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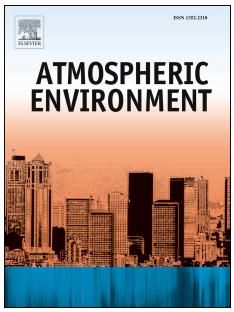

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Analysis of Meteorology-Chemistry Interactions During Air Pollution Episodes Using Online Coupled Models Within AQMEII Phase-2

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Abstract

This study reviews the top ranked meteorology and chemistry interactions in online coupled models recommended by an experts' survey conducted in COST Action EuMetChem and examines the sensitivity of those interactions during two pollution episodes: the Russian forest fires 25 Jul -15 Aug 2010 and a Saharan dust transport event from 1 Oct -31 Oct 2010 as a part of the AQMEII phase-2 exercise. Three WRF-Chem model simulations were performed for the forest fire case for a baseline without any aerosol feedback on meteorology, a simulation with aerosol direct effects only and a simulation including both direct and indirect effects. For the dust case study, eight WRF-Chem and one WRF-CMAQ simulations were selected from the set of simulations conducted in the framework of AQMEII. Of these two simulations considered no feedbacks, two included direct effects only and five simulations included both direct and indirect effects. The results from both episodes demonstrate that it is important to include the meteorology and chemistry interactions in online-coupled models. Model evaluations using routine observations collected in AQMEII phase-2 and observations from a station in Moscow show that for the fire case the simulation including only aerosol direct effects has better performance than the simulations with no aerosol feedbacks or including both direct and indirect effects. The normalized mean biases are significantly reduced by 10-20% for PM10 when including aerosol direct effects. The analysis for the dust case confirms that models perform better when including aerosol direct effects, but worse when including both aerosol direct and indirect effects, which suggests that the representation of aerosol indirect effects needs to be improved in the model.

Key words: aerosol direct effects, feedback, PM10, fire and dust

Highlights

- The interactions among aerosols, radiation, temperature and gas-phase chemistry during the Russian forest fire and Saharan dust episodes were examined.
- Eight WRF-Chem and one WRF-CMAQ simulations performed in context of AQMEII phase-2.
- For both episodes, the simulations including aerosol direct effects only have better performance than the simulations with no aerosol feedbacks or including both direct and indirect effects.

1. Introduction

Air quality modelling systems include both a meteorological model (MetM) and a chemistry transport model (CTM). There are many interactions between meteorology and chemistry in the atmosphere but they are often poorly understood and represented in models. Such interactions include aerosol-cloud-radiation feedbacks (Zhang 2008, Zhang et al., 2010 and Forkel et al., 2012) and interactions between temperature, gas-phase chemistry and aerosols (Baklanov et al., 2014). These interactions are complex and often form chains and loops between a number of meteorological and chemical components. How well they are represented in a model directly influences model performance and the ability of the model to replicate observations.

In order to simulate pollutant concentrations in the ambient atmosphere, MetMs and CTMs can be implemented either 'offline' or 'online'. Offline modelling implies that the CTM is run after the meteorological simulation is completed, while online modelling allows coupling and integration of some of the physical and chemical components to various degrees. Historically, MetMs and CTMs were developed separately and so most air quality modelling systems belong to the 'offline' category (e.g., LOTOS-EUROS: Schaap et al., 2008; MM5-CAMx: http://www.mmm.ucar.edu/mm5 and http://www.camx.com; WRF-CMAQ: Byun and Schere, 2006, San José et al, 2013, Skamarock et al., 2008 and EMEP: Simpson et al., 2012). An 'offline' system cannot take account of chemistry feedbacks on meteorology (e.g., gas and aerosol direct and indirect effects on radiative forcing). Supported by the dramatic increase in computer power in recent years, online coupled mesoscale meteorology and atmospheric chemistry models have undergone a rapid evolution. A number of new generations of online-coupled models have been developed worldwide, such as GATOR- MMTD (Jacobson, 1996, 1997a, b); MM5-MAQSIP (Mathur, et al., 1998), MCCM (Grell et al., 2000), Enviro-HIRLAM (Chenevez et al., 2004, Baklanov et al., 2008, Korsholm et al., 2008), WRF-Chem (Grell et al., 2005), GEM-AQ (Kaminski et al., 2007), GEM-MACH (Moran et al., 2010), WRF-CMAQ v5.0 (Mathur et al., 2010) and COSMO-ART (Vogel et al., 2009). A comprehensive overview of online coupled models has been given by Baklanov et al. (2014). Although the total CPU time required to run the online coupled models are not too different from running them in sequential meteorology followed by CTM simulations (traditional offline mode), the online mode has not been widely used in operational applications of NWP and regulatory use (Grell and Baklanov, 2011). Perhaps what has prevented this was the inadequate demonstration of the benefits for online coupled model applications (e.g., Does the weather forecast improve by including aerosol radiative effects? Are policy inferences derived from online vs offline systems different?).

The COST Action ES1004 - European framework for online integrated air quality and meteorology modelling (EuMetChem; http://eumetchem.info/) - is focusing on online integrated

CTMs and MetMs with two-way interactions between different atmospheric processes including chemistry, clouds, radiation, boundary layer processes, emissions, meteorology and climate. In collaboration with the COST ES1004, recent work carried out in Phase-2 of the Air Quality Model Evaluation International Initiative (AQMEII) (e.g., Galmarini et al., 2014 this issue, Im et al., 2014a this issue and 2014b this issue) focused on online coupled model evaluations. Sixteen modelling groups from Europe and North America have participated in this model evaluation exercise, running eight different online-coupled air quality models. The ENSEMBLE system of the Joint Research Centre (JRC), Ispra, provided the central database and facilities for collecting model output and observation data to support the quantitative analysis of the interactions between meteorology and chemistry.

Despite a growing number of studies of meteorology and chemistry feedbacks employing online coupled models, it is still not well known which meteorology and chemistry interactions are the most important to consider and how well they are implemented in current model systems. For example, the fifth Assessment Report (AR5) of IPCC (2013) has highlighted that "*climate models now include more cloud and aerosol processes, and their interactions, than at the time of the AR4, but there remains low confidence in the representation and quantification of these processes in models*". To address this gap in knowledge, an expert survey, based on expert judgement, has been conducted as part of COST Action ES1004 EuMetChem, to identify which coupling processes are thought to be most relevant for regional air quality and weather predictions and how well these coupling processes are represented in the current models.

The interactions between meteorology and chemistry can be particularly significant during strong air pollution episodes such as wild fire or dust events (Konovalov et al., 2011; Chen et al., 2014; Wong et al., 2012). For example, unprecedented hot and dry weather in summer 2010 caused intensive forest and peat bog fires over the vast territory of Central Russia. This very high aerosol concentration significantly changed the atmospheric gas composition, optical and radiative characteristics of aerosol, and as a result, solar irradiance at the atmosphere, which in turn imposed feedback effects on regional conditions of the climate system (Konovalov et al., 2011 and Chubarova et al., 2012). Makar et al., 2014a this issue and 2014b this issue show that the correlation coefficients between modelled meteorological variables from simulations without and with feedback significantly decreased during the Russian forest fire period. He found improvements in annual temperature when going from the no-feedback simulation to the direct-effect only simulation for each of the European subdomains examined in their analysis, which indicates the relevance of including feedback during these situations and concluded that the implementation of feedbacks has the potential to improve meteorological forecasts. In the events of Saharan dust, the high aerosol loading from mineral dust also interacts with climate and ecosystems and influences the atmosphere – Earth system radiative balance and decreases the photolysis rates of gases (Shao et al., 2011). Recent studies indicate that considering radiative feedbacks has the potential to improve the quality of weather predictions during strong Saharan dust events (Pérez et al., 2006; Bangert et al., 2012).

In this study, we will examine some of the top ranked interactions recommended by the expert survey (e.g., aerosol direct effects on radiation and temperature). As described above the coupling processes between meteorology and chemistry are more significant during strong pollution episodes. Two episodes have been selected from the 2010 AQMEII phase-2 annual runs for detailed analysis with a particular focus on meteorology- chemistry interactions: (a) the Russian forest fires from 25 Jul -15 Aug and (b) the period 1 Oct -31 Oct with significant Saharan dust transport towards Europe. So far most of the AQMEII phase-2 studies have been based on annual and domain averages in order to assess the overall model performance. To understand the role

and importance of the interactions between meteorology and chemistry and their impact on air pollution concentrations, this study undertakes detailed analysis of the two episodes which also provides an opportunity to examine online model performance during pollution episodes.

