Impacts of climate change on ground level gas-phase pollutants and aerosols in the Iberian Peninsula for the late XXI century

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ARTICLE INFO

ABSTRACT

Climate change alone influences future air pollution levels through modifications of gas-phase chemistry, transport, removal, and natural emissions. Hence, the goal of this study is to determine at what extent concentrations of air pollutants respond to changes over the Iberian Peninsula under a climate change scenario. The methodology includes the use of the regional modeling system MM5 (regional climate model version)-CHIMERE for two nested domains covering Europe and the Iberian Peninsula. Two time slices driven by ECHO-G global circulation model covering from 1991 to 2010 and 2071 to 2100 under the SRES A2 scenario have been compared. Climate change influences the concentrations of both gas-phase pollutants and aerosols through changes in temperature, precipitation, mixing height, transport, humidity, and oxidant levels. The trends of variation of ozone (changes up to 5 ppb, +10% increase during summertime) and aerosols over southwestern Europe are influenced by the higher mean temperature modeled for the future climate (up to +5.4 K), since it favors the formation of secondary gas-phase products. It also enhances sulphates ($+2\ \mu g\ m^{-3}$) and secondary organic aerosols (SOA) ($+2.5\ \mu g\ m^{-3}$ under SRES A2 scenario) and contributes to the decomposition of ammonium nitrate, remaining in the gas phase. Further, the 17% percent decrease of precipitation modeled for 2071–2100 has a strong effect in the frequency of the washout and therefore in the levels of natural aerosols: the concentrations of aerosols decrease with increasing precipitation as wet deposition provides the main aerosol sink.

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1. Introduction

Climate change impacts on air quality may influence long-term air quality planning. Climate change alone will influence future air quality through modifications of gas-phase chemistry (Racherla and Adams, 2008), transport (Jacob and Winner, 2009), removal (Avise et al., 2009; Dawson et al., 2009) and natural emissions (Zlatev and Moseholm, 2008; Liao et al., 2009). This influence is particularly important in the change of the intercontinental and transboundary transport and in hemispheric air pollution (Akimoto, 2003). However, several authors highlight the fact that the policies aimed at improving air quality have not accounted for the variations in the climate (Racherla and Adams, 2006; Steiner et al., 2006; Giorgi and Meleux, 2007; Shindell et al., 2008).

The coarse horizontal resolution of current global climate-chemistry simulations does not permit an estimate of the effects of climate change on tropospheric pollutants distributions on the regional scale (Forkel and Knoche, 2006). Therefore, the effects of climate change on air quality should be studied with higher resolution by using coupled regional chemistry transport models (Forster et al., 2007; Giorgi and Meleux, 2007; Gustafson and Leung, 2007).

Up to date, the most frequent approach adopted for the evaluation of climate change impacts on projected regional air quality has been the future-minus-present method. This is based on the assumption that biases in simulated present-day and future climate simulations should tend to cancel each other, and thus their difference captures the signal of the concentration anomalies. This method is widely supported in most future climate-air quality interactions studies (e.g. Forkel and Knoche, 2006; Liao et al., 2009; Chen et al., 2009; Pye et al., 2009), and therefore this approach is followed here. However, we should bear in mind that this approach is sensitive to the chosen control and future periods due to the inherent internal variability of the climate models, especially at regional scales (Raisanen et al., 2004). Moreover, the wide spread in the results of recent studies trying to analyze the influence of

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climate change on regional air pollution pointed out by Racherla and Adams (2008) and Wu et al. (2008), among others, indicate the inherent difficulty for these studies. Avise et al. (2009) highlight the need for including further works (especially in other regions apart from the United States) in order to assess the potential impact of global changes on regional air quality.

In this sense, the Mediterranean basin and the Iberian Peninsula are of special interest as climate change hot spots (Giorgi, 2006), with an important modification in the precipitation patterns and an increase in the desertification in lower latitudes. From an air quality perspective, the Western Mediterranean experiences frequent exceedances of the ozone and particulate matter thresholds which will be intensified under future scenarios (Meleux et al., 2007; Jiménez-Guerrero et al., 2009). Despite, to the author's knowledge, the scientific literature is scarce regarding air quality-climate simulations over the entire Iberian Peninsula and Spain, a number studies have evaluated the impact of climate change on air quality over Portugal. For instance, Carvalho et al. (2010) highlight that climate change alone may deeply impact the concentrations of pollutants under the SRES A2 scenario, and that emission changes should not be the only variable to take into consideration in this area. These authors have also analyzed the impacts of future forest fire emissions to the atmosphere for this same scenario (Carvalho et al., 2011).

Hence, this work complements and extends the previous knowledge for the entire Iberian Peninsula. The goal of this study is to determine the patterns of variability for ozone and aerosols under a changing climate (IPCC SRES A2 Scenario) for southwestern Europe at the end of the XXI century.

2. Models

The regional modeling system consists of a climatic version of the Fifth-Generation Pennsylvania State University − National Center for Atmospheric Research Mesoscale Model (MM5) (Dudhia, 1993; Grell et al., 1994). Different versions of the model have been extensively used in a number of regional climate simulations (e.g. Boo et al., 2006; Tagaris et al., 2007; Nunez et al., 2009; Lynn et al., 2010; Gómez-Navarro et al., 2010, among others), coupled to CHIMERE chemistry transport model (Schmidt et al., 2001; Bessagnet et al., 2004; Rouil et al., 2009). MELCHIOR2 gas-phase mechanism is implemented within CHIMERE (Derognat et al., 2003). The chemistry transport model includes aerosol and heterogeneous chemistry; distinguishes among different chemical aerosol components, namely nitrate, sulfate, ammonium, elemental and organic carbon with three subcomponents (primary, secondary anthropogenic and secondary biogenic) and marine aerosols. Unspecified primary anthropogenic aerosols and aerosol water are additionally kept as separate components. The model considers the thermodynamic equilibrium using the ISORROPIA model (Nenes et al., 1998). Last, the aerosol microphysical description for CHIMERE is based on a sectional aerosol module including 6 bins from 10 nm to 40 µm using a geometrical progression.

The spatial model configuration consists of two one-way nested domains of MM5−RCM simulations with spatial resolutions of 90 and 30 km, respectively. The inner domain covers the full Iberian Peninsula (Fig. 1). 24 sigma levels are considered in the vertical, with the top at 100 hPa. The fields are interpolated to CHIMERE working grids (resolutions of 0.75 and 0.2° for the European and peninsular domains, corresponding to approximately 80 and 25 km, in that order). The physico-chemical options for the regional modeling system (Table 1) have been chosen in order to minimize the computational cost, since none of the configurations included within the MM5−RCM provides the best performance for all seasons and locations (Fernández et al., 2007). A blending area of five grid points is excluded from the analysis hereafter.

The climate and air quality simulations cover two time slices, from 1991−2010 to 2071−2100, driven by ECHO-G General Circulation Model (Legutke and Voss, 1999) under the SRES A2 scenario. This scenario has been selected since it lies towards the upper end of the IPCC emission scenario range, with a CO_2 concentration reaching about 850 ppm by 2100, and hence maximized the effect of climate changes on air quality. A more detailed description of the ECHO-G experiment can be found in Zorita et al. (2005), meanwhile Gómez-Navarro et al. (2010) and Gómez-Navarro et al. (2011) provide information about the skill of the regional model for dynamic downscaling over the Iberian Peninsula. In the latter, the MM5−RCM model coupled to ECHO-G was validated in a reference period (1960−1990) against observational and reanalysis data in the context of a paleoclimate simulation. They showed that this system is able to realistically simulate the main regional features of the
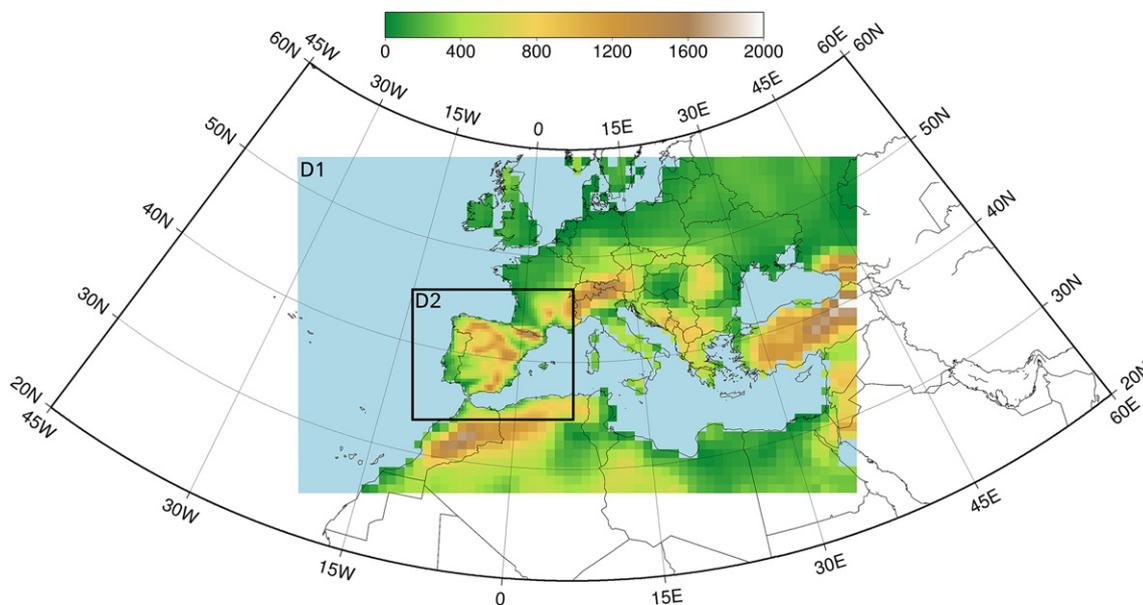

