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Impacts of natural emission sources on particle pollution levels in Europe

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HIGHLIGHTS

- Windblown dust impacts PM10 levels by 20% in southern Europe in summer and winter.
- Sea-salt increases PM10 levels by about 10 μg/m^3 in Mediterranean Sea in summer.
- In Atlantic Ocean, sea-salt enhances PM10 levels by 6 μg/m^3 during autumn.
- Biogenic emissions increase SOA by more than 90% during summer.
- Biogenic emissions reduce PM2.5 levels in central Europe and Eastern Mediterranean.

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ABSTRACT

The main objective of this work is the study of the impact of windblown dust, sea-salt aerosol and biogenic emissions on particle pollution levels in Europe. The Natural Emissions MOdel (NEMO) and the modelling system consisted of the Weather Research and Forecasting model (WRF) and the Comprehensive Air Quality model with extensions (CAMx) were applied in a 30 km horizontal resolution grid, which covered Europe and the adjacent areas for the year 2009. Air quality simulations were performed for different emission scenarios in order to study the contribution of each natural emission source individually and together to air quality levels in Europe. The simulations reveal that the exclusion of windblown dust emissions decreases the mean seasonal PM10 levels by more than 3.3 μg/m^3 (~20%) in the Eastern Mediterranean during winter while an impact of 3 μg/m^3 was also found during summer. The results suggest that sea-salt aerosol has a significant effect on PM levels and composition. Eliminating sea-salt emissions reduces PM10 seasonal concentrations by around 10 μg/m^3 in Mediterranean Sea during summer while a decrease of up to 6 μg/m^3 is found in Atlantic Ocean during autumn. Sea-salt particles also interact with the anthropogenic component and therefore their absence in the atmosphere decreases significantly the nitrates in aerosols where shipping activities are present. The exclusion of biogenic emissions in the model runs leads to a significant reduction of secondary organic aerosols of more than 90% while an increase in PM2.5 levels in central Europe and Eastern Mediterranean is found due to their interaction with anthropogenic component.

1. Introduction

Particulate matter (PM) pollution problems are generally related to the cumulative effect of natural and anthropogenic emission sources (Im et al., 2011a,b; Markakis et al., 2010a,b; Viana et al., 2014). PM of natural origin contributes many times more to the global aerosol burden by mass than the anthropogenic aerosol (Viana et al., 2014) providing a substantial amount of cloud condensation nuclei (CCN). Natural sources enhance PM levels contributing to the deterioration of the air quality and environment (EEA, 2012, 2008) as well as human health; particles are associated

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with an increase in daily mortality (Schwartz et al., 1996). Thus, proper quantification of natural PM emissions as well as the identification of their impacts on air quality is necessary.

Some of the main natural emission sources of primary and secondary atmospheric aerosols are vegetated areas, oceans, arid and semi-arid areas, soils and volcanoes (NATAIR, 2007; Viana et al., 2014). The natural emission sources studied in this work are windblown dust (WD), sea-salt aerosol (SSA) and biogenic volatile organic compounds (BVOCs) emissions from vegetation for which several studies in the past have revealed their impacts on air quality (Athanasopoulou et al., 2008, 2010; Im et al., 2011b, 2013; Viana et al., 2014). Note that NATAIR (2007) indicated that the major contributors of PM10 natural emissions over Europe were WD and SSA.

Windblown dust is generated by wind action on soils that have not been altered or disturbed by human activities (NATAIR, 2007). Note that WD is often difficult to distinguish from man-disturbed soil (i.e. agricultural activities, road traffic, construction), which enhance PM levels (Pikridas et al., 2013; Tolis et al., 2015) contributing significantly to air quality (Chatzimichailidis et al., 2014). Many efforts have been made in the past in order to study dust impact on air quality (Schaap et al., 2009; NATAIR, 2007) while a review of Tsiouri et al. (2014) indicated the adverse health effects that dust episodes have.

Sea-salt aerosol is one of the major components of the atmosphere as oceans cover 70% of the Earth's surface (Ovadnevaite et al., 2012; Tsyro et al., 2011) and it can be produced when wind disturbs water surface (Blanchard, 1989). SSA plays a significant role in the atmospheric chemistry through its reaction with anthropogenic component leading to changes in chemical composition of particles (Tsyro et al., 2011; Athanasopoulou et al., 2008).

BVOCs emissions from vegetation are the major contributor to total Volatile Organic Compounds (Tagaris et al., 2014). Moreover, they play an important role in PM composition due to their oxidation in the troposphere leading to the generation of secondary organic aerosols (SOA) (Froyd et al., 2010; Kanakidou et al., 2005).

Europe exhibits a large seasonal and spatial variability of natural emission sources (Schaap et al., 2009). Southern Europe and in particular the Eastern Mediterranean has been extensively used in the past as study area for simulating natural emissions. Its special climatic conditions, especially during the summer period (dry climate, northerlies) (Athanasopoulou et al., 2008, 2010; Kanakidou et al., 2011; Poupkou et al., 2014), enhance the production of WD. On the other hand, northern Europe and more specifically the Atlantic Ocean are characterized by strong winds during winter enhancing the transport of sea-spray to the continental areas (Tsyro et al., 2011). Biogenic emissions are also significant in the northern Europe (Poupkou et al., 2010).

Natural emissions are an important contributor to background pollution levels in cities and therefore the study of the air quality in Europe by incorporating both natural and anthropogenic sources into photochemical modelling systems and air quality forecasting systems (Marecal et al., 2015) is necessary in order to take the appropriate measures against air pollution.

The aim of the current study is to assess the atmospheric particulate pollution from windblown dust, sea-salt and biogenic emissions in Europe. Up to now, in most cases, the intra-European emissions from windblown dust are usually neglected in the air quality simulations due to the absence of dust emission schemes for Europe in addition to the fact that desert dust transport events are much more severe and frequent (Gerasopoulos et al., 2006; Perez et al., 2008; Remoundaki et al., 2013). However, also windblown dust emitted in Europe is considered to be an important natural source that should be taken into account in regional air quality simulations. Moreover, sea-salt and biogenic emissions are also significant natural sources that impact particle pollution levels in Europe. In the past, many studies have revealed the impact of natural emissions on air quality. However, most of them have studied separately the impact of different emission sources (Tsyro et al., 2011; Tagaris et al., 2014). Furthermore, previous modelling studies are usually presenting results focusing mostly on specific time periods within a year when the natural emissions are at maximum (Im, 2013; Tagaris et al., 2014). This work aims to present a comprehensive study of the impact of the most important natural sources in terms of particle pollution individually and all together by incorporating them into a well-documented photochemical modelling system using a new validated Natural Emissions MOdel (NEMO) that integrates in a single package updated emission methodologies (Liora et al., 2015). Thus, this study builds on the previous research of Liora et al. (2015) where the application and evaluation of NEMO was examined through its incorporation into an air quality modelling system applied on a high spatial resolution grid which covered Europe and the adjacent areas for a whole year. Section 2 presents a description of the modelling system and the emission data used as well as the simulation scenarios examined. In Section 3, the contribution of each natural emission source to total particle emissions and the air quality simulation results are discussed. Simulations are performed for different emission scenarios in order to study the impact of each natural emission source on PM levels. The conclusions of the study are summarized in Section 4.

2. Materials and methods

2.1. Modelling system

In the present study, the modelling system consisted of the Weather Research and Forecasting model (WRF v. 3.5.1; Skamarock et al., 2008) and the three-dimensional Comprehensive Air Quality Model with extensions (CAMx v.5.3; ENVIRON, 2010). The MACC reanalysis (Inness et al., 2013) run with the coupled model IFS-MOZART (Morcrette et al., 2009; Stein et al., 2012) and the EMEP MSC-W model (Simpson et al., 2012) provided the concentration data used as boundary conditions for the CAMx simulations. The Natural Emissions Model (NEMO; Liora et al., 2015) was used for the calculation of the natural PM emissions from WD and SSA as well as the BVOCs emissions from vegetation including isoprene, terpenes and other volatile organic compounds (OVOCs). The Model for the Spatial and Temporal Distribution of Emissions (MOSESS; Markakis et al., 2013) was used to spatially, temporally and chemically process the 2009 TNO-MACCII anthropogenic emission database (Kuenen et al., 2014). In addition, monthly potential anthropogenic PM emissions of mineral dust from agricultural activities and annual potential PM emissions from road traffic re-suspension were provided by The Netherlands Organisation (TNO). The anthropogenic dust emission data which had been estimated using the LOTOS-EUROS model (Schaap et al., 2009) were temporally and spatially analysed for the 30 km horizontal resolution grid using the model MOSESS as well as temporal profiles provided by TNO and taking into account the restriction that mineral dust emissions were forced to zero during precipitation events.

Aerosol processes, in CAMx, were modelled using a static two-mode fine/coarse scheme for the representation of the particle size distribution; fine were particles with diameter up to 2.5 μm while coarse were the larger particles with diameter from 2.5 μm to 10 μm. Fine particles ($PM_{2.5}$) were speciated as sulphates (PSO4), nitrates (PNO3), particulate ammonium (PNH4), sodium (Na), particulate chloride (PCl), primary organic aerosols (POA), primary elemental carbon (PEC), crustal, other primary fine particle and secondary organic aerosols (SOA). Coarse particles ($PM_{2.5\text{-}10}$) were

speciated as crustal and other primary coarse ones. The total simulated SOA derive from the sum of biogenic SOA and anthropogenic SOA (ENVIRON, 2010).