As reviewed by Baklanov et al. (2014), direct impacts of meteorology on chemistry or vice versa as well as feedback processes are complex, thus a simple classification is insufficient to describe the full range of two-way interactions between meteorological and chemical processes in the atmosphere. Some of the interactions cannot easily be switched on/off in the models (such as the effect of changes in wind speed on dust and sea salt emissions). Therefore, it is not possible to fully assess all the interactions. Of course, some interactions are important, but may not be well represented in the models. Therefore including the coupling processes does not necessarily lead to improved model performance. The scope of this paper is thus not to improve the representation of coupling processes directly, but to provide insight into the importance of the interactions between meteorology and chemistry for simulating air quality during air pollution episodes.

2. Data and Methodology

2.1 Descriptions of the Models

The Weather Research and Forecast (WRF; http://www.wrf-model.org/) community model coupled with Chemistry (WRF-Chem; Grell et al., 2005: Fast et al., 2006) provides the capability to simulate chemistry and aerosols from cloud scales to regional scales. In WRF-Chem, the chemistry model has been developed to be consistent with the WRF model I/O Applications Program Interface (I/O API).

An online model, WRF-Chem includes the treatment of the aerosol direct and indirect effects. Standard gas phase chemistry options of WRF-Chem include the RADM2 and the CBMZ mechanism, additional chemistry options are available with a pre-processing tool based on KPP (Kinetic Pre-Processors). For the aerosols, it offers the choice between bulk, modal, and sectional schemes. The Volatile Basis Set (VBS) approach is also available for the modal and sectional aerosol approaches to treat secondary organic aerosol (SOA) formation. The first and second aerosol indirect effects are implemented in WRF-Chem through a tight coupling of the aerosol module to the Cloud Condensation Nuclei (CCN) and cloud droplets of at least one of the microphysics and radiation schemes (Gustafson et al., 2007). Among other options MEGAN may be used for biogenic emissions and two pre-processors are available for wildfires (injection heights are being calculated online). Recent studies such as Grell et al. (2011), Forkel et al. (2012) and Zhang et al. (2010) have demonstrated that the WRF-Chem model can realistically account for a range of feedback mechanisms between simulated aerosol concentrations and meteorological variables.

In addition to WRF-Chem, the WRF-CMAQ simulation was selected for the dust case study as the WRF model is common to both systems. In the case of WRF-CMAQ, the CTM is the Community Multiscale Air Quality (CMAQ) (Byun and Schere, 2006) developed by the United States Environmental Protection Agency (EPA) EPA. The new version CMAQ 5.0 (officially released in February 2012, http://www.cmaq-model.org/) includes an option to run the model in a 2-way coupled mode with the WRF v3.3 model (Pleim et al., 2008; Mathur et al., 2010; Wong et al., 2012). A coupler is used to link these two models, ensuring exchange between the meteorology and atmospheric chemistry modelling components. In this two-way coupled system, simulated aerosol composition and size distribution are used to estimate the optical properties of

aerosols, which are then used in the radiation calculations in WRF. Based on the definitions from Baklanov et al. (2014), WRF-Chem is categorised as an 'online integrated model' and WRF-CMAQ as an 'online access model'. The use of WRF-Chem and WRF-CMAQ provides a useful comparison of both approaches to meteorological and chemical coupling.

2.2 Model Simulations

Seven WRF-Chem and one WRF-CMAQ groups in Europe participated in AQMEII phase-2 and have completed nine annual simulations (SI2, SI1, DE4, AT1, ES1, IT2, IT1, ES3 and UK5). The model configurations are shown in Table 1. With exception of the ES1 simulation using the Lin et al. (1983) cloud microphysics, identical physics options were chosen while the chemistry options were varied: Morrison double-moment cloud microphysics (Morrison et al., 2008), Rapid Radiative Transfer Method for Global (RRTMG) long-wave and short-wave radiation scheme (Iacono et al. 2008), Yonsei University (YSU) PBL scheme (Hong et al., 2006), NOAH land-surface model (Chen and Dudhia, 2001) and Grell 3D ensemble cumulus parameterization scheme (Grell and Devenyi, 2002) with radiative feedback.

Among these nine simulations, SI2 and IT1 were baseline cases without any aerosol feedbacks, SI1 and UK5 included aerosol direct effects only, while all the other simulations (DE4, AT1, ES1, IT2 and ES3) included both aerosol direct and indirect effects but using different aerosol schemes or gas phase chemistry. The first six simulations listed in Table 1 are using RADM2 gas phase chemistry (Stockwell et al., 1990) and the MADE/SORGAM aerosol module (Ackermann et al., 1998, Schell et al., 2001) and the remaining four cases with different chemistry options and aerosol/cloud modules.

For the Russian forest fire study, three additional WRF-Chem simulations were conducted for both the fire period (25 Jul – 15 Aug) and a non-fire period (16 Aug – 31 Aug):

- UK5a (no aerosol feedbacks using the same configuration as SI2),
- UK5b (direct effects only using the same configuration as SI1)
- UK5c (including both direct and indirect effects using a similar configuration as DE4. Different from the simulation DE4, the original RADM2 gas phase chemistry solver instead of the modified solver that had been applied for simulation DE4 (Forkel et al., 2014 this issue) was used for simulation UK5c in order to be consistent with UK5a/SI2 and UK5b/SI1. The modified RADM2 solver, which had been applied for the DE4 simulation in order to improve an under-representation of ozone titration in areas with high NO emissions is described in Forkel et al. (2014 this issue).

As UK5a and UK5b configurations are identical except UK5b includes the aerosol direct effects, therefore UK5b – UK5a can be used to quantify aerosol direct effects. UK5c includes additional aerosol cloud interactions and aerosol indirect radiative effects, thus UK5c – UK5a can be used to quantify combined aerosol direct and indirect effects.

All model simulations were performed for a large domain covering Europe [25°N, 70°N; 30°W, 60°E] which includes western Russia and northern Africa for the two selected episodes. The same data sets of anthropogenic emissions provided by the TNO (Netherlands Organization for Applied Scientific Research) (Kuenen et al., 2014) and of fire emissions provided by the Finnish Meteorological Institute (FMI) (http://is4fires.fmi.fi/) were used for all the simulations. 3-D daily chemical boundary conditions were provided by the ECMWF IFS-MOZART model run in the context of the MACC-II project (Monitoring Atmospheric Composition and Climate – Interim Implementation) on 3-hourly and 1.125˚ spatial resolution (Inness et al, 2013). An assessment of

the quality of these boundary conditions is provided by Giordano et al. (2014 this issue). According to the common simulation strategy for AQMEII phase-2, the fire and non-fire periods were simulated as a sequence of 2-day time slices with consistent meteorological spin-up files were provided within the AQMEII WRF-Chem groups.

A web-based model comparison system called ENSEMBLE (http://ensemble2.jrc.ec.europa.eu/public/) was used to compare the model output and observations in a standardized format. This system allows temporal and spatial analyses of individual models as well as their ensemble (e.g., Bianconi et al., 2004; Galmarini et al., 2012). For the Saharan dust period, existing model data were taken from ENSEMBLE for all eight WRF-Chem simulations and one WRF-CMAQ simulation listed in Table 1.

2.3 Observation Data

Measurements data used in this study (e.g., PM and ozone) were also extracted from the ENSEMBLE system. Data in the EU domain are obtained from EMEP (European Monitoring and Evaluation Programme, http://www.emep.int/) and AirBase (European AQ database; http://acm.eionet.europa.eu/databases/airbase/). The ENSEMBLE tool is able to extract the matched model and measurements data for specific time windows. For the Russian forest fire study, the selected time window is 25 Jul – 15 Aug 2010 and for the dust period it is 1 Oct – 31 Oct 2010. Rural and urban stations are analysed separately.

Unfortunately, there is no data available in the ENSEMBLE system from Russian stations since neither EMEP nor AirBase contain PM data from Russia. Although attempts were made to access State Environmental Institution "Mosecomonitoring" (www.mosecom.ru) data, it was only possible to use data from one station at 55.70˚ N, 37.51˚ E, which was provided by the Moscow State University. Data was extracted from the nearest WRF-Chem model grid cell from all the model outputs and matched in time (UTC+4) with the Russian station data. Given the coarse model resolution (23 km by 23 km), the point station data may not be directly representative of the nearest grid cell.

2.4 Statistical Analysis

All the observation data extracted from the ENSEMBLE system were spatially averaged (with data availability greater than 75%) in order to examine the temporal response of the model simulations to the extreme pollution episodes. In order to assess the individual model performances, the following statistical parameters were calculated: mean, standard deviation (stdev), correlation coefficient (r), mean bias error (MBE), root mean squared error (RMSE) and normalized mean bias (NMB) together with time series plots. Any missing data were removed before calculating these statistical parameters.