Fig. 1. Topography (m) of the domains included in the study as incorporated in the MM5−RCM model: Europe (D1) and Iberian Peninsula (D2).

Table 1
Configuration and parameterizations of the modelling system used.

ECHO-G	MM5	CHIMERE
Spectral atmospheric model ECHAM4 + HOPE-G ocean model; Atmos. resolution T30, 19 vertical levels; Ocean resolution 2.8°, grid refinement in the tropical regions, 20 vertical levels; Flux adjustment constant in time and with zero spatial average	Microphysics: Simple Ice; Cumulus: Grell; PBL: MRF; Radiation: RRTM; Soil: Noah LSM	Chemical Mechanism: MELCHIOR2; Aerosol chemistry: Inorganic (thermodynamic equilibrium with ISORROPIA module) and organic aerosol chemistry (MEGAN SOA); Natural aerosols: dust, resuspension and inert sea salt; BC: LMDz-INCA + GOCART

climate in the Iberian Peninsula, such as the temperature and precipitation annual cycle or the interaction between the large-scale circulation and orography, which modulates winter rainfall events over the target domain and has an important influence on air quality.

In the present work, boundary conditions for CHIMERE chemistry transport model are based on the global climate-chemistry model LMDz-INCA2 (96 × 72 grid cells, namely 3.75° × 2.5° in longitude and latitude, with 19 σ-p hybrid vertical levels, Szopa et al. (2009)) developed by the Laboratoire des Sciences du Climat et l'Environnement (LSCE). Climatic monthly mean data are interpolated in the horizontal and vertical dimensions to force the major chemical concentrations at the boundaries of the domain. A detailed description of the INteractive Chemistry and Aerosol (INCA) model is presented in Hauglustaine et al. (2004) and Folberth et al. (2006). Because the contribution of long-range transport on ground level concentrations (those considered in this work) can be considered as negligible, the influence of using climatological

boundary conditions is limited and overwhelmed by local processes. Changes on stratospheric ozone, which may impact ozone levels in the boundary layer (Isaksen et al., 2005) are not considered.

In order to isolate the possible effects of climate change on the ground concentrations of air pollution, unchanged anthropogenic emissions (derived from EMEP database) are assumed. Natural emissions depend on climate conditions, and consequently they are the only emissions to vary between reference and future climate simulations. Therefore, the effects of climate change on air pollutants are estimated without considering possible changes on vegetation, land use, anthropogenic pollutant emission changes or any feedbacks from the chemical compounds to the meteorological fields, but allowing changes in natural emissions (Meleux et al., 2007). Biogenic emissions were generated dynamically using MEGAN (Model of Emissions of Gases and Aerosols from Nature) (Guenther et al., 2006) with the parameterized form of the canopy environment model. The model estimates hourly isoprene,
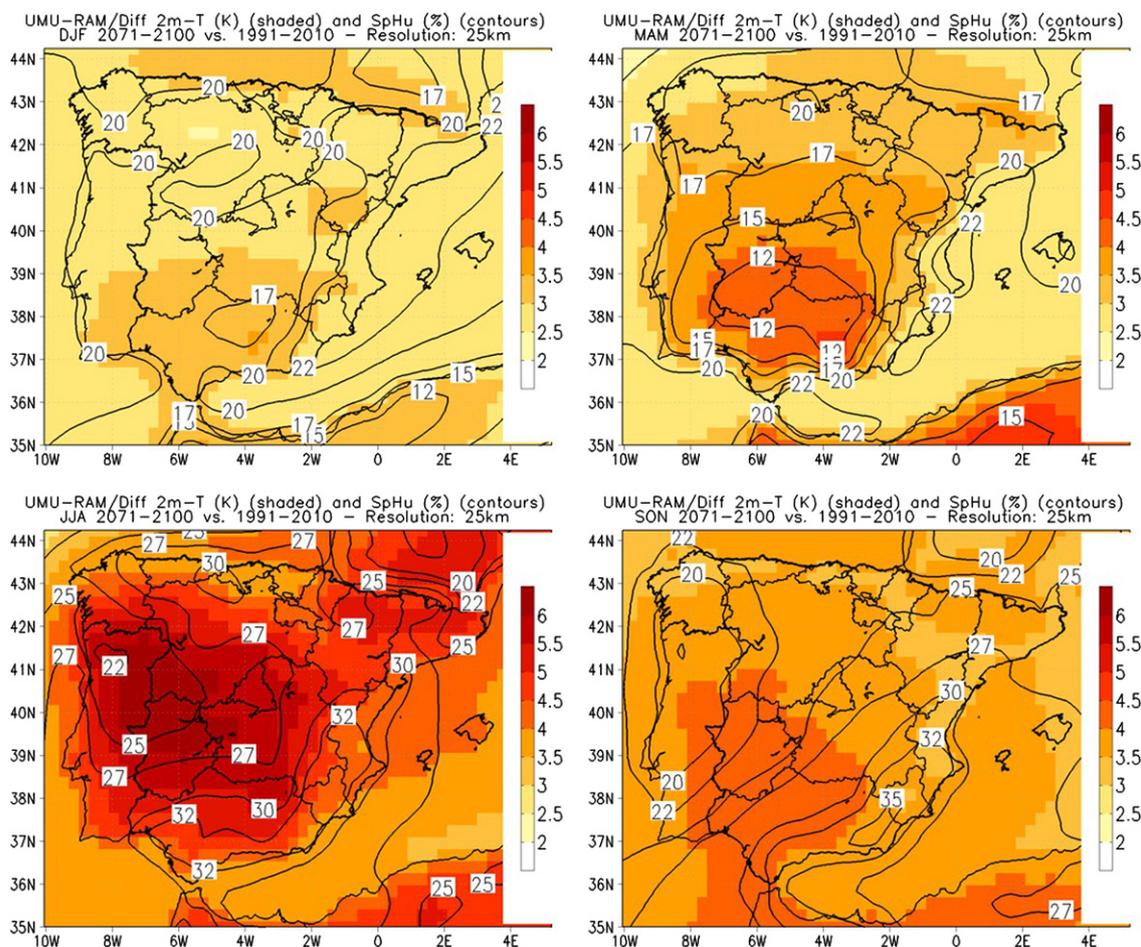
Fig. 2. Difference in seasonal temperature (K) (shaded) and specific humidity (%) (contours) in the Iberian Peninsula for the A2 scenario: 2071–2100 vs. 1991–2010 (DJF, top-left; MAM, top-right; JJA, bottom-left; SON, bottom-right).

monoterpene, and other BVOC emissions based on plant functional type and as a function of hourly temperature and ground level shortwave radiation from MM5–RCM.

