Climatological simulations of ozone and atmospheric aerosols in the Greater Cairo region

A. L. Steiner¹*, A. B. Tawfik¹,6, A. Shalaby², A. S. Zakey², M. M. Abdel-Wahab³, Z. Salah², F. Solmon⁴, S. Sillman¹, R. A. Zaveri⁵

¹Department of Atmospheric, Oceanic and Space Sciences, University of Michigan, Ann Arbor, Michigan 48109-2143, USA
²Egyptian Meteorological Authority, PO Box 11784, Cairo, Egypt
³Department of Meteorology, Cairo University, PO Box 12613, Cairo, Egypt
⁴Earth System Physics Group, International Centre for Theoretical Physics, 34151 Trieste, Italy
⁵Atmospheric Sciences and Global Change Division, Pacific Northwest National Laboratory, Richland, Washington 99352, USA
⁶Present address: Center for Ocean-Land-Atmosphere Studies, Calverton, Maryland 20705-3106, USA

ABSTRACT: An integrated chemistry–climate model (RegCM4-CHEM) simulates present-day climate, ozone and tropospheric aerosols over Egypt with a focus on northern Africa and the Greater Cairo (GC) region. The densely populated GC region is known for its severe air quality issues driven by high levels of anthropogenic pollution in conjunction with natural sources such as dust, and agricultural burning events. We find that current global emission inventories underestimate key pollutants such as nitrogen oxides and anthropogenic aerosol species. In the GC region, average ground-based observations of the daily July maximum nitrogen dioxide (NO₂) are 40 to 60 parts per billion by volume (ppbv) and are about 10 ppbv higher than modeled estimates, likely due to model grid cell resolution, improper boundary layer representation, and poor emissions inventories. Observed July daily maximum ozone concentrations range from 30 ppbv (winter) to 90 ppbv (summer). The model reproduces the seasonal cycle fairly well, but modeled July ozone is underestimated by approximately 10 ppbv and exhibits little interannual variability. For aerosols, springtime dust events dominate the seasonal aerosol cycle. The chemistry–climate model captures the springtime peak aerosol optical depth (AOD) of 0.7 to 1 but is slightly greater than satellite-derived AOD. Observed AOD decreases in the summer and increases again in the fall due to agricultural burning events in the Nile Delta; however, the model underestimates this observed AOD peak in fall, as standard emissions inventories underestimate the extent of this burning and the resulting aerosol emissions. Our comparison of modeled gas and particulate phase atmospheric chemistry in the GC region indicates that improved emissions inventories of mobile sources and other anthropogenic activities, specifically NOₓ and organic aerosols, are needed to improve air quality simulations in this region.

KEY WORDS: Ozone · Mediterranean · Urban · Air quality

1. INTRODUCTION

The Greater Cairo (GC) area in Egypt (population ~15 million) is the largest city in northern Africa and one of the 15 largest cities in the world (Demographia 2014). Urbanization and industrialization have increased rapidly over the past 50 yr, causing increased levels of air pollutants and numerous air quality exceedances (Robaa & Hafez 2002, Robaa 2003, Zakey et al. 2008). This population-dense urban area has high anthropogenic emissions from transportation, industry, and waste burning common to global megacities (Molina & Molina 2004), and is further subject to seasonal enhancements of atmospheric aerosols.
from wind-blown dust during the spring and agricultural burning during the fall (El-Askary et al. 2009, El-Metwally et al. 2010, Marey et al. 2011). While individual studies have investigated air quality at specific locations in northern Africa, regional modeling that includes atmospheric chemistry is still needed in the region. In this study, we implemented a coupled chemistry–climate model to simulate concentrations of tropospheric ozone and atmospheric aerosols over a decadal time period in the GC region, with the goal of evaluating the model performance in northern Africa and identifying emissions and model shortcomings for future work.