2.5 COST Expert Poll Survey

As an initial exercise within the COST Action ES1004, an expert survey was conducted in order to get an expert judgement on which coupling processes might be most relevant and how well these coupling processes were represented in current online coupled models. The survey questionnaire included 24 meteorology-chemistry interactions of potential importance for the three main application areas of online-coupled models: numerical weather prediction (NWP), chemical weather forecasting (CWF) and climate modelling. The survey questionnaire was sent to different experts in these communities in Europe and beyond, and the results of its analyses

were based on 30 responses. Although the survey results could be considered to be somewhat subjective, it still provided a valuable guidance to the community. The top six ranked important interactions for each of these three application domains are published in Baklanov et al. (2014). As some interactions were selected as important for multiple categories, a new list (see Table 2) was produced to remove duplicates and to merge all the top ranked interactions into one list for general model applications. The final 12 interactions were chosen, because the experts consider them to be the most important, yet at the same time, poorly represented in the current online coupled models. The present study mainly examines the following interactions: 'aerosol -> radiation', 'temperature -> chemical reaction rates and photolysis' and 'radiation -> chemical reaction rates and photolysis' as well the loops and chains formed from those coupling processes.

3. Results and Discussion

3.1 Russian Forest Fire Case Study

The first case study looked at the Russian forest fire episode. As several aerosol direct and indirect effects were ranked among the most important interactions in the COST expert survey (see Table 2), we focus primarily on the aerosol effects in this case study.

Model simulations were performed for both the fire period (25 Jul 25 – 15 Aug 2010) and a non-fire control period (16 Aug – 31 Aug 2010). The weather conditions during the fire period were mainly dry and particularly hot, with light winds. Figure 1 shows WRF-Chem simulated mean surface PM10 in $\mu g/m^{-3}$ and surface ozone in ppbv for both the fire period and the non-fire period for the baseline case without aerosol feedbacks (UK5a). In this severe air pollution episode, very high surface PM10 concentrations of 40-150 $\mu g/m^{-3}$ averaged over the fire period were found near Moscow (Figure 1a) in contrast to much lower concentrations of 2.5 – 10 $\mu g/m^{-3}$ for the non-fire period (Figure 1b). Ozone concentration (Figure 1c) in that region reached 40 - 60 ppbv during the fire period but was only 10-20 ppbv in the post-fire period (Figure 1d).

The impact of this high aerosol loading on other meteorology and chemistry variables is illustrated in Figure 2. The aerosol direct effects (UK5b – UK5a) in Figure 2 (left panels) show that downward shortwave radiation at the surface was significantly reduced by up to 100 W m^{-2} over the Russian fire regions (Figure 2a), which caused a reduction in 2-m temperature by 1-2 K (Figure 2c) and PBL height was reduced by 200 – 300 m (Figure 2e). Note that the effect of heat release due to the fires was not included in this sensitivity study. Reduced radiation can lead to less NO_2 photolysis and reduced temperature lower photochemical activity, thus both effects reduced ozone formation over the fire region (Figure 2g). In Figure 2 right panels (UK5c – UK5a) the combined aerosol direct and indirect effects during the fire period show that the north-eastern part of the EU domain (fire region) was dominated by aerosol direct effects during the fire period. Due to little cloud cover and simulated cloud droplet number densities that were of the same order of magnitude than the assumed number of 250 cm^{-3} which is used in WRF the absence aerosol cloud interactions, aerosol indirect effects on solar radiation were not significant in the fire region (Figure 2a). This also holds for temperature and PBL height, whereas precipitation was reduced in the fire region for UK5c as compared to UK5a and UK5b (not shown). Indirect effects on solar radiation were much stronger over the north Atlantic and British Isles than in the fire region due to the higher cloud cover there and also due to simulated cloud droplet concentrations that were much smaller than WRF's assumed default value.

Evaluation using observation data extracted from ENSEMBLE (domain averaged) in Figure 3 and Table 3 show that UK5b (aerosol direct effects only) has better performance for PM10 simulations for both rural and urban sites and mean bias error (MBE) is about 3$\mu g/m^{-3}$ (~20%)

smaller for rural sites and 2.5μg/m^{-3} (~10%) smaller for urban sites compared to UK5a and UK5c. UK5c including both aerosol direct and indirect effects had the best correlation coefficients (r = 0.75), but slightly larger MBE and RMSE. In all cases, these models underestimated PM10, particularly for the urban sites, which are a general feature for most of the model simulations in AQMEII phase-2 (Im et al., 2014) and other relevant studies (e.g., Stern et al., 2008). The smaller bias for UK5b can be explained by the decrease in PBL heights when the direct aerosol effect is considered, which result in higher near surface aerosol concentrations. Since scavenging of aerosol particles is higher in WRF-Chem when aerosol cloud interactions are considered explicitly (case UK5c) than for the cases without explicit aerosol cloud interactions (case UK5a and UK5b), the enhanced scavenging of aerosol particles compensates the increase due to the lower PBL height for case UK5c.

There was no significant difference between the three WRF-Chem simulations for ozone (see Figures 3c and 3d) for the fire region. The statistics for ozone evaluations in Table 3 were rather similar, again UK5b showing the smallest MBE, RMSE and UK5c showing a slightly higher correlation. As shown in Figure 2g and 2h, the impact of aerosol direct/indirect effects on ozone was rather small except within the fire region. Therefore, the evaluation using ENSEMBLE over the whole EU domain was not sufficient to investigate the interactions between meteorology and chemistry due to the fires.

Additional model evaluations were conducted using one Moscow station data for surface PM10, 2 m temperature and surface ozone for both the fire period and the non-fire period (see Figure 4 and Table 4). Statistics in Table 4 shows that the errors were much larger at this station comparing to the averaging statistics in Table 3 for the whole domain. It is obvious that average statistics over the large domain are likely to mask any local differences. However, due to only one available station data for the fire region, it is difficult to quantify the significance level sensibly in this study.

Due to too many missing records in the observed data, ozone statistics for the non-fire period was not produced. Results showed that in general all three model cases had better performance for the non-fire period compared with the fire period. All the model cases significantly underestimated PM10 by about 35 – 40 μg/m^{-3} (~35%) on average during the fire period. The underestimation could partly result from an underestimation of PM emissions by the FMI fire inventory. In addition, hotspots in the measurements data were absent in the model simulations probably due to their coarse resolution.

UK5b shows the smallest MBE and RMSE (see Table 4), which confirmed that it is important to include aerosol direct effects for the Russian fire episode as the feedbacks of high aerosol loading on meteorology and chemistry had been accounted. When aerosol direct effects (UK5b) or both direct and indirect effects (UK5c) were included, 2 m temperature was further reduced by 0.5 K compared to the baseline case (UK5a). Although the correlation slightly improved, the biases for 2 m temperature were not reduced by the inclusion of aerosol effects (UK5b and UK5c). However, as only one Moscow station was used, it is difficult to know the representativeness of this station compared with the model grid.

Again there was no significant difference between the three cases for the ozone simulations. Due to the complexity of the feedback of aerosol effects on ozone and limited measurements data, the magnitude of the aerosol effects on ozone predictions cannot be generalized to be not important based on the limited analysis presented here. For instance, aerosol radiative effects could impact ozone predictions in two opposing ways in certain situations: attenuation of photolysis and lower temperatures could reduce the chemical production; on the other hand, reduced mixing arising from the cooling could in fact increase concentrations within the boundary layer and lead to

higher ozone (Jacobson et al., 1996, Baklanov et al., 2014, Mathur et al., 2010 and Wong et al., 2012). The presence of scattering or absorbing aerosols is likely to result in different effects on photolysis and the modulation of clouds could further impact ozone predictions. It should be also taken into account the aerosol cloud interactions are less relevant in this context as high ozone is more related to dry and sunny conditions.

3.2 Saharan Dust Case Study

The second case study considered a Saharan dust episode that occurred during 1 Oct - 31 Oct 2010. In addition to the aerosol direct and indirect effects, we also investigated the interactions between wind speed and dust as it was ranked as one of the most important interactions in the COST ES1004 expert poll (see Table 2).