3. Results

The performance of the modeling system when reproducing present-climate air quality and the corresponding definition of the aerosol climatology over the target domain has been presented elsewhere (Jiménez-Guerrero et al., 2009, 2011b). The main results of the aforementioned works show an accurate skill of the model and can be summarized as follows. The system presents a slight tendency to underestimate the total concentration of aerosols (PM10) when the model is evaluated against observations by Querol et al. (2009) (mean bias MB = -2.01 μg m^{-3}). Looking into the chemical composition, sulfate is better reproduced than other species; a large consistency between model and observations was found (mean fractional error MFE = 31.9%, mean error ME = 0.91 μg m^{-3}; root mean square error RMSE = 1.01 μg m^{-3}) because sulfate formation chemistry is well understood and the emissions of precursors are well characterized within emission inventories. Since ammonia is strongly correlated to the sulfate cycle in the CHIMERE simulations, the performance of the model is similar for this compound. Nitrate presents a relatively poor performance with strong underestimations (mean fractional bias MFB = -99.7%), in the order of magnitude of other chemistry transport models. The model performance for organic aerosols shows a slight under-prediction through the whole period (MFB = -11.12%; mean

normalized bias MNB = -5.50%) because of the limitations in the mechanisms for formation of secondary organic aerosols. Natural aerosols perform similarly (mean fractional error MFE and mean normalized error MNE around 50%). Also, the high spatial correlation between measurements and simulations (over 0.85 for all compounds) indicates an accurate reproducibility of the patterns of spatial variation within the Iberian Peninsula.

3.1. Changes on meteorological variables

Projected changes in seasonal 2-m temperature, 2-m specific humidity, accumulated precipitation and boundary layer height are shown in Figs. 2–4. Differences are computed as the 30-year 2071–2100 period in the future A2 simulation minus the present-day simulation (1991–2010).

Average 2-m temperatures (Fig. 2) are projected to increase over all the Iberian Peninsula; however, the magnitude of the increase varies greatly by region and season. The largest increases are observed during summertime in south-central Iberian Peninsula, above +5.4 K, meanwhile the coastal areas show increases around +2 K due to the larger thermal capacity of the oceans. The rest of the regions show an intermediate behavior. The warming pattern is similar for all seasons and varies just in magnitude. As will be shown later, these higher temperatures in the future scenario generally favor the formation of secondary pollutants, as ozone or organic aerosols and sulfates (increasing their concentrations) but fosters the decomposition of ammonium nitrate, remaining in the gas phase.
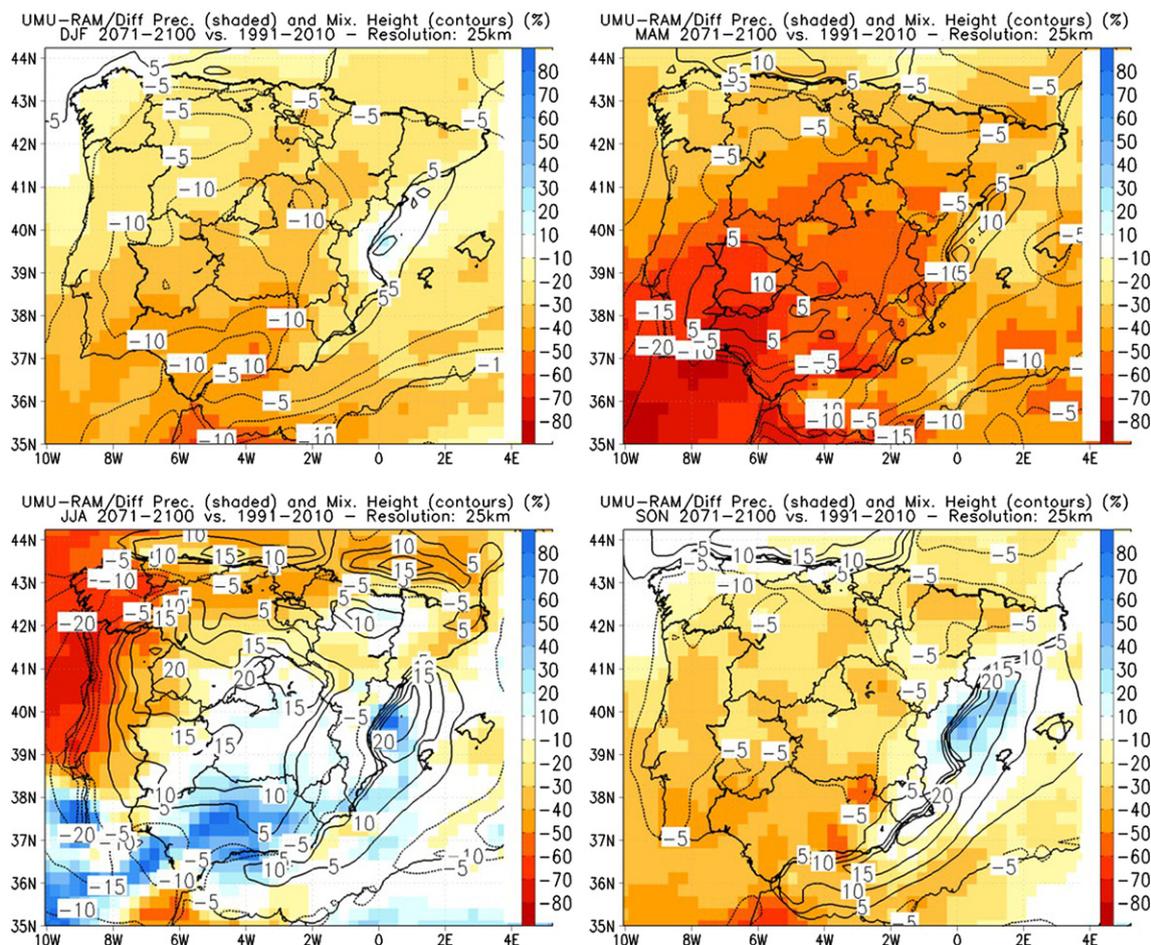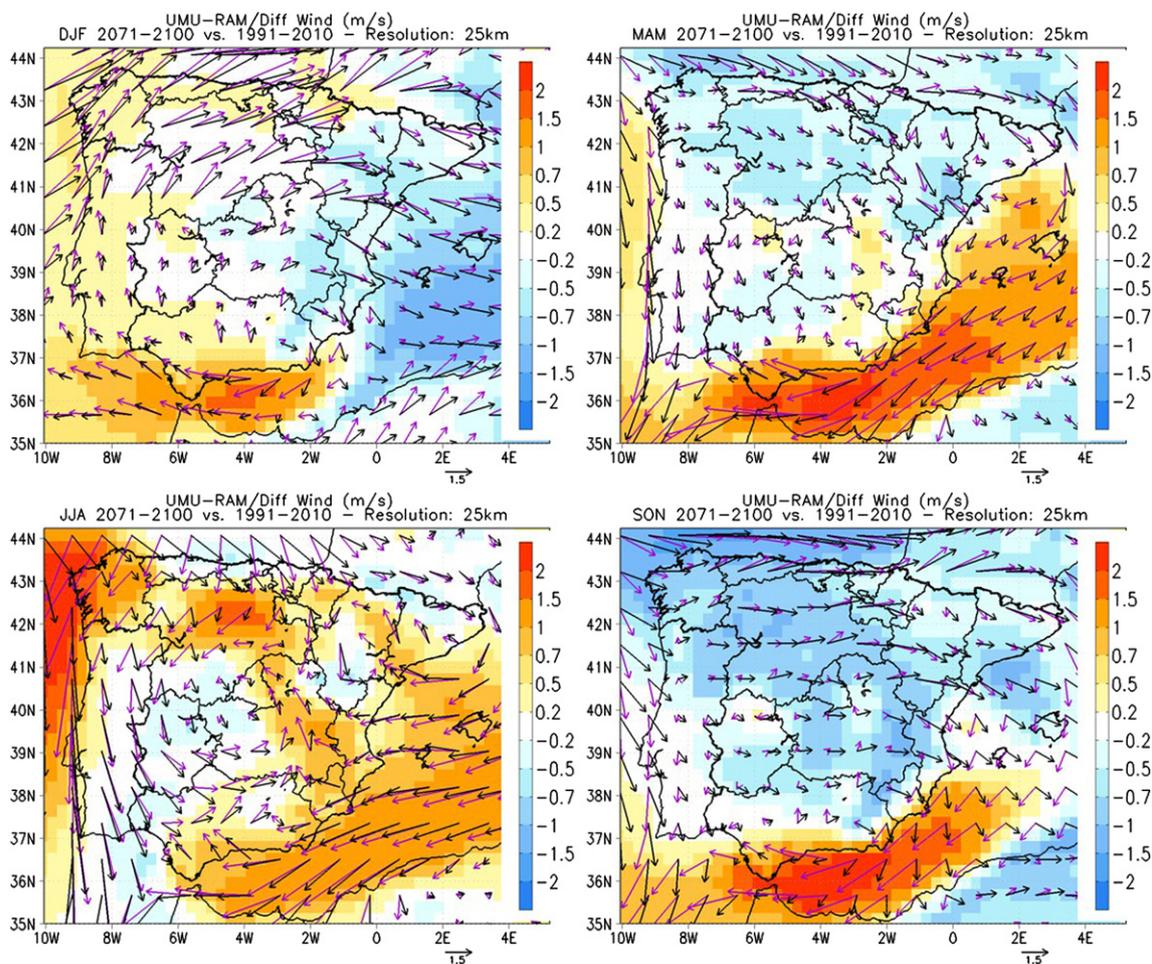