Dust emissions (natural and anthropogenic origin) were simulated as fine ($PM_{2.5}$) and coarse ($PM_{2.5-10}$) crustal particle emissions. $PM_{2.5}$ sea salt emissions were split into particulate chloride (55.04%), sodium (30.61%), sulphates (7.68%) and other primary fine particles (6.67%) emissions (Seinfeld and Pandis, 2006) while coarse sea salt emissions were simulated like other primary coarse particles. The oxidation of biogenic emissions from vegetation results in the production of SOA (Kanakidou et al., 2005) that are simulated in CAMx as biogenic SOA and polymerized biogenic SOA.

The modelling system was applied in a 30 km horizontal resolution grid (141 × 134 cells), which covered Europe and the adjacent areas for the year 2009 (Fig. 1).

2.2. Simulations

In order to investigate the impact of natural sources on PM levels in Europe, the WRF-CAMx modelling system was implemented for five emission scenarios by applying the widely used zero-out modelling method (Im and Kanakidou, 2012; Poupkou et al., 2008): simulations including and omitting the various natural emissions were performed. The scenarios studied are listed below:

1. Base run scenario (Base): it includes all natural and anthropogenic emissions mentioned in Sect. 2.1.
2. No windblown dust scenario (NoWD): Base scenario excluding WD emissions.
3. No sea-salt aerosol scenario (NoSSA): Base scenario excluding SSA emissions.
4. No biogenic scenario (NoBVOCs): Base scenario excluding BVOCs emissions.
5. No natural sources scenario (NoNAT): Base scenario excluding all natural emissions (WD, SSA, BVOCs).

The aforementioned simulations were performed with an hourly temporal analysis so as to examine the impact of natural sources individually and all together on the air quality in Europe. In particular, the annual and seasonal (winter, spring, summer and autumn) impact of each natural emission source in terms of mean total PM10 or PM2.5 mass concentrations is examined by calculating the differences in mean total PM concentrations between the examined scenario (i.e. NoWD, NoSSA, NoBVOCs or NoNAT) and the "Base" scenario. Moreover, the percentage contribution of natural sources to total PM10 or PM2.5 levels over Europe is presented. The percentage contribution (in %) is estimated by calculating the ratio of the difference in mean total PM concentrations to the mean total PM concentrations of the Base scenario. In addition, in some cases, the impact of natural sources on specific PM species (e.g. sulphates, nitrates etc.) is also examined. The increase or decrease in PM concentration data reveals the impact of each natural emission source.

It should be mentioned that this paper focuses on the study of the impact of local natural emission sources inside the European domain and therefore the chemical boundary conditions were kept

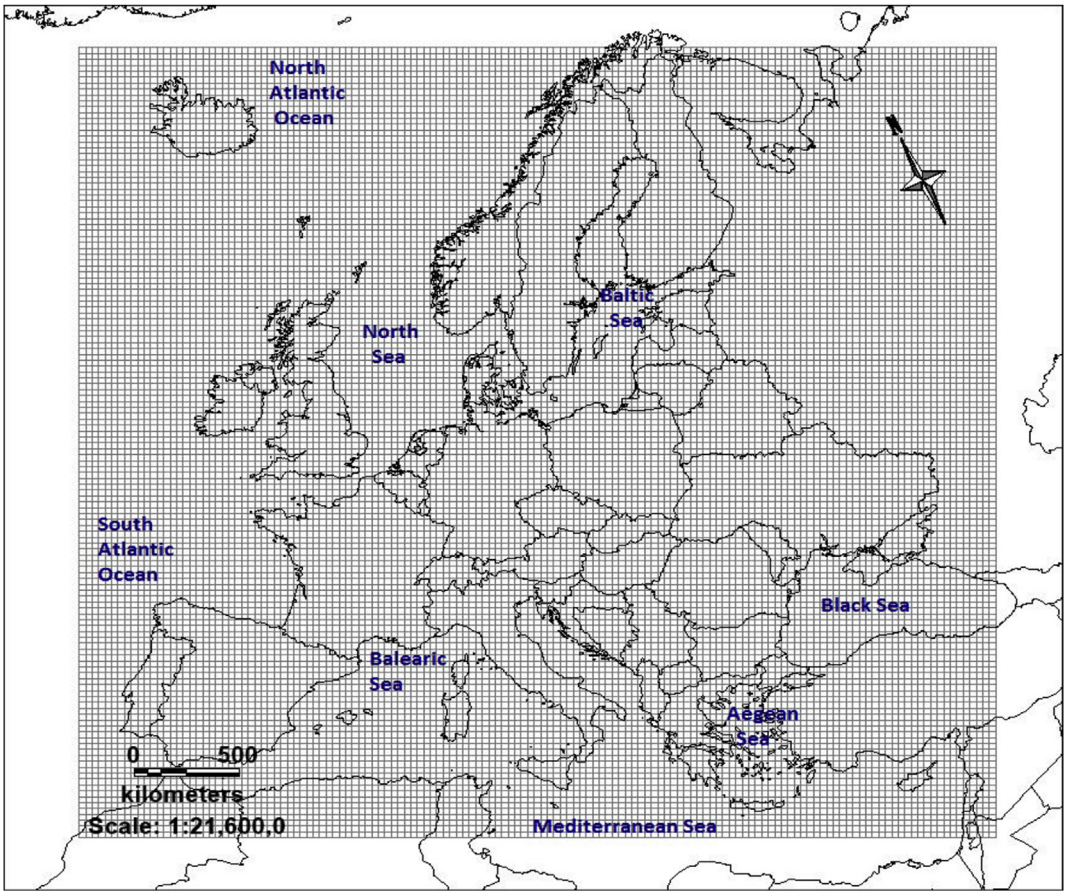

Fig. 1. Model domain (30 × 30 km^2 spatial resolution).

unchanged while running CAMx for the emission scenarios examined (i.e. NoWD, NoSSA, NoBVOCs, NoNAT).

The application and evaluation of the current modelling system, in terms of WD and SSA emissions, has been thoroughly described in Liora et al. (2015) indicating that the contribution of natural sources to PM levels was captured fairly well by the modelling system. In the current study, the spatial distribution of mean annual PM levels of the "Base" scenario is presented in Fig. S1 (in supplement). The comparison between the simulated and observed total PM concentration data in the selected monitoring sites described in Liora et al. (2015), where natural PM concentration data were available, is also presented. The evaluation reveals that the ratios between observed and simulated PM levels are close to unit in most of the monitoring sites studied. The Index of Agreement (equation S.(2)) ranges mostly from 0.53 to 0.68 (Tables S1 and S2 in supplement) suggesting that the modelling system produces fairly well the total PM concentrations and indicating the robustness of the current air quality modelling application. The absolute differences between observed and simulated PM values in most of the monitoring sites reach up to around 28% which are within the acceptable limits according to previous modelling studies (Pirovano et al., 2012; Prank et al., 2016). Thus, the performance of the modelling system is satisfactory in sites where natural data are available.

3. Results and discussion

3.1. Emissions

The natural and anthropogenic emissions over the model domain (Fig. 1) for the year 2009 are presented in Table 1. SSA is the major emission contributor to both PM10 and PM2.5 annual emissions, followed by the overall anthropogenic sources (dust and other sources). In particular, SSA contributes by around 73% and 70% to PM10 and PM2.5 emissions, respectively while the corresponding contribution of the overall anthropogenic sources is 25% and 30%, respectively. WD emissions have a very small impact (2% for PM10 emissions) while anthropogenic dust contributes by around 10% to total PM10 emissions. It should be noted that the aforementioned percentage contributions refer to the whole study domain with its high water coverage leading to high SSA contribution. BVOCs emissions from vegetation estimated as 20 Tg from which isoprene, monoterpenes and OVOCs represent a 15%, 28% and 57% share, respectively. BVOCs represent a 60% share to total Non-Methane VOCs (NMVOCs) emissions in the study domain. The estimation of OVOCs emissions is characterized by larger uncertainties compared to those for isoprene and monoterpenes emissions considering the fact that OVOCs consist of a large number of species, including hydrocarbons and oxygenated compounds, and have been proven difficult to quantify in atmospheric samples (EEA, 2013).

Before studying the impact of natural emissions on PM levels, it is important to examine their spatial contribution to total PM emissions across Europe. Fig. 2a–b depicts the spatial contribution (in %) of WD and SSA to total PM10 annual emissions in Europe for 2009 while Fig. 2c illustrates the corresponding contribution of vegetation (BVOCs) to total NMVOCs emissions.

Table 1
Natural and anthropogenic emissions (Tg/year) over the study area (Europe and adjacent areas (Fig. 1)) for the year 2009.

Emission source	Annual emissions (Tg/yr)		
	PM10	PM2.5	NMVOCs
Windblown dust	0.572	0.034	–
Sea salt aerosols	20.31	6.86	–
Vegetation	–	–	19.96
Anthropogenic mineral dust	2.88	0.36	–
Other anthropogenic sources	4.59	2.51	12.50

WD is the major contributor (up to 85%) to PM10 emissions in Turkey mostly due to the drier climate in combination with the high wind speed values (Liora et al., 2015). Southern Greece (i.e. Crete, Peloponnesus) is characterized by intense WD events (Liora et al., 2015) leading to a 40%–80% WD contribution. In southern Spain, most WD events have a small contribution (<10%) while others contribute by 20–80% and more to PM10 emissions. In central Europe, sparse WD episodes can be found with a moderate contribution to PM10 emissions (<40%) with the majority of them showing a small impact (<10%). In Southern Russia and Ukraine, WD contribution is also important (up to 60%).