While air quality is a known public health issue in Cairo, knowledge of the issue in this region is limited and few efforts have been made to model the regional emission sources and confounding climate factors that drive gas and aerosol chemistry. One of the factors limiting the development of a comprehensive air quality plan is the relative paucity of ground-based observations of air pollutants in the GC region. Limited air quality observations by the Egyptian Environmental Affairs Agency (EEAA) and the Egyptian Meteorological Authority (EMA) are available at select locations; however, these data have yet to be synthesized to provide a broader picture of the chemical environment in GC. Other observations of atmospheric constituents have been conducted by the Ministry of Health (Nasralla 1996) but these are currently unavailable to researchers for model evaluation. Surface ozone (O3) over Egypt has been explored in a few studies with observational climatology based on the EEAA monitoring sites (Khoder 2002, 2009, El-Hussainy et al. 2003). These observations confirm a typical diurnal cycle of O3 concentrations with midday maxima as observed at other urban areas, as well as spring or summer seasonal maxima depending on location.

Aerosol characterization and quantification in the GC region has received more interest in the last decade. Two recent field studies, the Cairo Air Improvement Project (CAIP) (Abu-Allaban et al. 2002) and the Cairo Aerosol Characterization Experiment (CACHE) (Alfaro & Wahab 2006), were conducted to specifically evaluate aerosols in Cairo. Results from the CAIP study focused on criteria pollutants, such as total fine particulate matter (PM$_{2.5}$) and coarse particulate matter (PM$_{10}$), while the CACHE study used aerosol filter samples at one site from 2004 to 2006 to investigate aerosol composition (Favez et al. 2008a, 2008b, Mahmoud et al. 2008). Results from the CACHE study highlight the contribution of biomass burning aerosol to organic carbon and the role of secondary organic aerosol (SOA) formation from insoluble species (Favez et al. 2008b). However, the results of these 2 studies have not been comprehensively integrated into atmospheric chemistry modeling to understand the climatic and seasonal factors that drive air quality in the region.

Other recent studies have circumvented the lack of ground-based observations using satellite-derived aerosol indices to investigate the seasonality of aerosols over Cairo (El-Metwally et al. 2010, Marey et al. 2010), and find that there are 2 distinct peaks in the aerosol seasonal cycle related to the spring dust storms and the fall agricultural burning of rice straw (Marey et al. 2011). The fall burning season and resulting poor air quality in Cairo is known colloquially as the ‘black cloud’ and, because of the intense air and health effects, has received much more attention recently (Mahmoud et al. 2008, Marey et al. 2010, 2011).

Fewer modeling studies exist on the air quality and climate of the GC region. Global atmospheric chemistry models suggest elevated summer surface ozone concentrations in the Middle East (Lelieveld et al. 2002), and recent studies have attributed this to a combination of local O$_3$ production, the transport of O$_3$ aloft due to the Asian monsoon circulation system, and the low-level transport of European pollution (Lelieveld et al. 2002, Duncan et al. 2008, Joiner et al. 2009, Liu et al. 2009, 2011). However, the local O$_3$ contribution is not explicitly examined in these studies as they are of coarse resolution and cannot account for the many uncertainties associated with local O$_3$ production. While these results suggest a Middle Eastern hotspot of tropospheric ozone that may be important from a climate forcing perspective, the lack of ground-based and vertical profile observations in the region makes it difficult to confirm and quantify. Further, few high-resolution regional modeling studies address this question exist.

The GC region is within a climate change hotspot region. Studies suggest that the Middle East, North African and Mediterranean regions are extremely sensitive to climate change compared to other regions in the world due to combined effects of heat and drought stress on human and natural systems (Giorgi 2006). Changes in temperature and the water cycle may also affect the formation of secondary pollutants such as O$_3$ and aerosols, which will likely affect air quality in the region. This highlights the important role of atmospheric chemical composition on the climate in the region and the urgent need for comprehensive atmospheric modeling studies that incorporate both dynamics and chemistry.
This study implemented a chemistry–climate model (Shalaby et al. 2012) that couples atmospheric chemistry with atmospheric dynamics, and simultaneously solves for transport and chemistry. This creates a 2-way interaction between the chemistry and dynamics of the atmosphere, and allows for the investigation of the complex interactions between atmospheric chemistry and climate in the region. The goals of this study were (1) to conduct simulations over a poorly understood geographic region from the atmospheric chemistry perspective using a high-resolution, fully coupled chemistry–climate model, and (2) to evaluate the model to the furthest extent possible given the limited ground-based and satellite-derived observational data.