Fig. 3. Difference in seasonal precipitation (shaded) and mixing height (contours) (both in %) in the Iberian Peninsula for the A2 scenario: 2071–2100 vs. 1991–2010 (DJF, top-left; MAM, top-right; JJA, bottom-left; SON, bottom-right).

Fig. 4. Present-climate (black vectors) and future-climate (purple vectors) 10-m wind over the Iberian Peninsula; and difference in 10-m wind module (shaded) (all variables in m s−1) in the Iberian Peninsula for the A2 scenario: 2071-2100 vs. 1991-2010 (DJF, top-left; MAM, top-right; JJA, bottom-left; SON, bottom-right). (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this article.)

Further, the changes in precipitation (Fig. 3) have a strong effect on the frequency of the washout and therefore in the levels of aerosols. The concentrations of aerosols increases with decreasing precipitation as wet deposition provides the main aerosol sink. For the Iberian Peninsula, the precipitation is projected to decrease on average by 17%, with a strong seasonality depending on the area. In this sense, precipitation over the eastern Iberian Peninsula is simulated to decrease over 50% for MAM and JJA. Conversely, the Mediterranean coast is projected to increase its precipitation by 23% on average during JJA, but we should bear in mind that the precipitation over southern Spain and eastern Iberian Peninsula is scarce on summertime, so these increases represent less than 0.1 mm day^{-1} on average.

The changes in ventilation, and specifically, in the mixing height have strong effects on aerosols because of their low background concentrations. Strong correlation of particulate matter with stagnant conditions is expected and has been previously reported by Leung and Gustafson (2005). The mixing height (MH) over the Iberian Peninsula is predicted to generally decrease from the present day to 2071−2100 (Fig. 3). Conversely, there is a particularly strong increase in the MH of about 30% over the Mediterranean coast during JJA and SON which may be associated with drying and warming (Wu et al., 2008). Increases and decreases in the MH are generally less than 20%. Note that decreases in the MH (hampering dispersion processes) are generally associated with a decreasing precipitation and increases in temperatures. In those

other regions changes in both meteorological parameters will generally lead to a positive relation in the change signal. The trend from a decreasing (increasing) mixing height found in several areas of the Iberian Peninsula is associated with a decrease (increase) in precipitation, and therefore represents an adding effect.

Also, the change in the circulation patterns (Fig. 4) affects the wind behavior, which strongly conditions the levels of natural aerosols, as will be discussed later. For winter, changes over the Iberian Peninsula indicate a decrease of zonal winds related to a latitudinal increase of sea-level pressure (Jerez, 2011). An intensification of the easterly winds appear, basically imposed by the boundary conditions driving the simulations. The projected changes during summer also point to an intensification of the easterly winds in both simulations. This is in agreement with the idea proposed by Haarsma et al. (2009), that the drier Mediterranean soils intensify easterly winds. Also, the simulations for JJA show an enhancement of the Iberian Thermal Low (Hoinka and Castro, 2003), producing a stronger cyclonic circulation around the Iberian Peninsula; the development of this thermal circulation is associated with the drier soils simulated for the future scenario, that will strongly affect the levels of natural resuspension of aerosols (Vautard et al., 2005).

3.2. Gas-phase pollutants

For ground level gas-phase pollutants (Fig. 5, top), the general increase in daily averaged ozone concentrations found under A2

Fig. 5. (Top) Projected changes in seasonal ground level gas-phase pollutants (ppb) in the Iberian Peninsula for the A2 scenario: 2071–2100 vs. 1991–2010. Shaded colors represent ozone changes and contours stand for nitrogen dioxide (DJF, top-left; MAM, top-right; JJA, bottom-left; SON, bottom-right). (Bottom) Id. expressed in relative changes with respect to 1991–2010 concentrations.

forcing (up to 5 ppb for central Iberian Peninsula during summertime) is due to a multiplicity of climate factors, such as increased temperature, decreased wet removal associated with the general decrease of precipitation, increased photolysis of primary and secondary pollutants due to lower cloudiness and increased biogenic emissions due to higher temperatures (as also stated by Giorgi and Meleux (2007)). This leads to a relative increase of 10% in mean ozone concentrations during JJA in the northwestern plateau of the Iberian Peninsula (Fig. 5, bottom), which is limited to 7.5% in the rest of the parts of the Iberian Peninsula and the rest of the seasons.

Moreover, the specific humidity is projected to largely increase in all the area under study (by approx. 30%, Fig. 2). According to Avise et al. (2009), these increases in relatively NO_x polluted environments (as occurring in most of the Iberian Peninsula), increased water vapor is expected to increase ozone through the competing reaction $NO + HO_2 \rightarrow NO_2 + OH$ (Stevenson et al., 2000; Racherla and Adams, 2008).

As observed from the NO_2 variation under the A2 scenario (Fig. 5, contours), the future combination of less dispersive conditions and lower removal efficiency (increased sea-level pressure and subsidence) leads to increased concentrations of pollutants in the boundary layer, and in particular an increase in NO_2 availability especially over the northwestern Iberian Peninsula (up to 60% more in 2071–2100 than in 1991–2010 period during summertime) for ozone production. Moreover, the variations (general decreases) in precipitation for the future scenario can strongly influence the levels of NO_2, since the wet deposition of nitric acid (HNO_3) is the main sink for NO_x. In the existence of sunlight, nitrogen dioxide reacts

mainly with hydroxyl radical to produce HNO_3, while nighttime chemistry involves the formation of N_2O_5, the anhydride of HNO_3, whose dry and wet deposition can be an important nighttime loss process for nitrogen oxides (Atkinson, 2000; Jacob, 2000).

Despite this large relative increase in NO_2, its changes are limited to 1.2 ppb as mean variation (3.6 ppb as maximum summertime increases) because low levels of nitrogen dioxide are simulated over the northwestern Iberian Peninsula for the period 1991–2000. Therefore our simulations suggests that the enhanced ozone chemical activity is mainly driven by the warming-induced increase in biogenic emissions (vegetation is kept invariable in the simulations, but MEGAN estimations of these biogenic emissions strongly depends on shortwave radiation and temperature, which are substantially modified in future climate scenarios) and not because of the variation in nitrogen dioxide levels, which hardly change or even decrease in the central and southeastern Iberian Peninsula (decreases up to 2.5 ppb, relative changes under 5–10%). Meleux et al. (2007) indicate that the high biogenic reactivity sustains the ozone production cycle and increases the O_3 yield per NO_x molecule. In most of the Iberian Peninsula, the ozone production is VOCs dependent (Jiménez and Baldasano, 2004). Therefore, in these regions of the domain an increase in nitrogen oxides concentrations does not imply an increase in ozone production, as depicted in the simulations.