According to Fig. 2b, SSA is the major contributor (>90%) over the majority of sea regions with few exceptions across the ship lines where the corresponding contribution ranges from 70% to 90%. However, in Baltic Sea, the low SSA emissions attributed to the low water salinity values (Liora et al., 2015) and the high shipping activities lead to a 20–40% contribution of SSA to PM10 emissions across the shipping lines.

Biogenic emissions from vegetation have a very high contribution to total NMVOCs emissions in the northern Europe (>95%) due to the highest monoterpenes emissions in the area in addition to the low anthropogenic emissions. This distribution of monoterpenes is mainly associated with the boreal forests, which are characterized by needle leaf tree species (Poupkou et al., 2010). In central Europe, BVOCs emissions contribute by around 20–60% while in the Eastern Mediterranean the contribution is much higher. Spain is also characterized by high BVOCs contribution; over 60% in most of the areas associated with evergreen oak woodland emissions (Poupkou et al., 2010).

3.2. Impacts of natural emissions on air quality

3.2.1. Windblown dust

The impact of WD emissions on the European air quality is examined with the simulation NoWD in which WD PM10 and PM2.5 emissions calculated with NEMO over the whole study area have been omitted. The effect of WD emissions from natural sources on the air quality is mostly limited over the southern Europe due to its climatic conditions (e.g. drier climate, high wind speeds).

In the Eastern Mediterranean, on an annual basis, the exclusion of WD emissions decreases PM10 concentrations by more than 0.9 $\mu g/m^3$ (~6%) in the western Turkey (Fig. 3). This important effect derives from some intense dust events appeared during winter (~20% decrease in PM10 mass) when PM10 levels decreased by more than 3.3 $\mu g/m^3$ (Fig. 4b). This peak in PM10 concentrations due to WD production is attributed, for the most part, to the strong wind speeds in combination with the absence of precipitation events. In southern Turkey and southern Greece, a lower annual contribution of up to 5% and up to 4.5% is shown, respectively, due to the summertime WD episodes (Fig. 4e, f); a maximum impact of 3 $\mu g/m^3$ on PM10 seasonal levels is shown in Turkey. Cyprus is affected by the WD events occurred in south Turkey, apart from the local dust events, leading to a contribution of up to 1.5 $\mu g/m^3$ during summer. In southern Greece, the corresponding seasonal contribution to PM10 levels in summer 2009 is up to 2.5 $\mu g/m^3$.

In the western Mediterranean, WD impacts PM10 concentrations by up to 0.7 $\mu g/m^3$ (4.5%) on an annual basis. In particular, in southern Spain, WD events occurred in winter and autumn led to a maximum seasonal contribution of 10% and 6%, respectively. In France, dust events of moderate intensity appeared in autumn resulted to a seasonal impact of up to 0.6 $\mu g/m^3$ on PM10 levels.

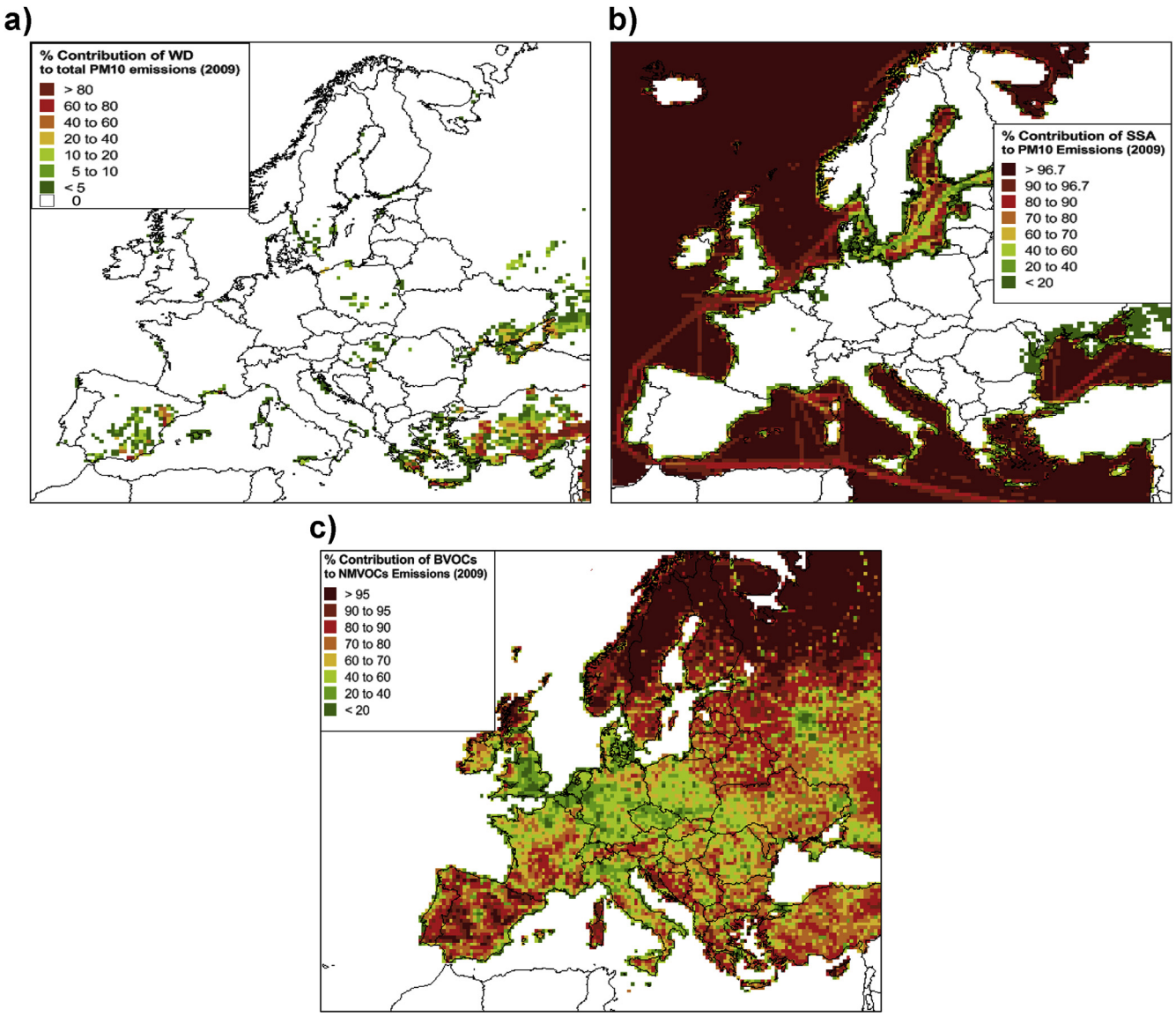

Fig. 2. Percentage contribution of a) windblown dust (WD) and b) sea-salt aerosol (SSA) to total PM10 annual emissions and c) % contribution of biogenic (BVOCs) emissions to total NMVOCs emissions over Europe in 2009.

In the rest part of the domain, the annual impact of windblown dust emissions to PM10 levels is negligible (<0.1 μg/m^3 and <0.1%). This is in accordance with previous studies (Schaap et al., 2009; NATAIR, 2007). According to Schaap et al., 2009, the average annual contribution of WD in coastal areas of Mediterranean Sea is up to 1 μg/m^3, similarly to the current study (Fig. 3b). WD has a negligible impact on PM2.5 levels over the study domain and therefore their spatial contribution is not presented.

It should be mentioned that WD impacts only crustal particles, as its contribution to other species is almost zero and for this reason it is not presented.

3.2.2. Sea-salt aerosol

The impact of SSA on air quality in Europe is investigated through the NoSSA simulation in which SSA PM10 and PM2.5 emissions estimated with NEMO have been excluded in CAMx runs.

On an annual basis, eliminating SSA emissions reduces PM10 levels in northern Atlantic Ocean and western Mediterranean by up to 35% while the corresponding reduction in PM2.5 concentrations is largest in the Atlantic Ocean (more than 35%) followed by the Mediterranean Sea (up to 20%) (Fig. 5a and c). This higher relative impact over North Atlantic is also due to the absence of anthropogenic emissions while in the Mediterranean Sea there are more anthropogenic shipping activities that emit mostly fine particles. However, it should be pointed out that despite the fact that anthropogenic emissions are very limited in the north Atlantic Ocean, the contribution of sea salt on PM levels is not higher than reported. This can be attributed to the influence on PM levels from the boundary conditions. Thus, the overall contribution of sea salt (including local sea salt particles plus sea salt transferred from the boundaries) would be much higher than 35% over the northern part of Atlantic Ocean. On absolute values, the exclusion of SSA decreases annual PM10 levels by up to 4 μg/m^3 and more than 6 μg/m^3 in southern Atlantic Ocean and Mediterranean Sea, respectively. In Black and Baltic Sea, low SSA emissions due to the lower salinity values (Liora et al., 2015) lead to a smaller impact, compared to other sea regions, of up to 2 μg/