2. MODEL DESCRIPTION AND APPLICATION

2.1. RegCM4-Chem

The RegCM4-CHEM is an online, coupled regional chemistry–climate model described in Shalaby et al. (2012). The core atmospheric model dynamics are based on the International Center for Theoretical Physics (ICTP) Regional Climate Model version 4.2 (Giorgi et al. 2012) with integrated atmospheric chemistry that is calculated at a specified chemistry model time step. For the simulations presented here, we implement the Community Land Model version 3.5 (CLM3.5) land surface model (Tawfik & Steiner 2011), the Grell convection scheme for convective precipitation (Grell 1993) and the SUBEX large-scale scheme for large-scale precipitation (Pal et al. 2000).

This version of RegCM-CHEM employs a condensed version of the Carbon Bond Mechanism (CBM-Z; Zaveri & Peters 1999) with a fast, radical-balance based solver (Sillman 1991, Barth et al. 2002) for gas-phase chemistry. For atmospheric aerosols, it utilizes a simplified aerosol model for the simulation of anthropogenic and biomass burning aerosols (Solmon et al. 2006) and an online, interactive dust scheme (Zakey et al. 2006). The anthropogenic aerosol scheme includes 3 primary aerosol species, including sulfate, black carbon (BC) and organic carbon (OC). Sulfate is emitted in the gas phase as SO2, and converted to sulfate within the model by gas and aqueous reactions (Qian et al. 2001). BC and OC are derived from anthropogenic activities or biomass burning, and are allowed to age from hydrophobic to hydrophilic species in the atmosphere after emission with a timescale of 1.14 d (Solmon et al. 2006). Dust is emitted based on meteorological conditions, soil type and soil texture and includes the transport and evolution of 4 size bins (Zakey et al. 2006). The model does not currently include the formation of SOA or the treatment of more complex aerosol thermodynamics such as ammonium or nitrate aerosol, which likely leads to an underestimation of the total aerosol mass. The inclusion of these aerosol types is currently under development.

The model domain (Fig. 1) has a 60 km resolution with 160 grid cells in the zonal direction spanning from approximately 20° W to 50° E and 80 grid cells in the meridional direction, spanning from approximately 5° to 40° N with 18 vertical levels. While the hydrostatic dynamical core can be run at finer resolutions, the 60 km simulations presented here were developed to include all major emission sources in the region and capture the necessary large-scale dynamics. Specifically, domain selection was designed to capture the transport of European emissions across the Mediterranean to the GC region, as well as the large-scale dynamical features such as the West African monsoon and eastern Africa circulation necessary to simulate the northern African climate.

Model simulations are conducted for an 11 yr time period (1996–2006) with a 2 yr model spin up (1996–1997) that is discarded to allow for the equilibration of soil moisture in the region. The final 9 yr (1998–2006) of the simulation are presented in this analysis. Meteorological boundary conditions were provided at 6 h intervals from the European Centre for Medium-Range Weather Forecasts (ECMWF) Interim data (ERA-Interim) (Dee et al. 2011). Monthly climatology of chemical boundary conditions are provided by the MOZART model at a 2.8° × 2.8° (T42) resolution (Emmons et al. 2012).

2.2. Emissions

Emissions for RegCM4-CHEM were developed specifically with 2 features in mind: (1) to implement global emissions inventories to allow flexibility in domain selection over multiple regions, and (2) to implement the highest resolution data available for each species required by the RegCM4-CHEM chemical mechanism (Shalaby et al. 2012). In this study, we include emissions inventories from the following sources: Monitoring Atmospheric Composition and Climate/CityZen inventory (MACCity) (Lamarque et al. 2010), Reanalysis of the Tropospheric Chemical Composition (RETCO) (Schultz et al. 2005), Emission Database for Global Atmospheric Research (EDGAR) (Olivier et al. 2005, van Aardenne et al. 2005), and
Precursors of Ozone and their Effects in the Troposphere (POET) (Granier et al. 2005). Because each emissions inventory provides a unique set of chemical species at varying resolutions, an emissions preprocessor was designed (Shalaby et al. 2012) to bilinearly interpolate each user-defined inventory from their native grids to the model grid. The particular inventories used for each species are shown in
Table 1. The POET, EDGAR, and RETRO inventories are only available through the year 2000, therefore simulated emissions after this time use the monthly emissions from 2000 for the remainder of the simulation. The pre-processing of emissions results in a blend of emission data sources implemented in these simulations, as described below. Emissions for the broader domain, including the Mediterranean region, the Middle East, and northern Africa are shown in Fig. 1, and depict the regional emissions that may affect transport into the GC region. Localized emissions in the GC region are shown in Fig. 2.