3.3. Aerosols

Observed correlations of aerosol concentrations with meteorological variables are weaker than for ozone and other gas-phase
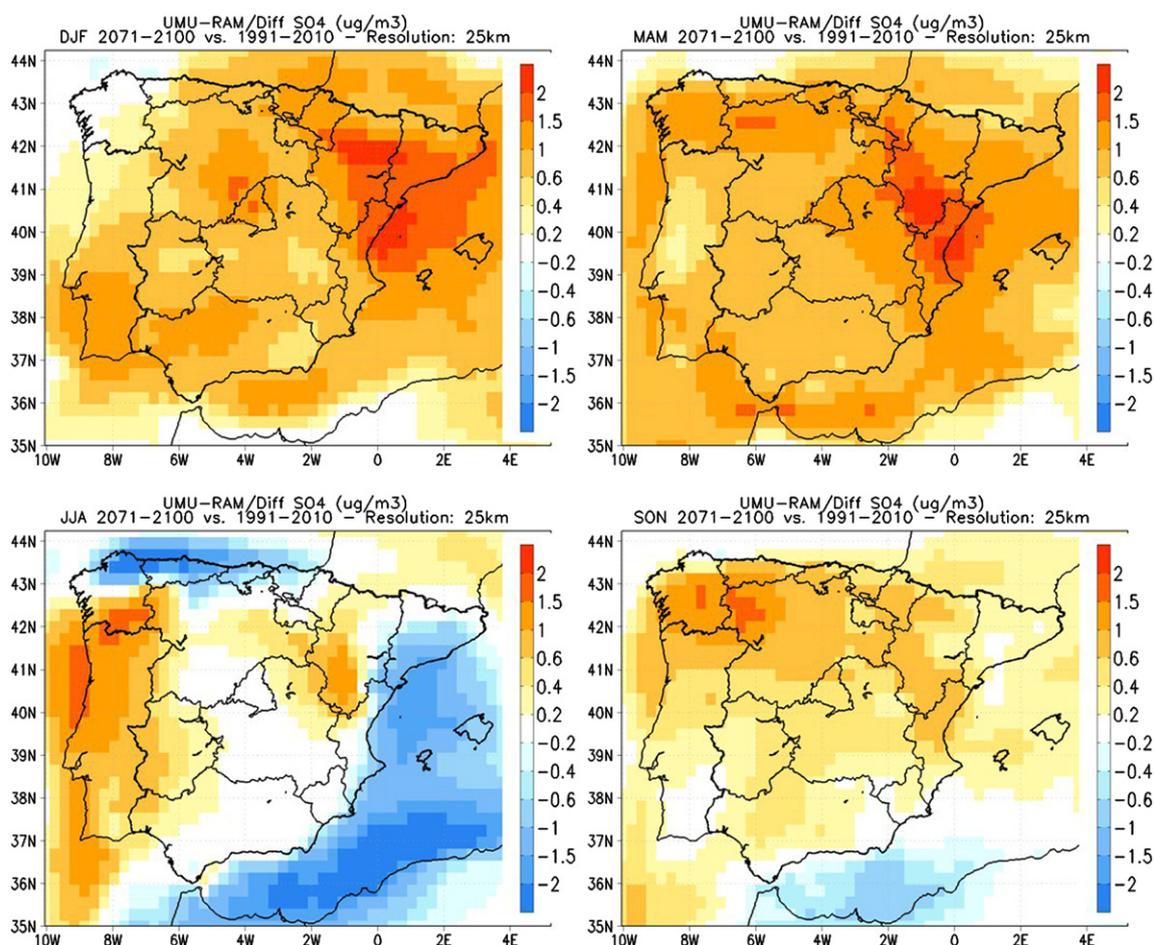

Fig. 6. Projected changes in ground level seasonal sulfate ($\mu g\ m^{-3}$) in the Iberian Peninsula for the A2 scenario: 2071–2100 vs. 1991–2010 (DJF, top-left; MAM, top-right; JJA, bottom-left; SON, bottom-right).

pollutants. This reflects the diversity of aerosol components, the complex coupling of aerosols to the hydrological cycle, and various compensating effects (Jacob and Winner, 2009) as those discussed above. Model perturbation studies find that the effect of temperature on PM depends on the PM component (Dawson et al., 2009). Hence, a careful look has to be taken when analyzing the chemical composition of ground level aerosols over the Iberian Peninsula.

3.3.1. Secondary inorganic aerosols (sulfate–nitrate–ammonium)

On an annual basis, sulfate concentrations are predicted to increase mainly inland the Iberian Peninsula by up to 2 µg m^{-3} whereas decreases over the northern and southern coasts by up to 0.5 µg m^{-3}. As stated by Tagaris et al. (2007), these increases because of climate change alone are strongly related to the changes of sulfur dioxide, which increases by 20% in the largest emitting areas of the Iberian Peninsula. Rae et al. (2007) have shown that increases in temperature and changes in oxidant concentrations are simulated to decrease 1% of Aitken-mode sulfate aerosols but increase of 9.2% of accumulation-mode sulfate by the end of the XXI century, assuming climate and emission-induced oxidant levels will follow the IPCC SRES A2 scenario.

For analyzing the future seasonal changes of sulfate (Fig. 6), the critical variable is precipitation frequency (since scavenging within a precipitating column is highly efficient (Jacob and Winner, 2009)) rather than temperature. If we take a look at the spatial patterns of sulfate and 2-m temperature variation,

the former seems to be somehow impacted by the higher temperatures modeled for the future climate. This could be attributed to the fact that temperature does not directly impact the concentrations of sulfate and nitrate, but modifying the kinetics and the concentrations of atmospheric oxidants (Jiménez-Guerrero et al., 2011a). As reported by some authors (e.g. Dawson et al., 2007; Kleeman, 2007), temperature enhances the formation of sulfates due to a to faster SO_2 oxidation (higher rate constants and higher oxidant concentrations, increasing the formation of condensable compounds). Liao et al. (2006) and Unger et al. (2006) also point out that higher water vapor in the future climate leads to higher concentrations of H_2O_2, the principal SO_2 oxidant, thus increasing sulfate concentrations. Higher H_2O_2 levels are predicted to lead to more in-cloud H_2O_2 production of sulfate (Pye et al., 2009).