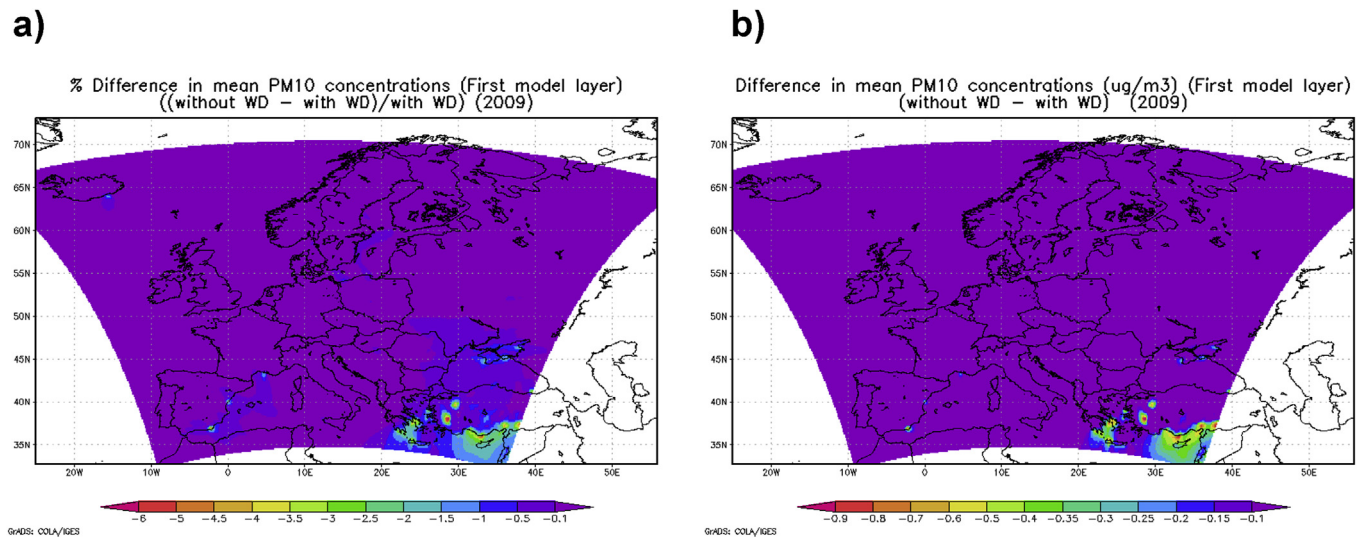

Fig. 3. Mean annual a) percentage contribution of windblown dust (WD) to PM10 levels and b) impact of WD on PM10 concentrations (in μg/m^3) in 2009.

m^3 on PM10. SSA can also affect areas near the coast, however, the contribution drops rapidly inland.

In continental areas of southern Europe, the impact of SSA on PM10 and PM2.5 levels is up to 1 μg/m^3 and 0.8 μg/m^3, respectively. However, a larger contribution is found in inland Crete (~4 μg/m^3 for PM10 levels) and Cyprus (~3 μg/m^3 for PM10 levels). In central Europe, in the NoSSA scenario, PM10 and PM2.5 concentrations decrease by 2–4% (<0.4 μg/m^3). In coastal areas located in Atlantic Ocean (UK, Ireland), the corresponding contribution is 2–3 μg/m^3, representing ~16% of PM10 mass.

The impact of SSA on the air quality is seasonally and spatially differentiated due to the different meteorological conditions between the different seasons and areas as well as the interactions with anthropogenic components (Athanasopoulou et al., 2008). Sea salt particles alter the composition of particulate matter and their size distribution leading to changes in sodium and chloride levels as well as to an enhanced formation of sulphates and nitrates over the marine areas (Fig. 6). It should be mentioned that in the current study only the impact of sea salt on the fine fraction of sea salt components (sodium, chloride etc) is examined due to the CAMx configuration applied.

Sodium mostly originates from sea salt (Tsyro et al., 2011) and therefore the effect of SSA on sodium levels is very high; in open sea areas of Mediterranean Sea, the decrease in Na mean annual concentrations, in NoSSA scenario, ranges from 70% to over 80% (0.6–0.9 μg/m^3) while in northern Atlantic Ocean the highest Na contribution is up to 75% (0.6 μg/m^3). The reactions of sea salt particles with sulphuric or nitric acids in the atmosphere lead to a chloride loss in the aerosol explaining the low impact of SSA on mean annual chloride concentrations over the Mediterranean region (<0.25 μg/m^3) where anthropogenic emissions are present.

Sea salt particles interact with anthropogenic component and more specifically they act as a sink for sulphuric and nitric acid in the atmosphere (Athanasopoulou et al., 2008; Im, 2013; Tsyro et al., 2011) resulting to changes in particulate sulphate and nitrate. Sulphates are mostly found as fine particles while nitrates can be found in both modes fine and coarse (Seinfeld and Pandis, 2006). Hence, SSA contributes to PNO3 annual levels by over than 65% in Mediterranean Sea. Comparing Figs. 2 and 6d, an enhanced formation of nitrates, due to SSA emissions, is shown in western Mediterranean Sea and North Sea (0.35–0.5 μg/m^3) where shipping lines are present. In Baltic Sea, despite the high contribution of shipping activities, a much lower impact compared to that over Mediterranean and North seas is presented (up to 0.2 μg/m^3) due to the very low SSA emissions over the area. The mean annual impact of SSA on sulphates particles ranges from 4% to 7% in Mediterranean Sea while it is almost double in northern Atlantic. On absolute values, the maximum impact of SSA on PSO4 annual levels is found in Mediterranean Sea (up to 0.25 μg/m^3) attributed to the higher SSA emissions found in the area.

In addition, the elimination of SSA result to a small increase in ammonium particles levels over areas with high sea-salt contribution. However, the impact is considered negligible over Europe (a maximum increase of around 0.14 μg/m^3 is found over the marine areas of Eastern Mediterranean Sea) and therefore is not presented. This increase in PNH4 levels with the omission of SSA emissions is attributed to the fact that more ammonia reacts with sulphuric or nitric acid due to absence of sea salt particles (no reaction between acids and sea-salt) and therefore more ammonium is produced.

On a seasonal basis, over marine areas, SSA contribution to PM10 levels is more pronounced during summer, being the highest in eastern Mediterranean Sea (>50%) leading to an important impact on PM10 concentrations (>10 μg/m3) (Fig. 7). This effect is presented in Aegean Sea and it is in agreement with previous studies (Athanasopoulou et al., 2008; Im, 2013). The eastern Mediterranean is characterized by very strong north winds, the so-called “Etesian” which blow during the summer period (Poupkou et al., 2011) leading to maximum SSA emissions in Aegean Sea during summer. A significant impact on PM10 levels (>10 μg/m^3) is also found in the western Mediterranean due to the strong wind speeds. In North Atlantic Ocean, SSA contributes by up to 50% (up to 4 μg/m^3) in summer due to the higher water temperature values in combination with the moderate wind speeds. According to NATAIR, the impact of sea salt on PM10 levels reached up to 10 μg/m^3 over marine areas of Atlantic Ocean, attributed to the different methodology used where only wind speed parameterization was taken into account. During winter and autumn, SSA contributes by up to 35% in North Sea and western Mediterranean. In spring, the impact of SSA on PM10 levels reaches up to 3 μg/m^3 and ~4 μg/m^3 over