Figure 5 presents WRF-Chem simulated monthly mean surface PM10, changes of surface downward shortwave flux due to dust (SI1 – SI2) and 10m wind speed for the Saharan dust period. The results show that dust mainly remained in the north of Africa (PM10 reached 50-100 $\mu g/m^{-3}$ monthly averaged), which could cause a 15 Wm^{-2} reduction of downward shortwave radiation at the surface (a relatively small impact compared to the Russian fire case in Figure 2a). The dust was spread out to some parts of the Mediterranean and North Atlantic due to strong south-east winds (monthly mean wind speed over the dust affected area was about 4-5 m/s in Figure 5c and dominant wind direction was between 90-180˚ not shown). In Figure 6, hourly model data at a hotspot in North Africa (29.5 N, 20.75 E) show that the higher surface PM10 were coincident with higher wind speed (r=0.75) and the wind rose plot shows the period was dominated by strong south-eastly wind. This may be partly explained by windblown dust emissions increasing with wind speed and did transport to some part of the European area. As all the WRF-Chem models in Table 1 use the same meteorological configurations, sensitivity to changes in wind fields between the different model simulations was not possible. Therefore, only overall model performance on PM10 was examined in the following analysis.

Figure 7 and Table 5 show observed and simulated surface PM10 concentration during the dust period over the whole EU domain (306 stations averaged for rural sites and 764 stations averaged for urban sites), including all the WRF-Chem / WRF-CMAQ simulations listed in Table 1. The results showed that simulations without any aerosol feedbacks (IT1, SI2) and with aerosol direct effects included only (SI1) had better performance ($r > 0.8$ and MBE ≈ -5.0 for rural sites; $r > 0.6$ and MBE ≈ -15.0 for urban sites) than other simulations that included both aerosol direct and indirect effects (DE4, AT1, ES1, IT2, ES3; $r < 0.8$ and MBE ≈ [-8, -14] for rural sites; $r < 0.8$ and MBE ≈ [-18,-24] for urban sites). The underestimations of PM10 concentrations were more pronounced for the urban sites (~50% or more) than for the rural sites (~25% or more) for all the simulations. This can be attributed to uncertainties in primary PM10 anthropogenic emissions for urban areas (Stern et al., 2008). The higher bias in the IT2 run compared to the other runs may be explained by an excess of dry deposition (Im et al., 2014b). It should also be highlighted that the IT2 run was performed with an experimental version of 3.4 WRF-Chem, where the module for SOA production (SOA-VBS) was coupled with cloud microphysics. As a consequence, the bias of the IT2 simulation should not be considered to be general bias of WRF-Chem, but only of this particular version which is still under development. The online access model WRF-CMAQ (UK5) had the best correlation and captured temporal variations well, but underestimated PM10 concentration constantly. The reason for UK5 underestimation could be partly due to the fact that UK5 did not consider windblown dust emissions but only the dust from the boundary (Im et al., 2014b this issue). Compared with the AQMEII phase-2 PM10 annual evaluations presented in Im et al. (2014b this issue), similar trend on the model performance were found in this study for a

specific episode, such as SI1 and IT1 have smallest biases and UK5 has the highest correlation coefficients.

A further analysis concentrated on a smaller domain more strongly affected by the dust transport in the southern parts of the domain ([30°N – 45°N; 25°W - 50°E]; 75 stations averaged for rural sites and 256 stations averaged for urban sites). Results in Figure 8 and Table 6 show that a similar trend was found in the smaller domain as in the whole EU domain. For the rural sites, all the simulations performed slightly better over the southern domain compared to the whole EU domain. However, bias in the model performance increased for the urban sites over the smaller dust domain, while the magnitude of PM predicted by the models for rural locations appears to be similar with that of urban locations, where in contrast measurement data indicate an urban enhancement. There were significant underestimations (more than 60%), particularly for the models that included both aerosol direct and indirect effects, which may indicate that emissions or aerosol feedbacks in the WRF-Chem/WRF-CMAQ models were not well represented in urban areas. In particular the urban increment is missing (not fully resolved due to too poor resolution and not urbanised version of WRF used in these simulations) in regional scale models. On the other hand, it is difficult to attribute such underestimations solely to the inclusion of online radiative feedbacks, given that other known effects connected to the structural and processes model attributes could cause discrepancies of comparable magnitude. Such attributes could include grid resolution, the treatment of sub-grid effects of turbulence, urban canopy and heat islands, fine-scale emission distributions, as well as the representativeness of urban monitoring sites. In that sense, the long-range dust transport event under this study does not appear to be adequately constrained for assessing model performance at urban sites. Nevertheless, one can reasonably expect that future studies of PM episodes with a stronger urban component could help illustrate the potential benefits of the online treatment of radiative feedbacks in the urban scale (Baklanov et al., 2014).

4. Conclusions

This study compared several model simulations with different feedback/process-interactions and examined the interactions among aerosols, radiation, temperature and gas-phase chemistry during the Russian forest fire and Saharan dust episodes based on eight WRF-Chem and one WRF-CMAQ simulations in context of AQMEII phase-2.

The results indicated that it is important to include interactions between meteorology and chemistry (especially aerosols and ozone) in the online coupled models. For example, the Russian forest fire case study has shown significant aerosol direct effects on meteorology (and loop back on chemistry). High levels of PM10 over the Moscow area caused significantly reduced downward short wave radiation and surface temperature, and also reduced PBL height. These in turn reduced the photolysis rate of NO_2 and slowed down photochemical O_3 production. The aerosol indirect effects were found relatively small over the fire region due to lack of clouds in the simulated episodes. Model evaluation using AQMEII phase-2 data and Moscow station data showed that UK5b (included aerosol direct effects) performed better and reduced NMB by 10 -20 % for PM10 compared to UK5a (no feedbacks) and UK5c (including both direct and indirect effects) for the fire period. Although the aerosol indirect effects on solar radiation were much stronger over the north Atlantic and British Isles regions, this study could not examine it further due to limited data and resources. In fact, given the large uncertainties (and challenges) in model representation of the timing, placement and extent of clouds (even when the models are constrained with observations in data assimilation), the challenges in assessing indirect effects are

enormous. It is also supporting the survey conclusion that the indirect aerosol effects are still poorly parameterised and need to be further developed and improved.

The dust case study also showed that the aerosol direct effects on radiative forcing are significant. Evaluation using AQMEII data showed that the WRF-Chem simulations with direct effects (SI1) or no feedback (IT1 and SI2) performed better than those simulations including both direct and indirect effects (DE4, AT1, ES1, IT2 and ES3). This suggests that the representation of aerosol indirect effects needs to be improved in online coupled models, in particular in the WRF-Chem model. Further study should select a period with significant aerosol indirect effects (e.g., cloudy days) in order to examine aerosol indirect effects and feedbacks to meteorology by different online models.

There still remains low confidence in the representation and quantification of these meteorology and chemistry coupling processes in current online models. Due to the complexity of the physical and chemical processes and high cost of computing time, more collaborative work is needed between the science community and model developers to improve the representation of these coupling processes.

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Table 1 AQMEII phase2 WRF-Chem/WRF-CMAQ model configuration

Model code in ensemble	**(UK5a)/SI2**	**(UK5b)/SI1**	**(UK5c)**	**DE4**	**AT1**	**ES1**	**IT2**	**IT1**	**ES3**	**UK5**
Version	3.4.1	3.4.1	3.4.1	3.4.1	3.4.1	3.4.1	3.5	3.4.1	3.4.1	3.4.1
Microphysics	Morrison[a]	Morrison	Morrison	Morrison	Morrison	Lin[b]	Morrison	Morrison	Morrison	Morrison
Gas phase chem.	RADM2[c]	RADM2	RADM2	RADM2 modified	RADM2	RADM2	RACM[d]	CBMZ[e]	CBMZ	CB-V-TU[m]
Inorg. aerosol	MADE[f]	MADE	MADE	MADE	MADE	MADE	MADE	MOSAIC[g] 4 bins	MOSAIC 4 bins	AERO6
Org. aerosol	SORGAM[h]	SORGAM	SORGAM	SORGAM	SORGAM	SORGAM	VBS[i]	-	-	Carlton et al., 2010
Grid scale wet deposition	Simple	Simple	Easter04	Easter04	Easter04	Easter04	Easter04	Simple	Easter04	Simple
Conv. Wet. dep	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
Grid scale aq. chem.	-	-	WT86	WT86[k]	FP01[l]	FP01	WT86	-	FP01	WT86
Conv. aq. chem	WT86	WT86	WT86	WT86	WT86	WT86	WT86	-	-	WT86
Aero Direct Effect	No	Yes	Yes	Yes	Yes	Yes	Yes	No	Yes	Yes
Aero IndirectEffect	No	No	Yes	Yes	Yes	Yes	Yes	No	Yes	No
Chem_opt*	2	2	41	41	11	11	43	7	9	-

Chem_opt:
=2: includes chemistry using the RADM2 chemical mechanism and MADE/SORGAM aerosols;
=7: CBMZ chemical mechanism without DMS; CBMZ chemical mechanism; MOSAIC using 4 sectional aerosol bins;
=9: CBMZ chemical mechanism without DMS; CBMZ chemical mechanism; MOSAIC using 4 sectional aerosol bins including some aqueous reactions;
=11: RADM2 chemical mechanism and MADE/SORGAM aerosols including some aqueous reactions;
=41: RADM2/SORGAM with aqueous reactions included;
=43: NOAA/ESRL RACM Chemistry and MADE/VBS aerosols using KPP library. The volatility basis set (VBS) is used for Secondary Organic Aerosols.