There is a generalized trend for a decrease of precipitation during all seasons over the Iberian Peninsula except in summer-time. Hence, sulfate levels at DJF, MAM and SON show a statistically significant increase ($p < 0.005$) by up to 2.3 µg m^{-3} in future climates mainly due to the reduction in precipitation, but also because of an enhanced photochemistry during these seasons, the lower air mass renovation at regional scale caused by an increase of the stagnant conditions, and the possible higher future contribution of marine secondary sulfate from DMS oxidation (Querol et al., 2009). For summer, 2071–2100 sulfate production in the Iberian Peninsula is somewhat offset as a result of the increased precipitation over the Mediterranean coast, the increment of the summer mixing layer depth hampering the mixing of
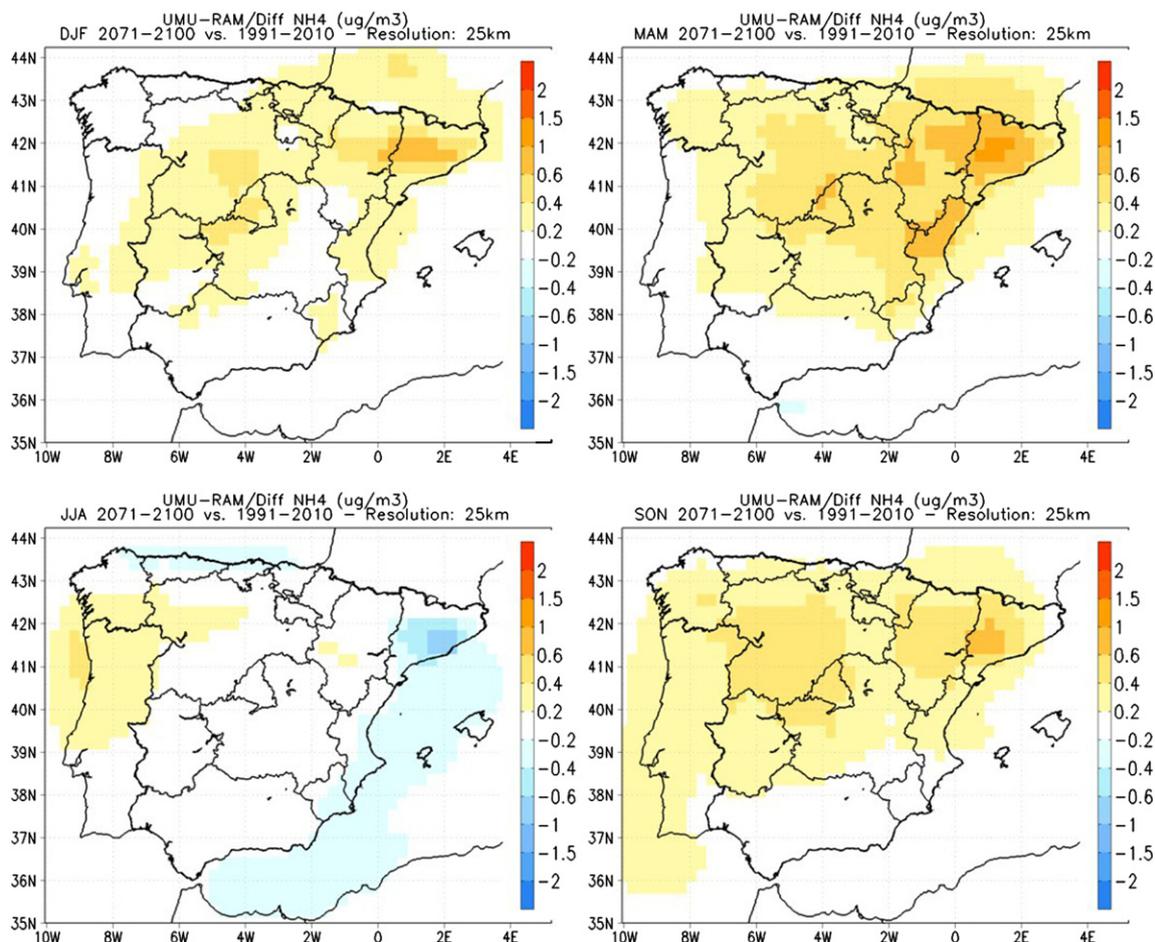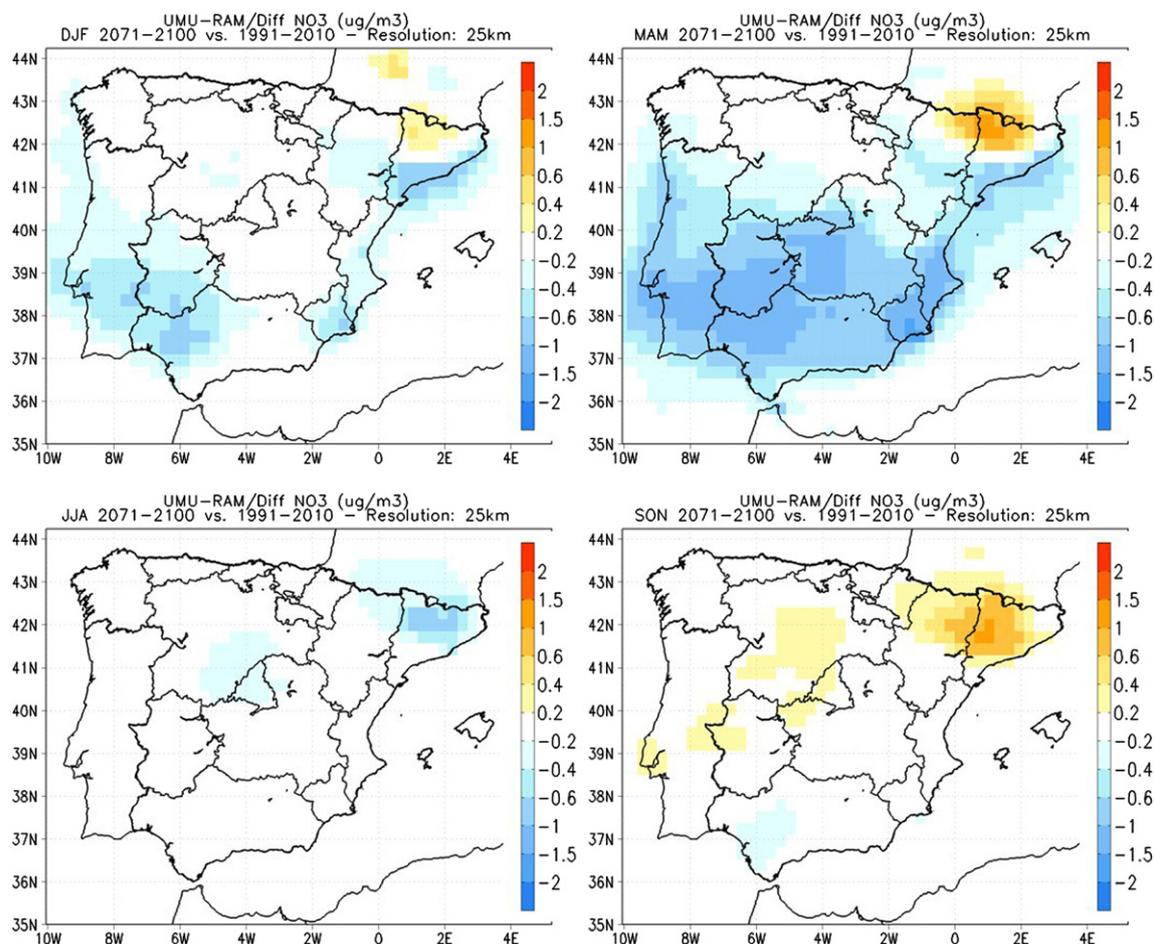

Fig. 7. Projected changes in ground level seasonal ammonium (µg m^{-3}) in the Iberian Peninsula for the A2 scenario: 2071–2100 vs. 1991–2010 (DJF, top-left; MAM, top-right; JJA, bottom-left; SON, bottom-right).

Fig. 8. Projected changes in ground level seasonal nitrate ($\mu g\,m^{-3}$) in the Iberian Peninsula for the A2 scenario: 2071–2100 vs. 1991–2010 (DJF, top-left; MAM, top-right; JJA, bottom-left; SON, bottom-right).

polluted air masses and the lower OH concentrations due to climate change. Although higher specific humidities are expected to increase HO_x production, higher temperatures and higher biogenic emissions can result in decreasing OH and increasing HO_2 (Pye et al., 2009).

The seasonally-averaged change in ammonium (Fig. 7) closely resembles that of sulfate. Most ammonium in the Iberian Peninsula would likely be in the form of ammonium sulfate and therefore 2071–2100 levels will increase up to 1.2 $\mu g\,m^{-3}$ in most seasons under the IPCC SRES A2 scenario, since more frequent stagnation leads generally to higher concentrations of sulfate and ammonium. The only important contribution to the formation of ammonium nitrate NH_4NO_3 in the simulated domain is observed in the northeastern Iberian Peninsula. For this compound, higher future temperatures favor NH_4NO_3 to remain in the gas phase and a decrease in ammonium levels is observed in the Spanish Mediterranean coast for summertime.

Because of higher temperatures, future nitrate aerosol is predicted to decrease across almost the entire Iberian Peninsula on an annual basis, with a maximum decrease of 0.64 $\mu g\,m^{-3}$ in southern Spain (Sevilla and the industrial area of Escombreras, Murcia). Aw and Kleeman (2003) present that nitrate aerosol may slightly increase with cool temperature (<290 K) but decrease with hot temperature (>290 K) as temperature increases. However, total nitrate can increase or decrease in individual seasons (Fig. 8), since the effects of climate on nitrate are more complicated than sulfate due to high vapor pressure for particle-phase ammonium nitrate (Seinfeld and Pandis, 2006). During spring and autumn, higher absolute humidity in the Catalan Pyrenees can favor nitrate partitioning to the aerosol phase (Dawson et al., 2007) and therefore nitrate concentration increases; however this increase is not statistically significant ($p > 0.1$). Despite the change signal of nitrate is not so obvious in the Iberian Peninsula (as also found by Jiménez-Guerrero et al. (2011a) for Europe), nitrate concentrations tend to decrease ($p < 0.05$) in most of the domain during springtime because of the higher temperature predicted for the 2071–2100 (Pye et al., 2009). Dawson et al. (2007) indicate that this reduction likely indicates the effects of increased temperature on thermodynamic interactions between sulfate and nitrate (changing the temperature-dependent oxidation chemistry of SO_2, the amount of ammonia available, and the partitioning of semi-volatile ammonium nitrate). The reduced nitrate levels are fostered during summertime because of the increased wet deposition of gaseous HNO_3 in the Iberian Peninsula domain. Furthermore, the largest change in nitrate for the Iberian Peninsula is predicted to occur in a present-day ammonia-limited regime according to the gas ratio (<1), so increased 2071–2100 sulfate levels may result in less ammonia being available for nitrate (Pye et al., 2009).