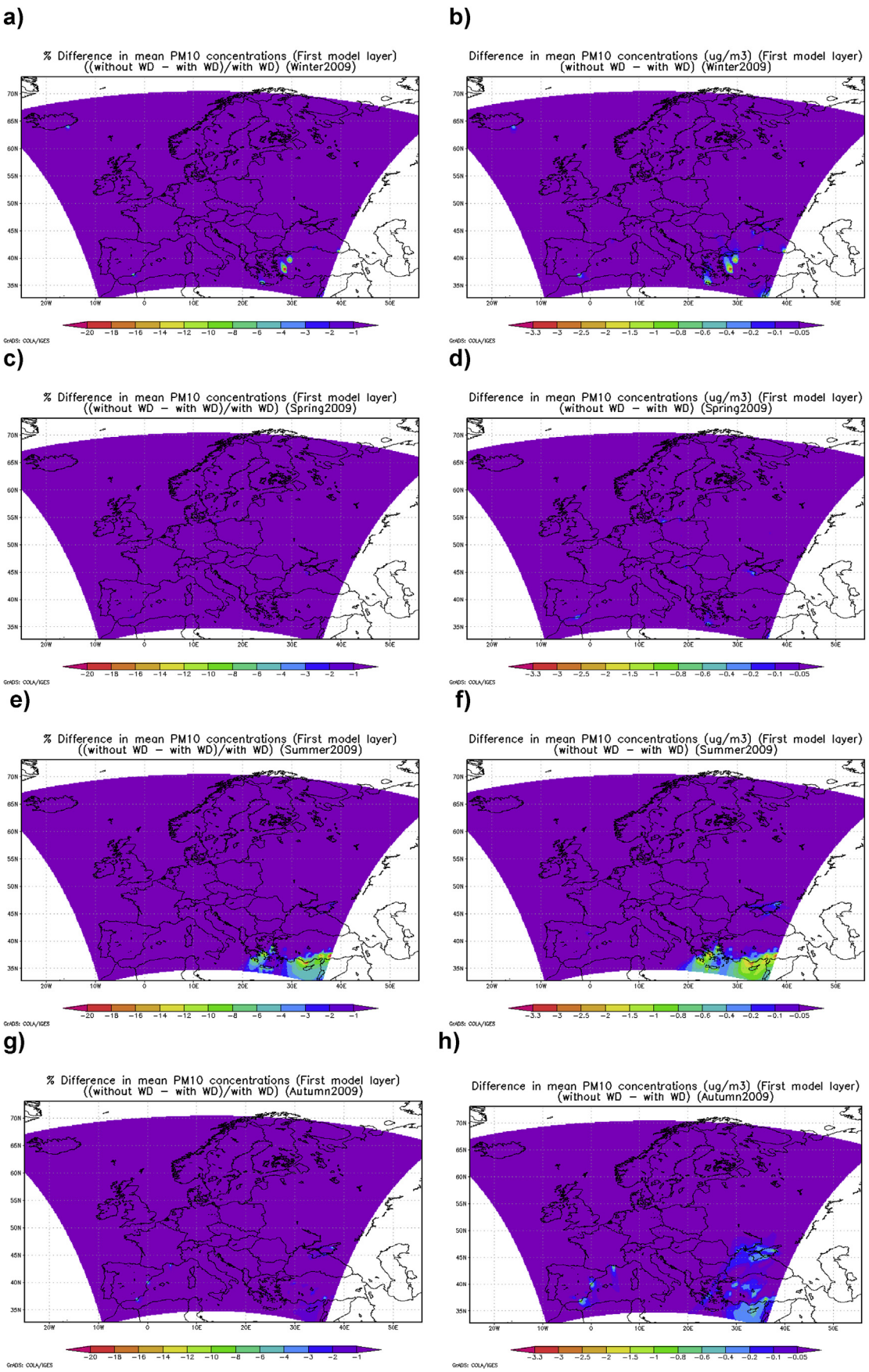

Fig. 4. Mean seasonal percentage contribution (in %) of windblown dust (WD) to PM10 levels and seasonal impact of WD on PM10 concentrations (in $\mu g/m^3$) for winter (a,b), spring (c,d), summer (e,f) and autumn (g,h) for 2009.

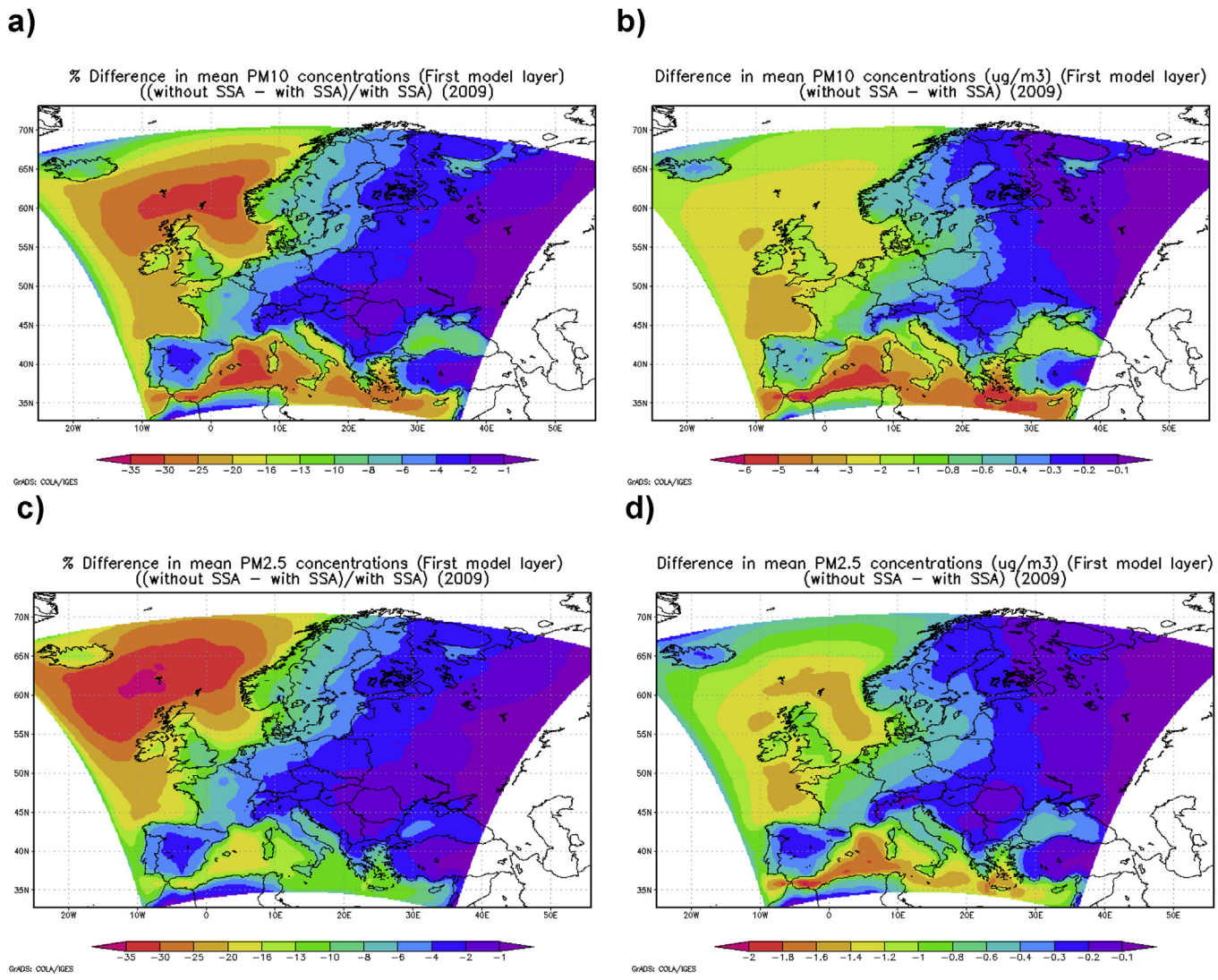

Fig. 5. Mean annual percentage contribution (in %) of sea-salt aerosol (SSA) to PM10 and PM2.5 levels (a, c) and mean annual impact of SSA on PM10 and PM2.5 levels (in $\mu g/m^3$) (b, d) for 2009.

Atlantic Ocean and Mediterranean Sea, respectively. Over inland areas, the maximum impact of SSA is found during autumn; the exclusion of SSA leads to a decrease of up to 6 $\mu g/m^3$ in PM10 levels in parts of southern Atlantic Ocean and in North Sea. This effect in combination with the strong winds over the Atlantic Ocean impacts the continental areas of western and northern Europe (0.4–4 $\mu g/m^3$).

Finally, it seems that even though the maximum percentage contribution of sea salt to PM levels is found mostly over the north Atlantic Ocean, the decrease in PM levels, when SSA emissions are absent, is the highest in Mediterranean Sea. This is attributed to the fact that Mediterranean Sea is characterized by high SSA emissions mainly due to its warmer waters leading to higher impact on PM concentrations. On the other hand, the presence of anthropogenic sources in the area reduces the overall contribution of sea salt to total PM levels which is maximum in Atlantic Ocean where anthropogenic sources are limited.

3.2.3. Biogenic emissions

BVOCs emissions from vegetation are considerable during the summertime (Oderbolz et al., 2013; Poupkou et al., 2010) and therefore their influence on air quality is examined only during this period. Moreover, they contribute, for the most part, to fine particles as they result to the formation of secondary organic aerosols, which are mostly found in fine mode (Seinfeld and Pandis, 2006).

According to Fig. 8, the exclusion of biogenic emissions in the air quality simulations is predicted to decrease PM2.5 levels by more than 0.5 $\mu g/m^3$ (>16%) in the northern Europe during summer in which vegetation is the major contributor to NMVOCs emissions (Fig. 2). On the other hand, an increase in PM2.5 concentrations is shown in a large part of the rest Europe when biogenic emissions are omitted in the photochemical model runs. The highest increase in PM2.5 mass is found in central Europe as well as in the Eastern Mediterranean (>0.4 $\mu g/m^3$ (>4%)). This negative or positive change in particle levels with the elimination of biogenic emissions, mainly due to their interaction with anthropogenic component, is in agreement with previous studies (Im and Kanakidou, 2012; Tagaris et al., 2014; Megaritis et al., 2014). In particular, Tagaris et al. (2014) had found a maximum simulated reduction of 1 $\mu g/m^3$ in PM2.5 concentrations over southern Europe in July 2006 due to biogenic emissions while in other areas an increase in PM2.5 levels was