2.2.1. Anthropogenic emissions

Anthropogenic emissions are based on the MACCity inventory, which spans the period 1990–2010, providing interannual variations in emissions during this simulation period. The MACCity inventory (Lamarque et al. 2010) is based on a historical emissions dataset developed to support the Intergovernmental Panel for Climate Change (IPCC) fifth assessment report (AR5) for the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP). It is available globally at 0.5° resolution and provides information on emissions of alkanes, alkenes, aldehydes, alcohols, carbon monoxide (CO), SO₂, nitrogen oxides (NOₓ), BC and OC. In our simulations, emissions for most species are based on monthly anthropogenic emissions and do not have prescribed diurnal emissions cycles. The one exception is that of nitric oxide (NO) emissions, where a bimodal diurnal cycle is imposed on the monthly MACCity NO emissions to replicate 2 typically observed rush hour traffic peaks while conserving the total monthly mass flux of NOₓ to the atmosphere.

Climatological anthropogenic NOₓ emissions over the model domain for the summer, i.e. in June, July and August (JJA), during the simulation years 1998 to 2006 are high in industrialized regions in Europe and the Middle East, with perceptible ship tracks across the Mediterranean and Red Sea (Fig. 1A). Other emissions hotspots for NOₓ are urban centers in Saudi Arabia, the Mediterranean coast of the Middle East, and Southern Europe. Over the Egyptian Nile Delta, anthropogenic NOₓ emissions gradually increased by about 15% from 1998 to 2006 (Fig. 2A) with an average winter emissions maximum of 15 mg m⁻² d⁻¹ (Fig. 2B).

Anthropogenic alkanes from this inventory are displayed to indicate the location of anthropogenic volatile organic compound (VOC) emissions, although we note that there are several VOC categories included in the CBM-Z mechanism that are not presented here (see Shalaby et al. [2012] for VOC speci-
For anthropogenic alkanes (Fig. 1B), summer (JJA) emissions are high (12 to 20 mg m\(^{-2}\) d\(^{-1}\)) in the GC region, and there are several other emissions hotspots along the Arabian Coast of the Red Sea and in Gulf cities in the United Arab Emirates, Qatar, and Kuwait. These high emissions of anthropogenic alkanes of up to 100 mg m\(^{-2}\) d\(^{-1}\) are likely linked to petrochemical production; however, ground-based data to evaluate the accuracies of these inventories are lacking. As with anthropogenic NO\(_x\), VOC emissions in GC exhibit clear seasonal cycles with maximum emissions in the winter months (Fig. 2B). Over the decade of the simulated period, anthropogenic alkane emissions as estimated by the MACCity inventory increase by about 30% in Cairo from 1998 to 2006 (Fig. 2A).

The spatial distributions of anthropogenic SO\(_2\) and BC emissions are shown in Fig. 1C,D, indicating the location of regional aerosol emission sources. As with anthropogenic NO\(_x\), SO\(_2\) emissions are largest in major urban areas, and the Egyptian Nile Delta region shows significant SO\(_2\) emissions of up to 24 mg m\(^{-2}\) d\(^{-1}\) that rival other European emissions centers (Fig. 1C). BC emissions are significantly lower, with few anthropogenic sources on the African continent according to the MACCity inventory and a
small emissions source in the Cairo region (Fig. 1D). Recent studies (Doumbia et al. 2012) show that urban areas in western and central Africa have very high concentrations of BC due to emissions from a variety of mobile fuel sources; however these are not included in our current MACCity inventory and may represent a further unrecorded emissions source in Cairo. Overall, anthropogenic BC sources are likely underestimated over the GC region. Seasonal cycles of the aerosol precursor SO$_2$ and primary anthropogenic BC and OC show a slight seasonal cycle with higher emissions in the winter (Fig. 2D), and a ~5% decrease in OC and a ~12% increase in BC over the simulation period (Fig. 2C).