3.3.2. Organic aerosols

The concentrations of primary organic aerosols (POA) changed little under the influence of future climates in the simulations included here (not shown). Conversely, average concentrations of secondary organic aerosols (SOA) increase considerably under the IPCC SRES A2 scenario (Fig. 9) and therefore the analysis in this
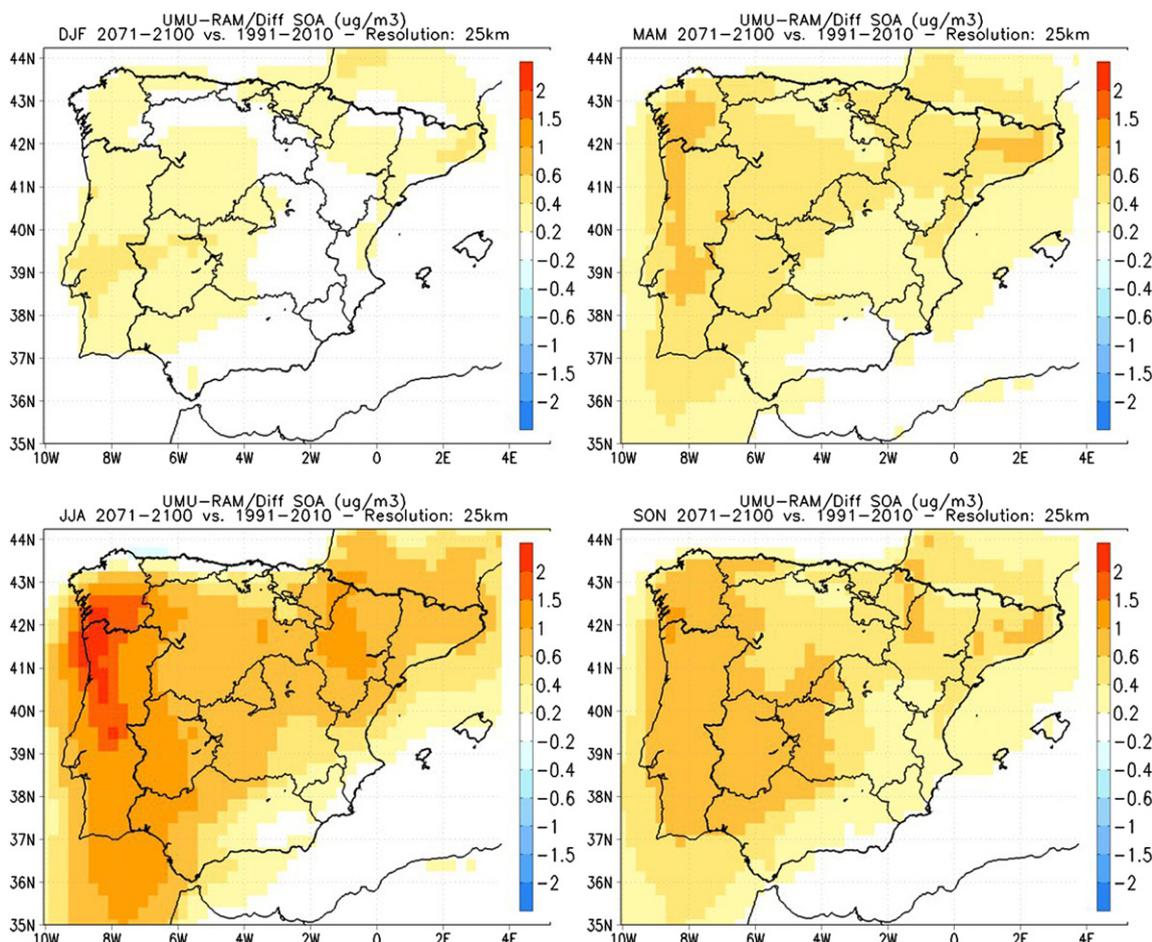
Fig. 9. Projected changes in ground level seasonal secondary organic aerosols (μg m^{-3}) in the Iberian Peninsula for the A2 scenario: 2071–2100 vs. 1991–2010 (DJF, top-left; MAM, top-right; JJA, bottom-left; SON, bottom-right).

section will focus mainly in the SOA and not in POA. As a first guess, higher temperatures modeled by MM5−RCM favor semi-volatile compounds such as SOA to remain in the gas phase. On the other hand, the modeled increases in temperatures and specific humidity (as previously observed in Fig. 2) result in higher emissions of SOA precursors and faster oxidation of volatile organic compounds (VOCs), increasing formation of condensable compounds, such as semi-volatile organic species (SVOCs) (Liao et al., 2009). The increase in SOA concentrations indicates that chemical production effects are outweighing volatility effects, as also stated by Dawson et al. (2009). SOA is predicted to be influenced by changes in biogenic VOCs emissions as well as modifications in the formation rates under the effects of the extreme IPCC SRES A2 climate scenario. Since SOA formation is conditioned by temperature-dependent oxidation chemistry, this again shows the importance of temperature changes. Moreover, the changes in the SOA formation (i.e. increases in the condensable compounds) are also facilitated by the decrease of precipitation, the main sink for these compounds.

3.3.3. Natural inert aerosols: sea salt + dust

The changes in natural inert aerosols (Fig. 10) are strongly related to those of sea salt and mineral dust matter. They are strongly correlated to the temperature increase (dust) and precipitation; however for sea salt the change in circulation patterns affect the local marine aerosol production. In this section, the contribution of dust to natural inert aerosols is only due to local resuspension and not to the transport from Saharan dust.

The source of sea salt emissions to the atmosphere is a function of 10-m wind to the 3.41 power, and this is calculated every time step in our CHIMERE simulations (for a more detailed description, see Jiménez-Guerrero et al., 2011c). As a result of this, the variation in sea salt aerosols is strongly correlated to the 10-m wind (≈0.8 for the whole Mediterranean domain) and hence shows a strong seasonal dependence as the wind does, as previously discussed. This change is only appreciable in the areas where the sea salt aerosols are emitted, over open-ocean areas (as observed in Fig. 10), while the changes inland are negligible. As reported by Vautard and Yiou (2009), the large-scale circulation controls the surface weather changes in Europe especially during the winter season, using wind as an accurate predictor. The variation of future circulation patterns load, on average, to differences up to 31% in some areas of the Mediterranean domain of southwestern Europe. These results agree to those of Mahowald et al. (2006), who indicate differences in zonally averaged concentrations of sea salt aerosols as large as 40% between the current climate a doubled carbon dioxide climate. However, these same authors indicate that globally averaged sea salt sources, deposition and loading are not very sensitive to climate change and change <5% for dispare climates and they largely depend on the response of the sea-salt flux scheme implemented in the model.