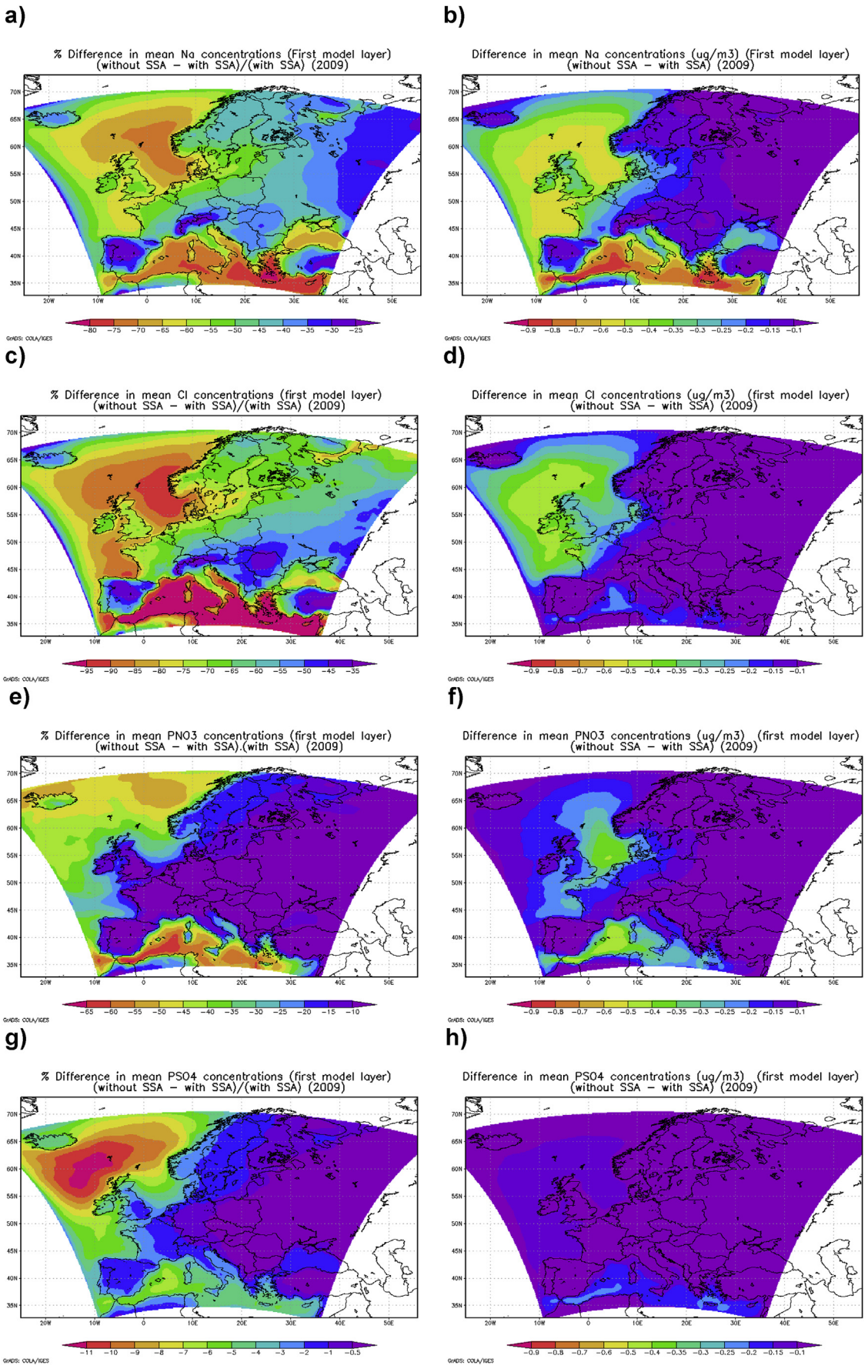

Fig. 6. Mean annual percentage contribution of SSA and impact of SSA on sodium (Na) (a, b), chloride (Cl) (c, d), nitrates (PNO3) (e, f) and sulphates (PSO4) (g, h) aerosol concentrations (in % and μg/m^3) for 2009.

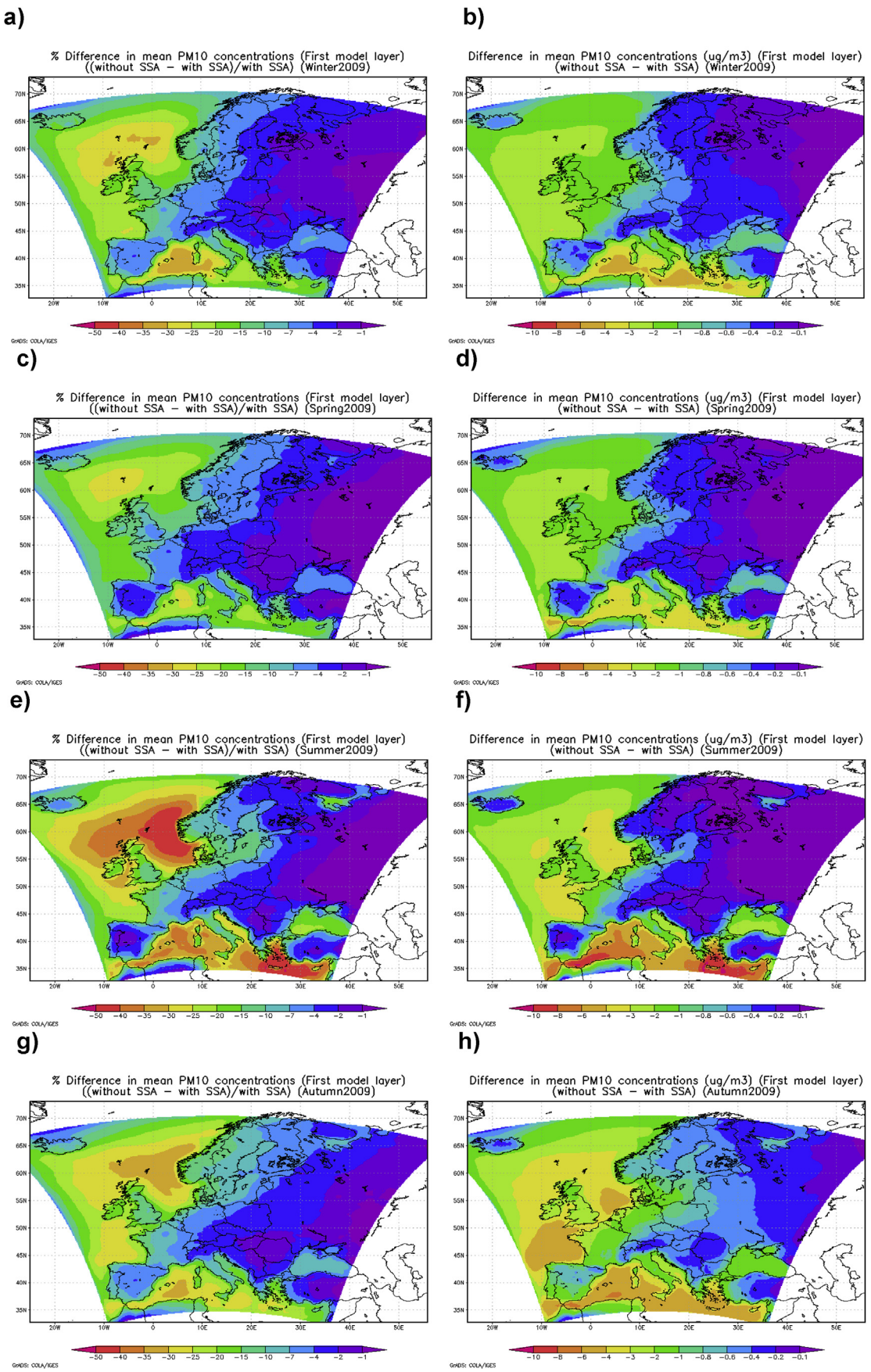

Fig. 7. Mean seasonal percentage contribution (in %) of SSA to PM10 levels and mean seasonal impact of SSA on PM10 (in $\mu g/m^3$) for winter (a,b), spring (c,d), summer (e,f) and autumn (g,h) for 2009.

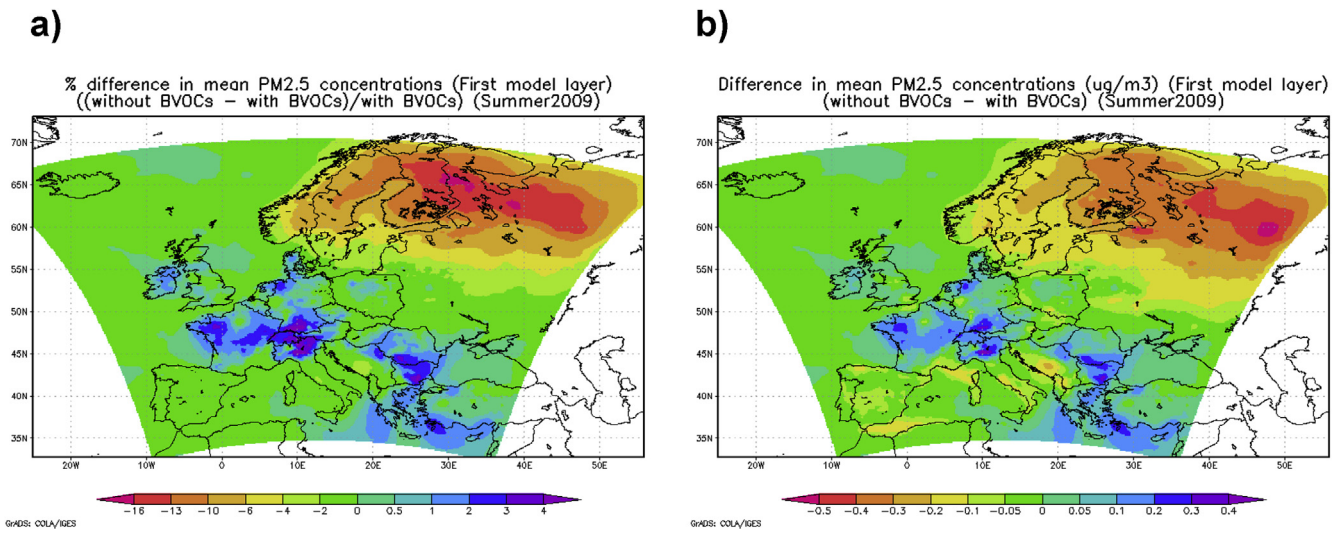

Fig. 8. Mean seasonal a) percentage contribution (in %) of biogenic (BVOCs) emissions to PM2.5 levels and b) impact of biogenic emissions on PM2.5 concentrations (in $\mu g/m^3$) during summer 2009.

found. In order to investigate thoroughly the response of PM levels in biogenic emissions it is important to study their chemical composition. Fig. 9 illustrates the impact of biogenic emissions on SOA, PNO3 and PSO4 levels. Their impact on other particles species is negligible.