2.2.2. Biogenic volatile organic compounds (VOCs)

Biogenic VOCs are calculated online in the land surface component of the climate model (Shalaby et al. 2012, Tawfik et al. 2012) using the MEGAN emissions model (Guenther et al. 2006), which allows emissions to respond every 10 min to temperature, net surface radiation, and soil moisture as well as to the monthly climatological, satellite-derived leaf area index (LAI). Generally, biogenic isoprene emissions are relatively small in the Nile Delta as compared to the forested regions in the Mediterranean and the strong source in the tropics of Africa (Fig. 1F). Seasonal emissions in the Nile Delta reach up to 0.03 mg C m$^{-2}$ d$^{-1}$ (Fig. 2B), which is relatively small when compared to the anthropogenic emissions in the region. Further, these emissions decrease significantly in magnitude over the course of the simulation (Fig. 2A) due to the effects of soil moisture on biogenic emissions (Guenther et al. 2006), suggesting that the slow drying of model soil water reduces the local emissions in northern Africa. This is an artifact of a soil moisture initialization with soils that are too wet for the region. The remaining biogenic species are provided by monthly average emissions from POET, which includes both terrestrial and ocean sources (Table 1).

2.2.3. Biomass burning

Biomass burning emissions are provided by the RETRO, EDGAR and POET inventories. The RETRO inventory includes many of the main biomass burning species, such as NO$_x$, alkanes, CH$_4$, CO$_2$, CO, a suite of VOC, and BC and OC. Generally, biomass burning as predicted by these inventories is low in JJA and emissions are concentrated in the tropics (Fig. 1E). The contribution of fall burning, i.e. in September, October and November (SON), in the Nile Delta is not present in the existing global inventories, and may be due to the underestimation of crop residue burning in some regions (McCarty 2011). For example, satellite observations have identified fall aerosol events over the Nile Delta (Marey et al. 2010, 2011), but these emissions are not likely present in global emissions inventories. A new, daily fire emissions product based on satellite fire counts (Wiedinmyer et al. 2011) over the region was also tested to see if higher spatial and temporal resolution products improved the burning emissions. We found that the use of this product did not improve the simulation of fire and fall agricultural burning that frequently occurs for this region; this may be due to the timing of the satellite crossover and/or the difficulty of remotely sensing these types of fire events (McCarty 2011). As will be discussed in Section 4.2.4, our simulation likely misses biomass-burning sources due to agricultural burning in the region that cannot be captured by current inventories and satellite products.

2.2.4. Dust emissions

Dust emissions are simulated in an online, interactive dust scheme as defined by Zakey et al. (2006). Emissions are calculated at each time step within RegCM using soil texture, representative soil aggregate distributions, soil humidity, fractional vegetation cover, and threshold frictional winds from CLM3.5. These variables drive a dust emission flux in specified size bins from the fine to coarse mode, and in the simulations presented here we use 4 dust size bins including the fine mode (0.01 to 1.0 µm), the accumulation mode (1.0 to 2.5 µm), the coarse mode (2.5 to 5.0 µm), and a large size mode (5 to 20 µm). Once emitted, dust in each size bin is advected based on prognostic model equations and removed via wet (washout and rainout) and dry deposition. Recent studies indicate that the standard RegCM4 dust emission scheme performs reasonably well over northern Africa and the Mediterranean after new model development on soil representation and emission distribution and the use of a different land surface model (Solmon et al. 2006, 2012, Nabat et al. 2012). These parameterization and sub-model changes result in variations in the dust emissions, as discussed in Section 4.2.3 below.
2.3. Observations

2.3.1. Climate

Climate model temperature and precipitation are evaluated against the 0.5° gridded Climate Research Unit (CRU) surface air temperature and precipitation products (Mitchell & Jones 2005). This data set incorporates observations from multiple ground-based observations over the time period 1900–2010 and is regridded to the climate model grid. We note that the station distribution over northern Africa is one of the sparsest of all regions in the globe, and this may introduce some bias into our model evaluation.