The contribution of local mineral aerosols is very high in the Iberian Peninsula due to the existence of semiarid soils all over our target domain. In the CHIMERE chemistry transport model, the resuspension flux is governed (apart from the type of soil,
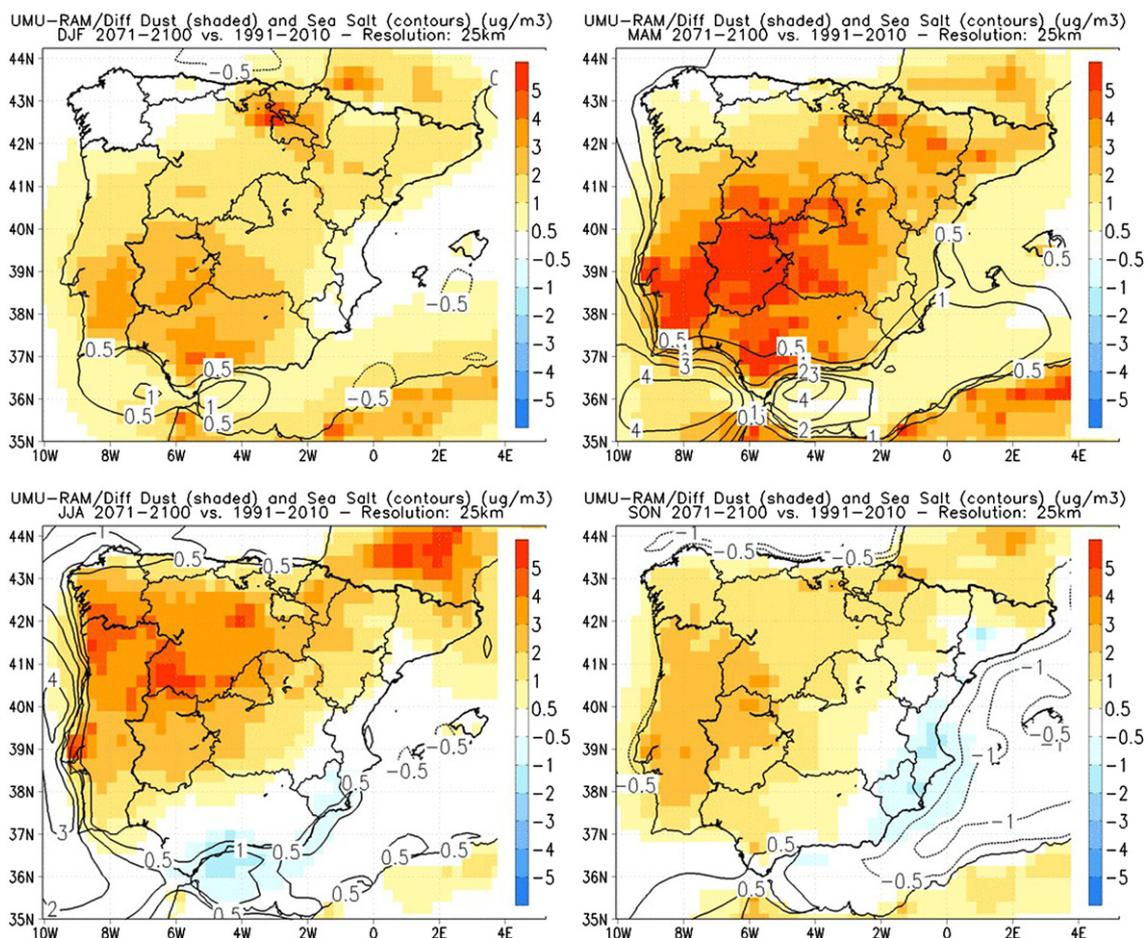
Fig. 10. Projected changes in ground level dust (shaded) and sea salt (contours) (μg m^{-3}) in the Iberian Peninsula for the A2 scenario: 2071—2100 vs. 1991—2010 (DJF, top-left; MAM, top-right; JJA, bottom-left; SON, bottom-right).

which is kept constant in the simulations) by the soil water content (Vautard et al., 2005) and hence the natural dust levels are strongly linked with precipitation changes. Future variations of local dust concentrations in the Iberian Peninsula may achieve up to 4 μg m^{-3} during springtime as a consequence of the enhanced fluxes of natural aerosols (because of the diminished soil water content, not shown) and the reduced wet deposition, which is strongly spatially related ($r = 0.87$) to the decrease in precipitation over southern Europe (see Fig. 3) and specially over the southwestern part of the domain, where the variations in precipitation may achieve −60% during MAM. The changes are not pronounced for the rest of the seasons and parts of the Iberian Peninsula (around 1 μg m^{-3} as an average annual increase), coinciding with those regions with arid or semiarid soils. Despite the strong evidence shown by the simulations for a future increase in local dust levels, it should be highlighted that these results are conditioned by the fact that local dust strongly depends on dust emissions that are occurring on small spatial and temporal scales and the uncertainties of surface wind speed used in the models.

4. Conclusions

This work intended to highlight how future climate change can influence the levels of regional air quality in such a complex terrain as southwestern Europe (focusing on the Iberian Peninsula) for the late XXI century. For that, a regional climate modeling system (coupling MM5−RCM and CHIMERE chemistry transport model) has been applied to two time slices covering a present-climate condition (1991−2010) and the future SRES A2 scenario (2071−2100) in order to obtain regional distributions of both meteorological parameters and air quality concentrations. Results show the plausible influence of climate change alone on the levels of gas-phase pollutants and aerosols.

For regional ozone levels and its main anthropogenic precursor, nitrogen dioxide, large changes are simulated over southwestern Europe for the A2 scenario (over 5 ppb for ozone) during summertime, with strong increases for ozone over the north-western central plateau and lesser increases towards the coast as a consequence of the changes in meteorological parameters (mainly temperature − which enhances isoprene emissions and other biogenic non-methane volatile organic compounds − and specific humidity) under a changing climate. Since the modeled and measured values are close to the target values established by the EU Directive 2008/50/EC in this area (60 ppb as 8-hr average) (Baldasano et al., 2011), the projected increases of 10% in ground level ozone concentrations will contribute significantly to future exceedances of this values. For nitrogen dioxide, slight increases (or even decreases) are only observed in relatively clean areas in the northwestern plateau (hence relative increases are high due to the low background value). Therefore, in these areas (together with the Mediterranean part of the domain), ozone is largely determined by its background concentration. However, this results should be carefully considered because of the large uncertainty of the

sensitivity of isoprene to climate change (Guenther et al., 2006; Forkel and Knoche, 2006; Horowitz et al., 2007).

Regarding aerosols, the response of the diverse components to changes in future climate conditions varies in a wide range. In general, precipitation drives the modification in the concentration of most aerosol components, since the 17% percent decrease of precipitation modeled for the A2 scenario in the Iberian Peninsula leads to a regional increase in the levels of secondary inorganic aerosols and mineral matter (over 4 μg m^{-3} in southwestern Iberian Peninsula during springtime). Also, the enhanced oxidative capacity of the atmosphere causes SO_2 gas-phase emissions to turn into the particulate phase, thus increasing sulfate concentrations. Furthermore, the higher emissions of biogenic precursors of secondary organic aerosols causes a significant increase of around 2 μg m^{-3} in northern Portugal and other continental regions during summertime, as also pointed by Heald et al. (2009). Last, the increased stagnation in wintertime under future climates (decrease in the mixing height by 15% percent over continental Iberian Peninsula) favors the increase of particulate matter in polluted regions.

An important remark has to be done regarding the inherent uncertainties in the climate projections which may affect the results depicted here. Not all climatic variables are affected by the same degree of uncertainty. Future climate simulations tend to agree in a warm trend due to the increase of greenhouse gases concentrations, and only differ in the intensity and spatial distribution details of the warming. However, there is not such a good agreement in the projections of precipitation changes, which strongly affect the results for aerosols as commented before. This larger uncertainty is partly due to the complex mechanism that governs precipitation, which involves a wide variety of spatial scales and it is approximated by different approaches among the state-of-the-art models. In addition to this uncertainty, which can be in principle reduced in the future by improving the models, there is a component of uncertainty in the precipitation projections due to internal variability which is impossible to overcome. In the case of the Iberian Peninsula, the variations of the North Atlantic Oscillation (NAO) modulate the amount of precipitation (Trigo and Palutikof, 2001). However, this important large-scale circulation mode is to a great extent dominated by internal variability, and thus the precipitation amount is partly governed by unpredictable fluctuations at centennial time scale (Luterbacher et al., 2010), making the precipitation projections less reliable than those for temperature. This implies that projection of changes in atmosphere pollutants are affected differently by uncertainty. Those pollutants whose behavior depends more strongly on precipitation projections, such as aerosols, are more charged by uncertainty. On the contrary, projections of pollutants whose behavior is loosely related to this variable, such as gas-phase pollutants, can be in general considered as more reliable.

It is therefore important to characterize and reduce the uncertainties of model projections by having a better idea of the regional air pollution meteorology (especially related to temperature and precipitation, which importantly conditions the difference between aerosol simulations). As pointed out by Jacob and Winner (2009), considering the effect of climate change on air quality in the design of air quality and climate policy will require confidence in model simulations. However, it is widely known that some inadequacies and errors are common to all models and simulations. Hence, future works should be devoted to investigate the differences in air pollution meteorology between diverse parameterizations in the chemistry transport models (physics, anthropogenic and natural emissions, chemistry, deposition), the impact of long-range transport in future air quality projections and the improvement from ensemble-based results of regional air quality-climate simulations (Jiménez-Guerrero et al., 2011b).

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