According to Fig. 9, omitting biogenic emissions results to a significant decrease in SOA concentrations (>90%) over land in Europe in summer 2009 concluding that secondary organic aerosols are originated mostly from biogenic emissions. The maximum absolute decrease in seasonal SOA values has been estimated in northern Europe ranging between 0.35 and 0.6 μg/m3. Previous studies have indicated the importance of biogenic emissions to the formation of SOA (Hallquist et al., 2009; Kanakidou et al., 2005; Mochizuki et al., 2015).

Moreover, biogenic emissions affect the lifetime of hydroxyl (OH) (Megaritis et al., 2014) and therefore its decrease in the atmosphere. OH contributes to the formation of sulphuric or nitric acid in the atmosphere through the oxidation of SO_2 or NO_x. Thus, the increase of OH leads to the increased formation of sulphuric and nitric acids reacting with ammonia resulting in an increased production of particles sulphates and nitrates. Hence, depending on the presence of the anthropogenic pollutants in the atmosphere an increase or decrease in nitrates and sulphates concentrations is expected when BVOCs emissions are omitted. Hence, a significant increase in nitrates concentrations (0.1–0.4 $\mu g/m^3$) is found over central Europe where anthropogenic sources are the major contributor to NMVOCs emissions (Fig. 2c), with the exclusion of BVOCs emissions. Biogenic emissions have both negative and positive response in PSO4 levels over Europe. In the eastern Mediterranean, an increase in PSO4 levels can be seen with the highest positive values reaching up to 0.4 $\mu g/m^3$ (~9%). Tagaris et al. (2014) had found also a reduction in particulate sulphate with the addition of biogenic emissions in the Eastern Mediterranean (up to 17% for July). In the remaining part of the domain a small decrease in sulphate concentrations is visible (~0.15 $\mu g/m^3$) for the NoBVOCs scenario. Finally, BVOCs, similarly to SSA, reduce PNH4 levels over Europe. However, their impact is negligible; the maximum decrease of up to 0.14 $\mu g/m^3$ is shown in central Europe.

3.2.4. All natural sources

In order to investigate the cumulative effect of natural sources on the air quality in Europe, the NoNAT simulation results are examined. In NoNAT simulation scenario, PM10 and PM2.5 emissions from windblown dust and SSA as well as BVOCs emissions from vegetation were omitted. According to Fig. 10, the highest annual percentage contribution of natural sources to PM10 levels (~30%) is shown in the northern part of Atlantic Ocean and the western Mediterranean Sea due to the high contribution of sea salt to total PM10 emissions. The maximum impact of natural sources, on absolute values, in Mediterranean Sea is around 6 $\mu g/m^3$ and 2 $\mu g/m^3$ to PM10 and PM2.5 annual levels, respectively. Coastal PM10 mass over southern Europe decreases by up to 2 $\mu g/m^3$ (up to 13%) in areas located close to coastlines, due to the cumulative effect of natural sources. WD and SSA have a negative impact in the area while biogenic emissions result in a slight increase in particle levels when natural sources are omitted. The lowest impact of natural sources on PM levels is shown in central Europe (<0.2 $\mu g/m^3$). Over northern Europe, a decrease of around 0.6 $\mu g/m^3$ (~10%) in PM levels is presented due to the effect of biogenic emissions in addition with that of sea-salt particles transported from Atlantic Ocean.

As it has been shown in the previous sections, natural sources alter the composition of particulate matter. In particular, the absence of biogenic emissions in the atmosphere leads to an overall increase of up to 10% in nitrates in central Europe and Balkans (Fig. 11). In coastal areas, a small decrease (~10%) in PNO3 is shown due to the impact of SSA. Natural sources have a negligible impact on particle sulphates, for the most part, over land (±0.05 $\mu g/m^3$). However, an increase of up to 0.1 $\mu g/m^3$ is found in Balkan area due the interaction of biogenic particles to the anthropogenic ones. A larger decrease in PSO_4 levels is observed in coastal areas located close to Atlantic Ocean due to SSA effect. Natural sources (SSA and vegetation) increase ammonium particles levels, however, their contribution is negligible and therefore it is not shown. It should be noted that WD does not alter any of the aforementioned particle species. WD impacts only crustal particles, as its contribution to other species is almost zero.

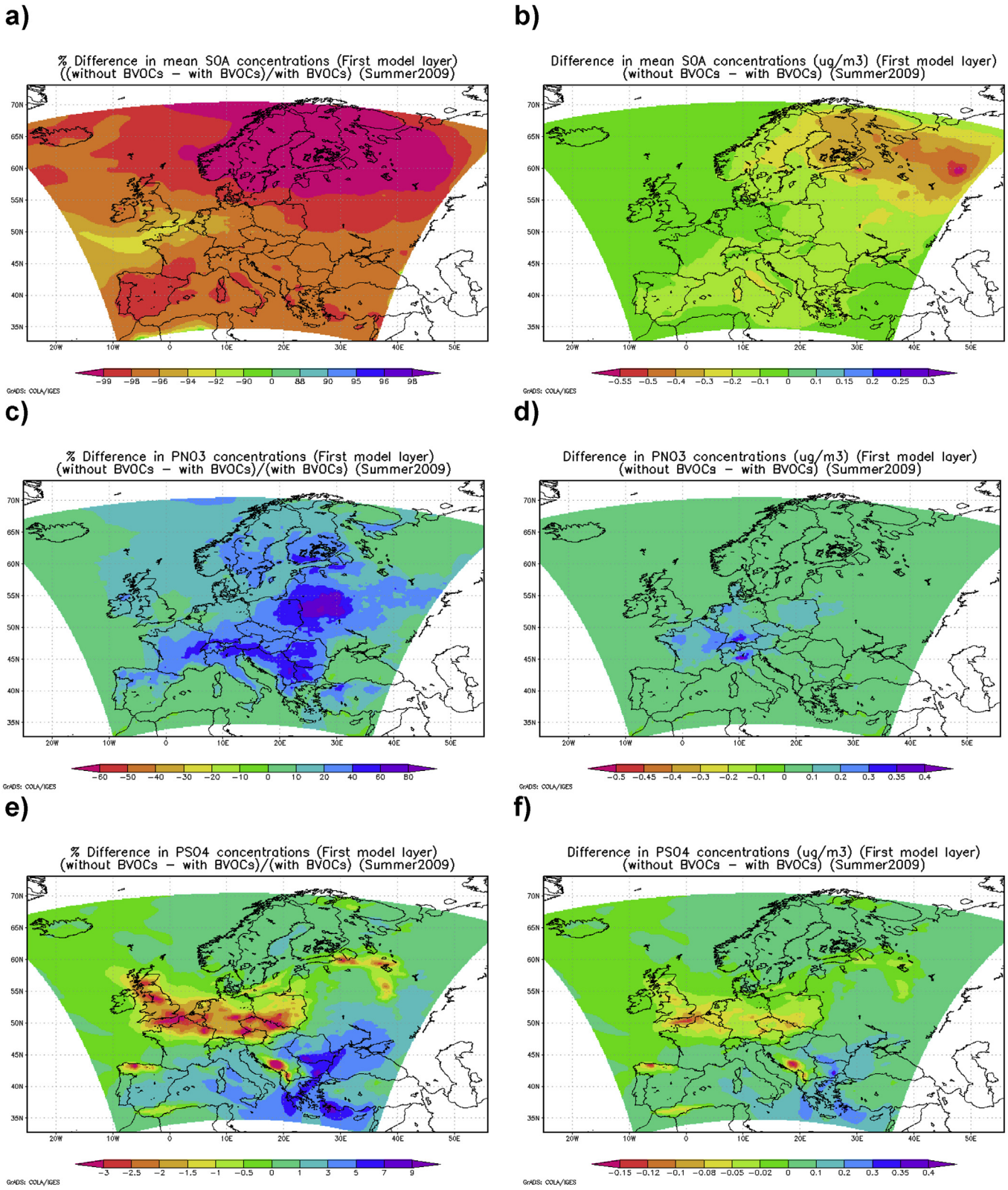

Fig. 9. Mean seasonal percentage contribution of biogenic emissions (BVOCs) and mean seasonal impact of BVOCs emissions secondary aerosols (SOA) (a, b), nitrates (PNO3) (c, d) and sulphates (PSO4) (e, f) concentrations (in $\mu g/m^3$ and %) in summer 2009.

