywhere, except Alps and UK. The same consideration is also valid for PAR: in fact WRF systematically simulates lower radiation than ERA-INTERIM.
Considering the soil moisture, WRF is drier than its forcing over all the domain, except coastal areas. As already discussed for temperature, this pattern is likely related to the downscaling which enhances the model-data misfit in area of complex topography or over specific surface cover. In addition, the different land-sea cover fraction of coastal grid points between WRF and ERA-INTERIM might explain the systematic bias over these areas.

In order to have a visual comparison of model skills in reproducing the mean annual cycle of the selected variables over the different bioclimatic regions (Figure S1), we use the Taylor diagram (Taylor, 2001). The WRF performances are expressed in terms of their correlations, centered RMS differences and standard deviations compared to its forcing (i.e. ERA-INTERIM). Figure S4 clearly highlights how WRF systematically better reproduces 2-m temperature and radiation than soil moisture and 2-m relative humidity. Since seasonal variation in temperature and radiation are primarily determined by the insolation pattern, while seasonal variations in soil moisture and humidity are strongly influenced by vertical movement of air due to atmospheric instabilities of various kinds and by the flow of air over orographic features, models in general have higher skills in reproducing the right temperature pattern than precipitation. In fact, to simulate accurately the seasonally varying pattern of precipitation, models must correctly simulate a number of processes (e.g., evapotranspiration, condensation, and transport, Randall et al., 2007). Therefore, the correlation of WRF-simulated 2-m temperature and radiation is systematically larger than 0.95, while soil moisture and 2-m humidity are between 0.7 and 0.8 with the exception of Alpine bioregion, where the performances are much poorer. The same consideration is also valid for the RMSD, with 2-m temperature and radiation having a lower error than soil moisture and 2-m humidity.
References for Supporting Information


Tables for Supporting Information

Table S1. WRF 3.6 physical configurations used in the model simulations.

<table>
<thead>
<tr>
<th>Process</th>
<th>Configuration</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Microphysics</td>
<td>Morrison 2-moment scheme (mp_physics = 10)*</td>
<td>Morrison et al. (2008)</td>
</tr>
<tr>
<td>Cumulus Parameterization</td>
<td>Grell-Freitas (cu_physics = 3)*</td>
<td>Grell and Freitas (2013)</td>
</tr>
<tr>
<td>Shortwave Radiation</td>
<td>RRTMG (ra_sw_physics = 4)*</td>
<td>Iacono et al. (2008)</td>
</tr>
<tr>
<td>Longwave Radiation</td>
<td>RRTMG (ra_lw_physics = 4)*</td>
<td>Iacono et al. (2008)</td>
</tr>
<tr>
<td>Land-surface</td>
<td>Noah land model (sf_surface_physics = 2)*</td>
<td>Ek et al. (2003)</td>
</tr>
<tr>
<td>Planetary Boundary Layer</td>
<td>YSU (bl_pbl_physics = 1)*</td>
<td>Hong et al. (2006)</td>
</tr>
</tbody>
</table>

*A complete description of parameterizations and model’s flags is given in the WRF 3 user guide ([http://www.mmm.ucar.edu/wrf/users/docs/arw_v3.pdf](http://www.mmm.ucar.edu/wrf/users/docs/arw_v3.pdf))
Table S2. Parameterization of wilting point (WP in volume/volume) and field capacity (FC in volume/volume) at different soil types for estimating the soil water content function $f_{SWC}$.

<table>
<thead>
<tr>
<th>Soil category</th>
<th>Soil type</th>
<th>Wilting Point</th>
<th>Field Capacity</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Sand</td>
<td>0.010</td>
<td>0.339</td>
</tr>
<tr>
<td>2</td>
<td>Loamy Sand</td>
<td>0.028</td>
<td>0.421</td>
</tr>
<tr>
<td>3</td>
<td>Sandy Loam</td>
<td>0.047</td>
<td>0.434</td>
</tr>
<tr>
<td>4</td>
<td>Silt Loam</td>
<td>0.084</td>
<td>0.476</td>
</tr>
<tr>
<td>5</td>
<td>Silt</td>
<td>0.084</td>
<td>0.476</td>
</tr>
<tr>
<td>6</td>
<td>Loam</td>
<td>0.066</td>
<td>0.439</td>
</tr>
<tr>
<td>7</td>
<td>Sandy Clay Loam</td>
<td>0.067</td>
<td>0.404</td>
</tr>
<tr>
<td>8</td>
<td>Silty Clay Loam</td>
<td>0.120</td>
<td>0.464</td>
</tr>
<tr>
<td>9</td>
<td>Clay Loam</td>
<td>0.103</td>
<td>0.465</td>
</tr>
<tr>
<td>10</td>
<td>Sandy Clay</td>
<td>0.100</td>
<td>0.406</td>
</tr>
<tr>
<td>11</td>
<td>Silty Clay</td>
<td>0.126</td>
<td>0.468</td>
</tr>
<tr>
<td>12</td>
<td>Clay</td>
<td>0.138</td>
<td>0.468</td>
</tr>
<tr>
<td>13</td>
<td>Organic Material</td>
<td>0.066</td>
<td>0.439</td>
</tr>
<tr>
<td>14</td>
<td>Water</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>15</td>
<td>Bedrock</td>
<td>0.006</td>
<td>0.200</td>
</tr>
<tr>
<td>16</td>
<td>Other (land-ice)</td>
<td>0.028</td>
<td>0.421</td>
</tr>
</tbody>
</table>
Figure S1. Dominant soil categories (upper panel) and distribution of biogeographical regions based on a modified version of EEA dataset (http://www.eea.europa.eu/data-and-maps/data/biogeographical-regions-europe, lower panel). The black box into the upper panel indicates the CHIMERE domain.
Figure S2. Spatial distribution of trees based on EFI data (http://www.efi.int/portal/virtual_library/information_services/mapping_services/tree_species_map_for_european_forests/, upper panel) and converted vegetation according to the DO3SE species (lower panel).
Figure S3. Spatial pattern of annual anomalies (ERA-INTERIM - WRF) of 2-meter air temperature (°C), 2-meter relative humidity (%), downward shortwave radiation (W m\(^{-2}\)), and soil moisture (m\(^{-3}\) m\(^{-3}\)).
Figure S4. Taylor diagrams showing the WRF performances with respect to ERA-INTERIM mean annual 2-meter temperature (1, T2M), 2-meter relative humidity (2, RH), soil moisture (3, SM), and shortwave radiation (4, SW) over different biogeographical regions (see Figure S1).