2.3.2. Surface gas-phase observations

Generally, ground-based observations of air quality criteria pollutants and atmospheric aerosols are limited in Egypt and the GC region. Ground-based observations of 2 gas-phase species, O₃ and NO₂, are provided from a suite of observational stations from the Environmental Information and Monitoring Program (EIMP) directed by the EEAA. The EIMP-EEAA measures several trace gases (including PM₁₀, NOₓ, SO₂ and CO) at approximately 42 sites across Egypt, with 14 in the GC area. Here we present observations during the simulation time period for O₃ and NO₂ concentrations from eleven of the EIMP-EEAA sites (locations shown in Fig. 3), noting that not all sites were operational during the entire simulation time period. Generally, observations are collected on an hourly basis at each of these sites throughout the year; however, hourly data is not archived and is unavailable. Mean monthly 24 h maxima of these species are recorded and are used for this analysis. These sites are located in a variety of settings, including urban, residential and industrial. The measurements presented here include a cluster of sites in the center of Cairo, including Abbassaya (urban/residential), Giza CU (residential), Qualaly (urban center; also called El-Kolaly in some reports), Gemhoroya (street canyon; also called El-Gomhorya). Also included are 2 sites to the north, Kaha (background/residential) and Shoubra El-Kheima (industrial/residential), Nasr City (residential) to the west, Fum Khalig (roadside/urban near the Nile shore) and Tabbin (industrial/residential) to the south, and Maadi (residential) and the Sixth of October (residential/urban/industrial) in the southwest portion of the GC area. To understand the behavior of the model over GC and the Nile Delta region and compare with observations, modeled data is averaged over 9 grid boxes (outlined box in Fig. 3) corresponding to the region.

2.3.3. Satellite-derived aerosol optical depth (AOD)

We implement satellite-derived observations of aerosol optical depth (AOD) at 550 nm from the Moderate Resolution Imaging Spectroradiometer (MODIS) on the Terra and Aqua satellites to evaluate the simulated AOD from the RegCM (data available from the NASA Giovanni portal http://disc.sci.gsfc.nasa.gov/giovanni/overview/index.html). Two different AOD values are available from each satellite: the ‘Dark Target’ product and the ‘Deep Blue’ product. The ‘Deep Blue’ AOD product was developed to separate out the radiative effects of aerosols over highly reflective surfaces, such as those that are present in our northern Africa domain. This uses the blue channels of radiation, where the contribution of the surface reflectivity to the signal is relatively low, and is generally thought to provide better retrievals over highly reflective surfaces (Shi et al. 2012). Satellite data is compared to modeled data surrounding the Cairo region (as defined by the outlined region in Fig. 3), and these products are discussed in the model evaluation in Section 4.2.4.
3. Model climatology

Here we provide an evaluation of the ability of RegCM4-CHEM to simulate the region’s temperature and precipitation, which are 2 key climate aspects that can be important for the evaluation of atmospheric chemistry in the region. In December, January and February (DJF), surface air temperatures in northern Africa are from 275 to 285 K (Fig. 4A), and the model under-predicts temperatures in the Sahara by up to 5 K but only by 0.1 K in the GC region (Table 2). These winter cool biases over desert regions are similar to those in other RegCM4 simulations (Giorgi et al. 2012) and have been attributed to the simulation of desert albedo and land-surface parameterizations. This bias is unlikely to affect atmospheric chemistry in the region, as most of the photochemical activity occurs during the summer months. In the summer, the average JJA temperature is between 300 and 310 K (Fig. 4B), with biases ranging from +3 to −3 K depending on the location (Fig. 4D) and a warm bias of about 2 K in the GC region (Table 2). The summer temperature bias has the potential to accelerate atmospheric photochemistry in the region, as discussed in Section 4.1.2.

As expected in northern Africa, precipitation is rare with slight amounts in DJF (Fig. 5). The majority of continental precipitation is focused in the tropics during the JJA rainy season (Fig. 5B). Domain precipitation biases are less than 10 mm mon⁻¹, with the model exhibiting slightly more precipitation than observed over northern Africa in both seasons (Fig. 5C,D). This slight positive precipitation bias found over the Sahara during winter may explain some of the cooler than observed temperatures (Fig. 4C). However, most of the model precipitation bias occurs in the tropics in JJA, with some of central Africa exhibiting large wet biases and the western coastal region exhibiting a dry bias. JJA precipitation in this region is controlled by the placement and strength of the West African Monsoon, and prior RegCM testing in this region shows mean absolute biases of 30 to 50% (Giorgi et al. 2012), consistent with our study. Other RegCM simulations in this region have shown it is particularly sensitive to the land surface, treatment of desert dust and selection of the convection scheme (Davis et al.
Biases in the GC region are small (typically less than 5 mm mo\(^{-1}\)) and range from a 90% dry bias in the summer (due to the small magnitude) and a 10% dry bias in the winter (Table 2). Precipitation biases of this magnitude are typical of many regional climate model simulations (Giorgi et al. 2012). Therefore despite some modeled precipitation uncertainties in the tropics outside our area of interest, overall, the model can reproduce the GC region climatology.