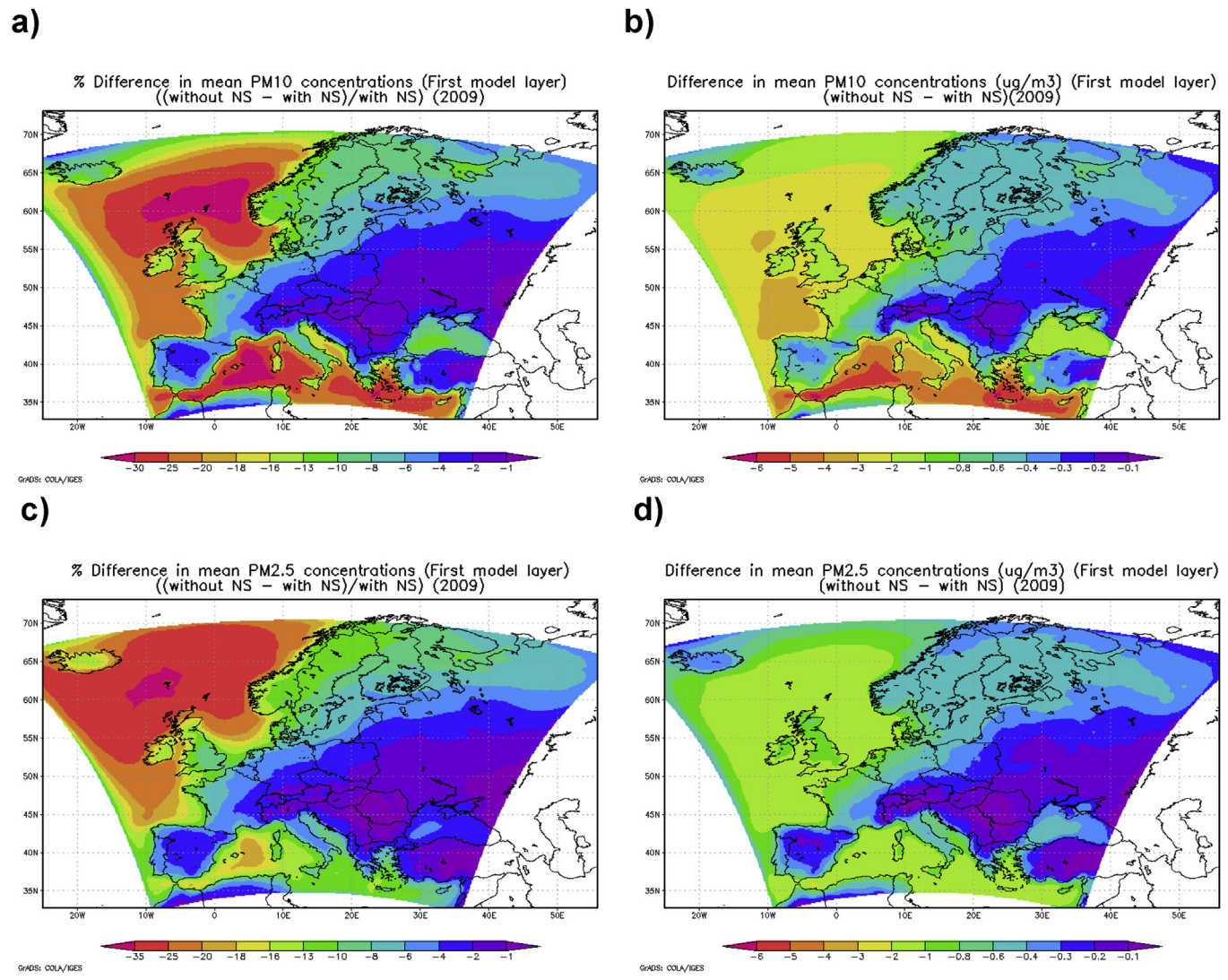

Fig. 10. Mean annual percentage contribution of natural sources (NS) to PM10 and PM2.5 levels (a, c) and impact of natural sources on mean PM10 and PM2.5 concentrations (in μg/m^3) (b, d) for 2009.

4. Conclusions

The effects of windblown dust, sea-salt and biogenic emissions on particulate matter levels in Europe have been investigated through the application of the air quality WRF-CAMx modelling system for the year 2009. PM natural emissions as well as biogenic emissions have been derived from the application of NEMO. The impact of natural sources on the air quality over Europe has been examined through the implementation of five simulation scenarios suggesting that the effects of natural emissions are seasonally and spatially differentiated.

In particular, WD emissions impact PM10 seasonal concentrations over the Eastern Mediterranean by around 3.5 μg/m^3 and 3 μg/m^3 during winter and summer, respectively. The high contribution of WD in southern Europe in winter and summer derives from the high wind speed values and the dry climate, respectively. The exclusion of SSA decreased PM levels in Atlantic Ocean mainly during autumn while a significant effect of SSA during summer was found in the Mediterranean Sea. Moreover, sea-salt particles altered the PM composition through their interaction with anthropogenic particles leading to changes in nitrate and sulphate generation. The effect is more pronounced over the areas characterized by shipping activities where a reduced formation of nitrates in aerosol composition was found with the elimination of SSA emissions in model runs. Biogenic emissions had both positive and negative response in PM levels mainly due to their interaction with anthropogenic component. More specifically, high biogenic emissions in the northern Europe in combination with the absence of anthropogenic NMVOCs emissions led to an impact of around 16% on PM levels during summer due to the enhanced formation of SOA. On the other hand, in central Europe the low contribution of biogenic emissions to total NMVOCs resulted to a slight decrease in PM levels due to the corresponding decrease in nitrates when biogenic emissions are inserted in the model runs.

To conclude, the cumulative effect of windblown dust, sea-salt and biogenic emissions was shown to be up to 3 μg/m^3 mainly over coastal areas of Europe on an annual basis. Thus, natural sources have a significant impact on PM levels and composition increasing particle pollution burden over Europe indicating how important is the study of these sources in order to improve air quality simulations.

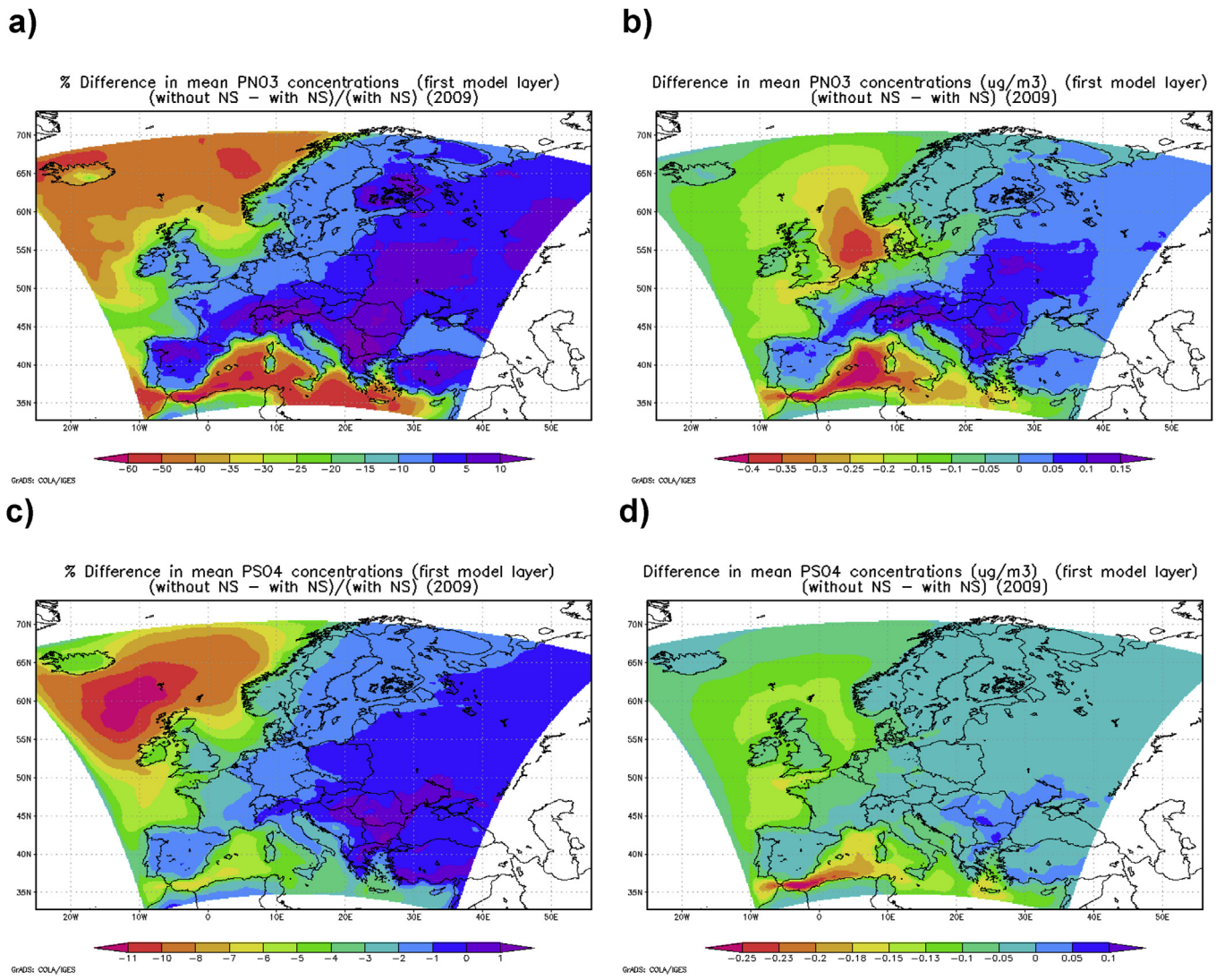

Fig. 11. Mean annual percentage contribution of natural sources (NS) and impact of natural sources on nitrates (PNO3) (a, b) and sulphates (PSO4) (c, d) concentrations (in % and μg/m^3) in 2009.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2016.04.040.

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