Reference for each scheme: [a]Morrison et al. 2008, [b]Lin et al 1983, [c]Stockwell et al.,1990, [d]Stockwell et al.1997, [e]Zaveri & Peters 1999, [f]Ackermann et al. 1998, [g]Zaveri et al.2008, [h]Schell et al., 2001; [i]Ahmadov et al, 2012, [k]Walcek & Taylor 1986, [l]Fahey & Pandis 2001and [m]Whitten et al., 2010. UK5a, UK5b and UK5c are additional runs for the Russian fire case study.

Table 2 The top ranked important interactions based on COST expert survey

	Meteorology and chemistry interactions: Cause/Effect of … on (->) …
1	aerosol -> precipitation (initiation, intensity)
2	aerosols -> radiation (shortwave scattering/absorption and longwave absorption)
3	temperature vertical gradients -> vertical diffusion
4	aerosol -> cloud droplet or crystal number density and hence cloud optical depth
5	aerosol -> haze
6	aerosol -> cloud morphology (e.g., reflectance)
7	wind speed -> dust and sea salt emissions
8	precipitation (frequency/intensity) -> atmospheric composition
9	temperature -> chemical reaction rates and photolysis
10	radiation -> chemical reaction rates and photolysis
11	liquid water -> wet scavenging and atmospheric composition
12	radiatively active gases -> radiation

Table 3 Statistics of observed and simulated daily surface PM10 and hourly surface ozone (EU domain averaged) for both rural (left) and urban (right) over the forest fire period (22 days in total).

model	mean	stdev	r	MBE	RMSE	NMB (%)	mean	stdev	r	MBE	RMSE	NMB (%)
	PM10 ($\mu g/m^{-3}$) rural (291 stations averaged)						PM10 ($\mu g/m^{-3}$) urban (595 stations averaged)					
obs	15.24	2.60	-	-	-	-	21.80	2.71	-	-	-	-
UK5a	12.24	2.18	0.69	-3.00	3.54	-19.7	11.30	1.75	0.63	-10.50	10.70	-48.2
UK5b	15.19	2.24	0.68	-0.04	1.92	-0.3	13.87	1.73	0.61	-7.93	8.20	-36.4
UK5c	11.70	2.32	0.75	-3.53	3.93	-23.2	10.59	1.77	0.76	-11.21	11.35	-51.4
	Ozone (ppbv) rural (473 rural stations averaged)						Ozone (ppbv) urban (472 urban stations averaged)					
obs	33.24	6.37	-	-	-	-	28.81	7.78	-	-	-	-
UK5a	34.01	4.66	0.78	0.77	4.07	2.3	33.27	4.93	0.74	4.46	6.92	15.5
UK5b	33.89	4.65	0.78	0.65	4.03	2.0	33.18	4.93	0.74	4.37	6.84	15.2
UK5c	34.06	4.63	0.79	0.82	4.03	2.5	33.27	4.95	0.75	4.47	6.85	15.5

Table 4 Statistics of observed and simulated surface PM10, 2m temperature and surface ozone at a Moscow station for both fire period (left) and non-fire period (right). Due to too many missing records in observed data, ozone for non-fire period is not produced.

model	mean	stdev	r	MBE	RMSE	NMB (%)	mean	stdev	r	MBE	RMSE	NMB (%)
	PM10 ($\mu g/m^{-3}$) fire period: N=409						PM10 ($\mu g/m^{-3}$) non-fire period: N=382					
obs	103.5	115.58	-	-	-	-	20.92	24.89	-	-	-	-
UK5a	66.28	80.49	0.46	-37.22	112.6	-36.0	5.16	6.37	0.64	-15.75	26.53	-75.3
UK5b	67.68	76.56	0.5	-35.82	107.6	-34.6	6.48	6.37	0.59	-14.43	26.11	-69.0
UK5c	63.24	63.57	0.46	-40.25	110.6	-38. 9	5	6.79	0.64	-15.92	26.48	-76.1
	2m temperature (˚C) fire period: N=524						2m temperature (˚C) non-fire period: N=384					
obs	28.7	4.26	-	-	-	-	16.58	5.68	-	-	-	-
UK5a	26.28	5.01	0.81	-2.42	3.81	-8.4	14.4	5.27	0.87	-2.18	3.53	-13.2
UK5b	25.8	4.73	0.84	-2.9	3.9	-10.1	14.43	5.22	0.88	-2.14	3.43	-12.9
UK5c	25.78	4.74	0.84	-2.92	3.92	-10.2	14.62	5.1	0.88	-1.95	3.36	-11.8
	Ozone (ppbv) fire period: N=406											
obs	25.27	28.14	-	-	-	-						
UK5a	67.16	30.87	0.51	41.89	51.16	165.8						
UK5b	67.17	29.02	0.51	41.89	50.56	165.8						
UK5c	67.5	29.01	0.52	42.22	50.72	167.1						

Table 5 Statistics of observed and simulated surface PM10 during Saharan dust period (1 Oct – 31 Oct 2010) for all nine models listed in Table 1 over EU domain.

model	mean	stdev	r	MBE ($\mu g/m^{-3}$)	RMSE ($\mu g/m^{-3}$)	NMB (%)	Direct effects	Indirect effects
Averaged 306 rural stations over EU domain								
obs	20.33	4.48	-	-	-	-	-	-
SI2	15.32	2.99	0.81	-5.01	5.67	-24.6	No	No
SI1	15.36	3.01	0.81	-4.96	5.64	-24.4	Yes	No
DE4	12.65	2.21	0.68	-7.68	8.37	-37.8	Yes	Yes
AT1	12.05	1.7	0.73	-8.28	8.94	-40.7	Yes	Yes
ES1	11.4	1.72	0.79	-8.92	9.49	-43.9	Yes	Yes
IT2	6.32	0.8	0.49	-14	14.58	-68.9	Yes	Yes
IT1	15.83	3.21	0.89	-4.49	4.98	-22.1	No	No
ES3	11.57	2.54	0.74	-8.75	9.27	-43.0	Yes	Yes
UK5	10.11	2.51	0.92	-10.22	10.48	-50.3	Yes	No
Averaged 764 urban stations over EU domain								
obs	32.2	5.2	-	-	-	-	-	-
SI2	16.59	3.08	0.68	-15.61	16.07	-48.5	No	No
SI1	16.62	3.06	0.67	-15.58	16.04	-48.4	Yes	No
DE4	13.83	1.97	0.57	-18.38	18.88	-57.1	Yes	Yes
AT1	13.18	1.81	0.64	-19.02	19.49	-59.1	Yes	Yes
ES1	12.57	1.86	0.68	-19.64	20.06	-61.0	Yes	Yes
IT2	7.42	1.11	0.45	-24.78	25.23	-77.0	Yes	Yes
IT1	17.17	3.1	0.8	-15.03	15.38	-46.7	No	No
ES3	11.83	2.15	0.66	-20.37	20.77	-63.3	Yes	Yes
UK5	11.56	2.31	0.86	-20.64	20.91	-64.1	Yes	No

Table 6 Statistics of observed and simulated surface PM10 during Saharan dust period (1 Oct – 31 Oct 2010) for all nine models listed in Table 1 over southern EU dust domain.