### 4. Atmospheric chemistry: climatology and interannual variability

In this section we evaluate the gas-phase chemistry (Section 4.1) and atmospheric aerosols (Section 4.2) as simulated by the model over the GC region.

#### 4.1. Gas-phase chemistry and ozone formation

4.1.1. Nitrogen oxides and carbon monoxide

Simulated climatological NO\(_x\) concentrations are typically less than 10 parts per billion by volume (ppbv), with larger concentrations in the tropics in DJF due to biomass burning emissions (Fig. 6A). In the GC region, a clear urban NO\(_x\) signal is present of up to 10 ppbv (Fig. 6A), with lower concentrations in the summer (2 ppbv). The main source of NO\(_x\) in the GC region is generated from automobile traffic and fossil fuel burning. Surface observations of the daily maximum NO\(_2\) concentrations from 10 EIMP-EEAA sites in the region show that they range from ~20 to

<table>
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<th>Time period</th>
<th>Surface air temperature bias (K)</th>
<th>Precipitation bias (mm mo(^{-1}))</th>
<th>(%)</th>
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<td>−4.41</td>
<td>−54</td>
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Table 2. Bias in annual and seasonal modeled climatological data for the Greater Cairo region (1998–2006) compared to observed monthly Climatic Research Unit data (0.5° resolution) (Mitchell & Jones 2005)
120 ppbv, with some sites and locations reaching over 250 ppbv (Fig. 6C). Generally, values at most sites range from 20 to 50 ppbv in the summer to ~80 to 120 ppbv in the winter (Fig. 6C), consistent with an earlier single-site analysis (Khoder 2002). Several sites exhibit exceptionally high concentrations. For example, NO$_2$ concentrations of 250 to 400 ppbv are measured at the Giza CU and Maadi sites in the winter of 2004, and of 150 to 220 ppbv at Giza CU, Abbasseya and Sixth of October in the winter of 2001.

Unfortunately, average 24 h monthly maximum NO$_2$ concentrations from the model were not saved, therefore we cannot do a direct comparison of observed and modeled daily maximum NO$_2$ for all years and months. However, an average modeled diurnal cycle of NO$_2$ concentrations for July was retained (Fig. 6D), indicating NO$_2$ maxima in the early morning and late afternoon of 15 to 20 ppbv, decreasing during midday to ~5 ppbv. In comparison, the observed July station-averaged model maximum is 38 ppbv (with a range from 4 to 98 ppbv), a factor of 2 greater compared with modeled values. In prior studies with RegCM4-CHEM, an evaluation of NO$_2$ over Europe showed that the model tended to over-predict observed NO$_2$ concentrations, with relatively low observed concentrations (ranging from 2 to 10 ppbv at remote locations and 5 to 20 ppbv in urban locations) and modeled concentrations 2 to 20 ppbv higher than observed, depending on the region (Shalaby et al. 2012). However in the GC region, observed surface NO$_2$ is much higher than that observed in Europe and are of the same order of magnitude as other extremely polluted regions, such as China (Liu et al. 2010, Xu et al. 2011).

There are several possible reasons for the large measured versus modeled discrepancies in NO$_2$. One is instrument error and the overestimation of observed NO$_2$, as the standard chemiluminescence measurement technique implemented at the Cairo sites is known to overpredict NO$_2$ concentrations.