model	mean	stdev	r	MBE	RMSE	NMB	Direct effects	Indirect effects
obs	18.03	4.48	-	-	-	-	-	-
SI2	14.22	4.00	0.52	-3.81	5.58	-21.1	No	No
SI1	14.44	3.97	0.52	-3.60	5.45	-20.0	Yes	No
DE4	12.87	3.00	0.67	-5.16	6.11	-28.6	Yes	Yes
AT1	11.33	2.64	0.68	-6.70	7.46	-37.2	Yes	Yes
ES1	10.96	2.65	0.65	-7.08	7.83	-39.3	Yes	Yes
IT2	6.77	1.42	0.34	-11.26	12.00	-62.5	Yes	Yes
IT1	14.84	3.49	0.65	-3.20	4.66	-17.7	No	No
ES3	10.06	2.37	0.80	-7.97	8.48	-44.2	Yes	Yes
UK5	9.14	2.17	0.76	-8.89	9.42	-49.3	Yes	No
obs	36.95	6.16	-	-	-	-	-	-
SI2	17.32	4.36	0.21	-19.63	20.72	-53.1	No	No
SI1	17.56	4.28	0.21	-19.39	20.48	-52.5	Yes	No
DE4	14.74	2.99	0.29	-22.22	22.99	-60.1	Yes	Yes
AT1	13.40	2.87	0.35	-23.55	24.23	-63.7	Yes	Yes
ES1	13.00	3.05	0.33	-23.95	24.64	-64.8	Yes	Yes
IT2	8.91	2.22	0.33	-28.05	28.62	-75.9	Yes	Yes
IT1	17.21	4.42	0.37	-19.74	20.63	-53.4	No	No
ES3	10.54	1.71	0.51	-26.42	26.96	-71.5	Yes	Yes
UK5	11.32	1.74	0.42	-25.63	26.22	-69.4	Yes	No

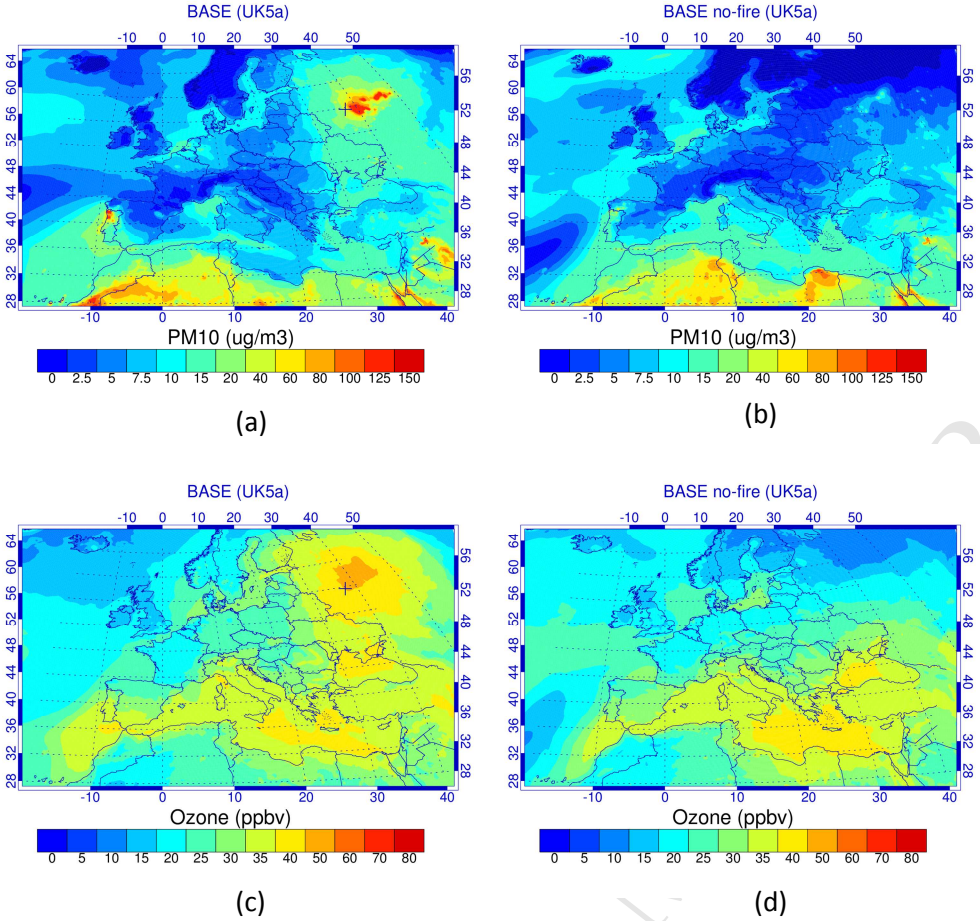


Figure 1 WRF-Chem simulated mean surface PM10 (top) in μg/m^{-3} and surface ozone (bottom) in ppbv for the forest fire period (left; 25 Jul – 15 Aug 2010) and non-fire period (right; 16 Aug – 31 Aug 2010) for the baseline case without aerosol feedbacks (UK5a). The '+' symbol marks the location of the Moscow station.

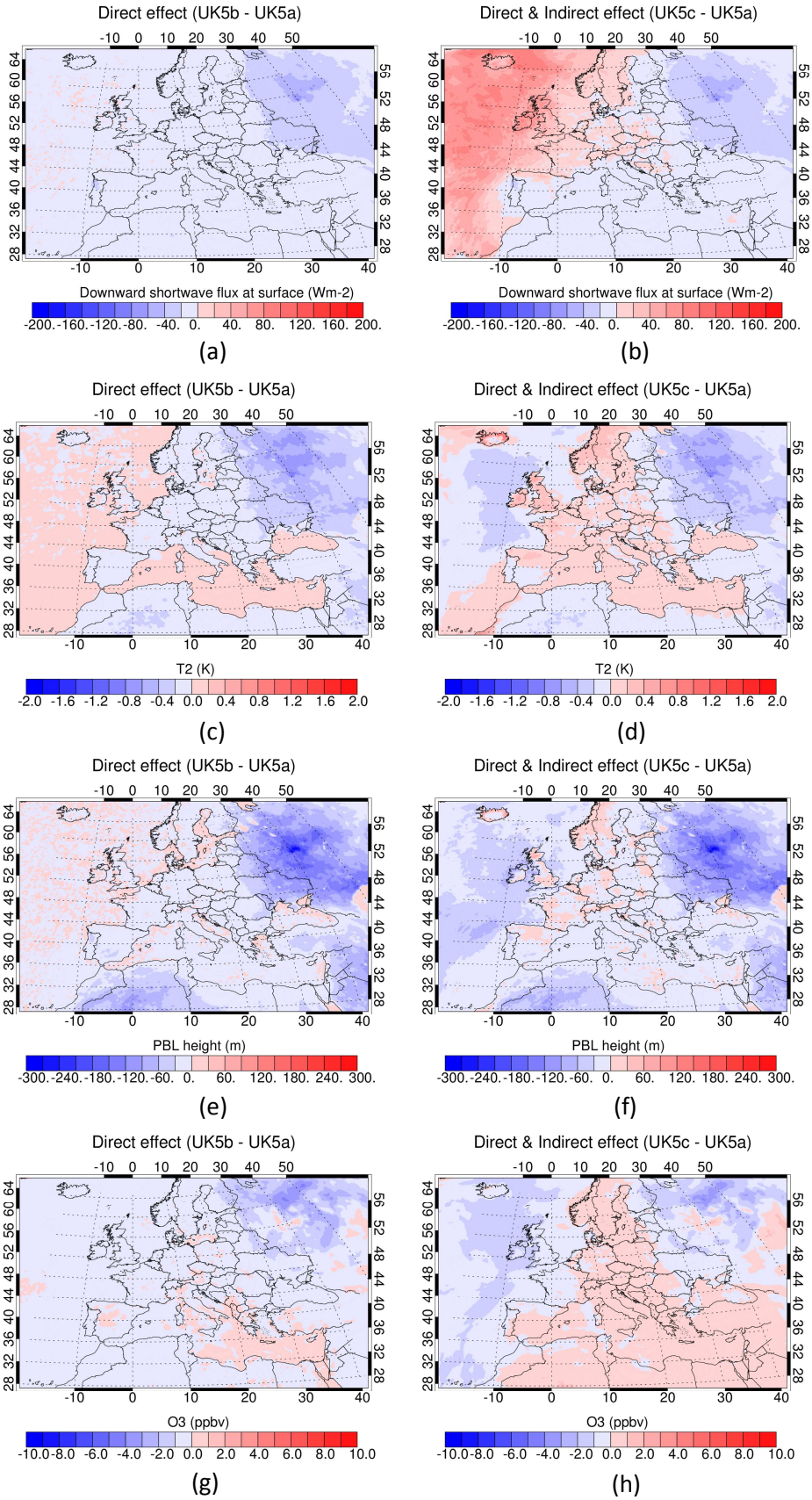


Figure 2 WRF-Chem simulated mean changes due to aerosol direct effect (UK5b – UK5a; left panels) and both direct & indirect effect (UK5c – UK5a; right panels) during the fire period (25 Jul – 15 Aug 2010) for downward shortwave flux at surface in W m^{-2} (a & b), 2 m temperature in K (c & d), PBL height in meters (e & f) and surface ozone in ppbv (g & h).

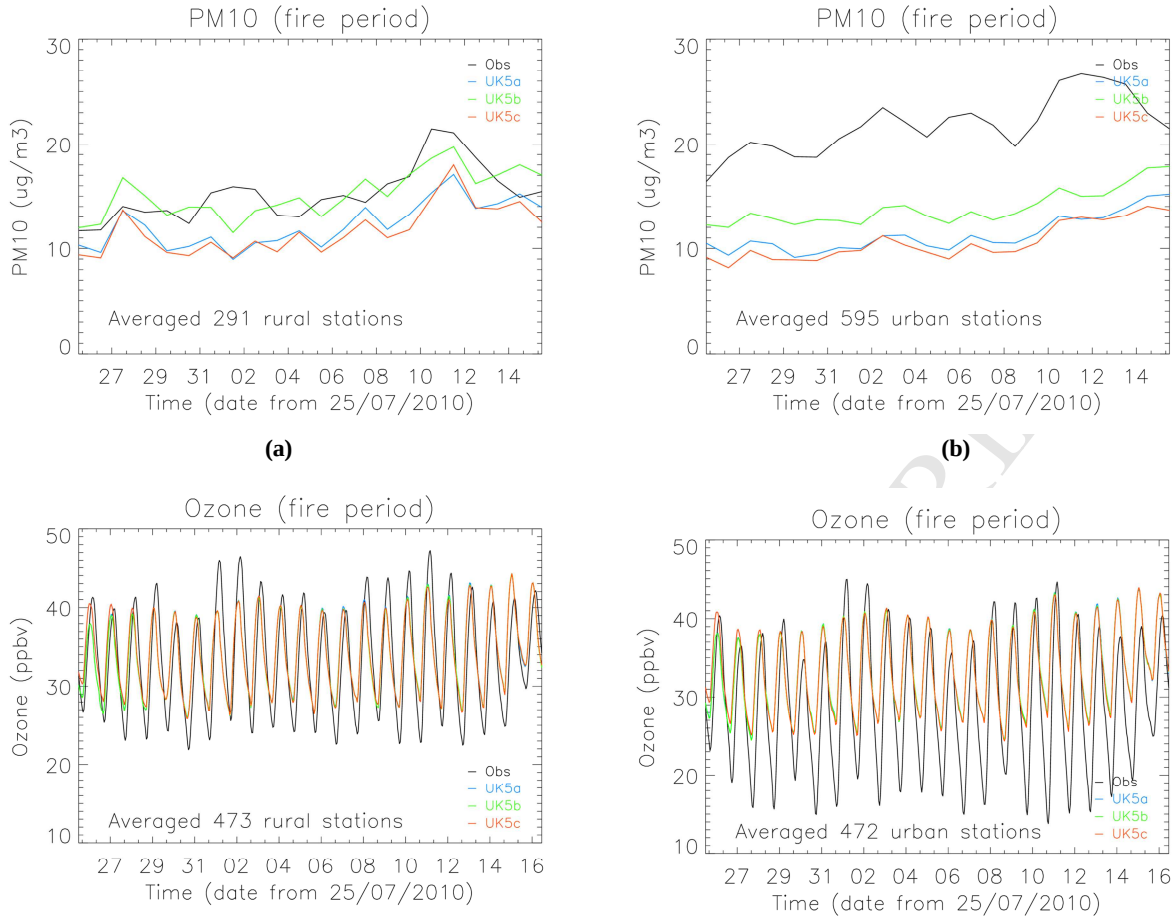


Figure 3 Observed and simulated surface PM10 (top panels) and surface ozone (bottom panels) for rural (left) and urban (right) respectively during the fire period (EU domain averaged)

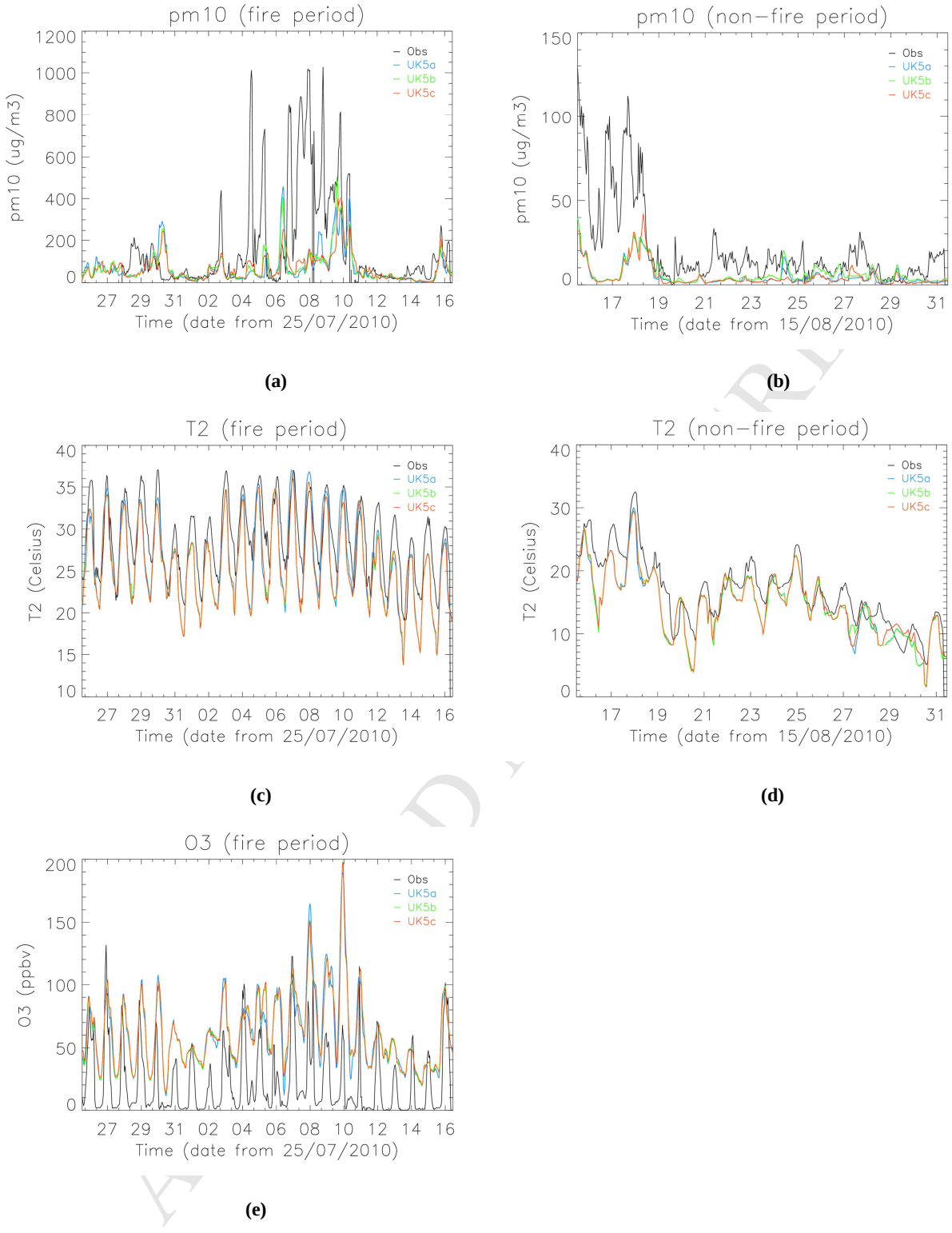


Figure 4 Observed and simulated surface PM10 (top), 2m temperature (middle) and surface ozone (bottom) at a Moscow station for both fire period (left) and non-fire period (right). Due to too many missing records in the observed data, ozone for non-fire period is not produced.

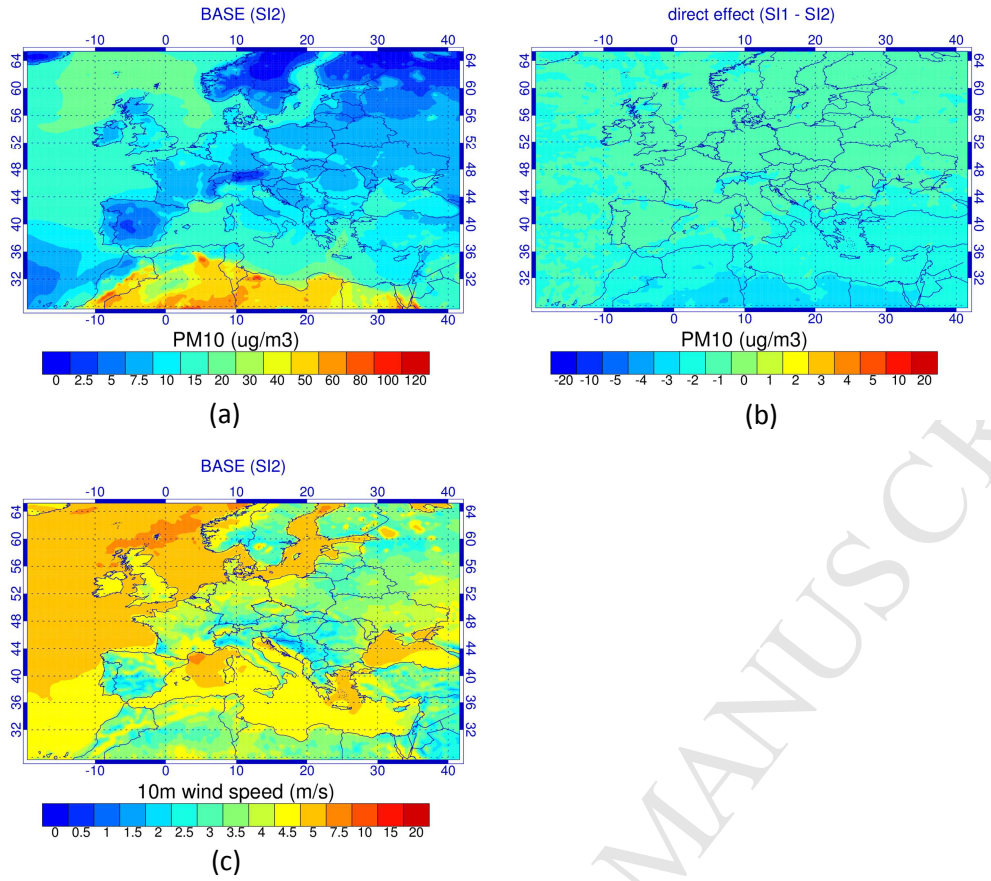
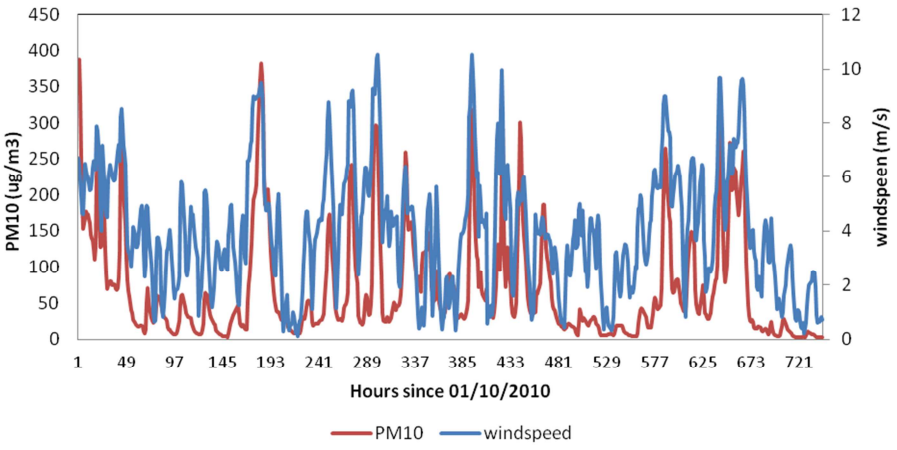
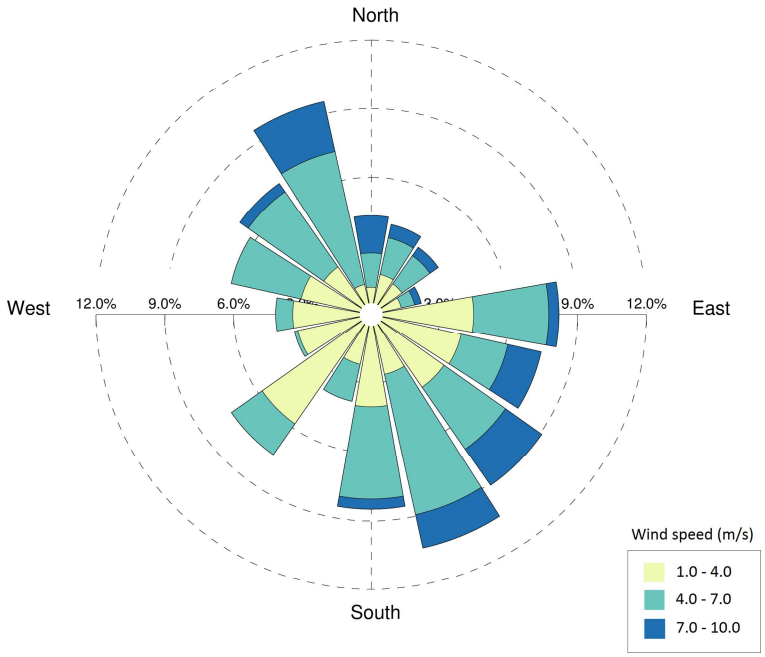


Figure 5 WRF-Chem simulated monthly mean (a) surface PM10 in μg/m^{-3}, (b) changes of downward shortwave flux at surface in Wm^{-2} due to aerosol direct effect, (c) 10m wind speed in m/s.



(a)



(b)

Figure 6 WRF-Chem simulated hourly surface PM10 and 10m wind speed (a) and wind rose (b) at a hotspot in North Africa (29.5 N, 20.75 E).

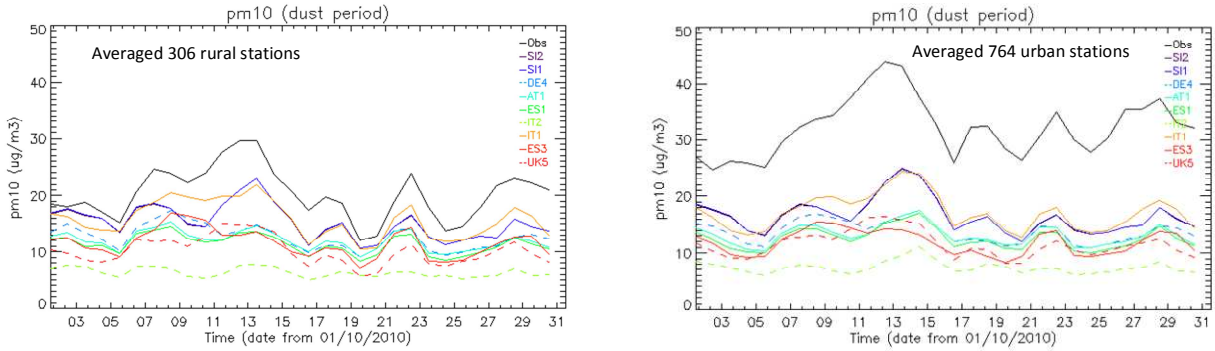


Figure 7 Observed and simulated surface PM10 for rural (left; 306 stations) and urban (right; 764 stations) during the dust period 1 Oct – 31 Oct 2010 (EU domain averaged)

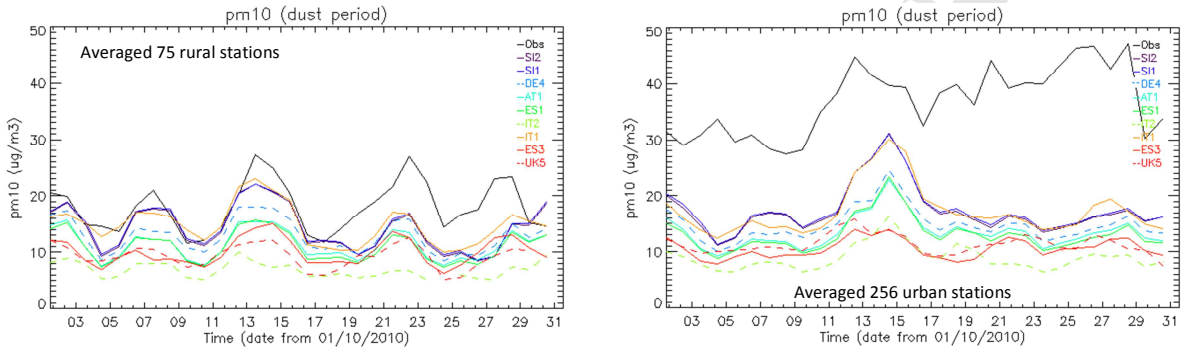


Figure 8 Observed and simulated surface PM10 for rural (left; 75 stations) and urban (right; 256 stations) during the dust period 1 Oct – 31 Oct 2010 (southern EU dust domain averaged)