Fig. 6. Climatological data (1998–2006): modeled mean midday (12:00 to 17:00 h local time) NO$_x$ concentrations for (A) DJF and (B) JJA; (C) observed ground-based mean monthly 24 h maximum NO$_2$ concentrations at stations in the Greater Cairo (GC) region (see Fig. 3 for locations); (D) modeled average July diurnal cycle of NO$_2$ concentrations (black) with the observed station-averaged mean July 24 h maximum NO$_2$ concentration (solid red line) and the range of minimum and maximum observed concentrations (dotted red lines) at all GC stations.
(e.g. Dunlea et al. 2007). However, given the known air quality problems in the GC region, it is plausible that concentrations are this high. Model errors could also drive the underestimation, including the relatively coarse grid resolution (60 km) that may dilute point NO₂ sources within a large grid cell. The large discrepancies between the local-scale and large-scale grid measurements will likely lead to a substantial under-prediction of reactive gases such as NO₂. Additionally, the boundary layer dynamics in RegCM4 may not sufficiently represent the region and lead to low O₃ concentrations in the boundary layer. Because the European study (Shalaby et al. 2012) conducted at a similar model resolution over Europe showed an NO₂ bias opposite to this study, this suggests that emissions inventories may not accurately represent the urban pollutants in this region. Further investigation is needed to fully attribute and address this issue.

We also evaluate CO concentrations at the surface. Modeled CO concentrations are greatest in the winter (DJF; Fig. 7A), with surface concentrations over 350 ppbv in the tropics due to biomass burning and up to 300 ppbv in the GC region (Fig. 7A,C). Concentrations decrease in the summer to ~150 ppbv (Fig. 7B,C). We compare modeled surface CO concentrations with those derived from the MOPITT satellite (Bowman 2006). The model generally captures the seasonal cycle in the GC region, although lacks the interannual variability displayed in the satellite observations (Fig. 7C), despite a slight interannual variability of the emissions (Fig. 2A). Overall, the model tends to over-predict the surface CO concentrations (Fig. 7D) with a weak correlation (R² = 0.36). This result is in contrast to other anthropogenically related emissions such as NO₂, which is underestimated in the GC region (Fig. 6D), suggesting discrepancies in the emission sources.

![Fig. 7. Climatological (1998–2006) modeled mean midday (12:00 to 17:00 h local time) CO concentrations for (A) DJF and (B) JJA; (C) average modeled and MOPITT-derived surface CO concentrations over the Greater Cairo (GC) region; (D) correlation between modeled and MOPITT surface CO versus modeled CO in GC with the regression slope (correlation coefficient)](image-url)
4.1.2. Ozone

Modeled midday mean tropospheric O$_3$ concentrations in DJF are relatively low over the Mediterranean and northern Africa, with daily maximum concentrations from 40 to 50 ppbv (Fig. 8A) and higher concentrations in the tropics (60 to 90 ppbv) due to biomass burning and isoprene sources. In JJA, O$_3$ concentrations increase to 60 to 80 ppbv in the Mediterranean with elevated concentrations over the GC region (Fig. 8B). Monthly averages of daily maximum observations of O$_3$ concentrations at the ground sites in the GC area (locations in Fig. 3) exhibit a seasonal cycle of O$_3$ ranging from 10–30 ppbv in winter to 70–100 ppbv in summer (Fig. 8C).

As with NO$_2$ concentrations, the daily maximum modeled values were not saved, so a direct comparison of monthly mean values is not possible. However, the modeled July average diurnal cycle shows a range of O$_3$ concentrations from 40 ppbv at night to 65 ppbv during the day. The modeled July daily maximum is 10 ppbv lower than the climatological, station-averaged O$_3$ maximum of 75 ppbv (range 47 to 96 ppbv) (Fig. 7D), indicating an overall underestimation of summer O$_3$ maxima in the GC area. Assuming that the main driver of the seasonal amplitude is photochemistry, the model likely underestimates O$_3$ in large part due to the very low modeled NO$_2$ concentrations (Fig. 6D), as local anthropogenic emissions are likely underestimated in the region based on current inventories. Another important factor in the simulation of the urban O$_3$ observations is the relatively coarse model grid cell resolution of 60 km. This resolution will not capture many of the local, air quality responses that are observed at the
stations and likely is an important factor in the underestimate of modeled NO2 and O3.

There is a noticeable interannual variability in the observed O3 surface concentrations, with the years 2003 and 2004 exhibiting relatively low summer maximum O3 concentrations (70 to 80 ppbv) and the year 2006 exhibiting relatively high summer maximum O3 concentrations (up to 105 ppbv at some sites). Generally, the model does not simulate the seasonal amplitude exhibited by the observations. Average modeled concentrations are 40 to 45 ppbv in winter and increase up to 65 ppbv in summer, while exhibiting very little interannual variability (results not shown). This lack of modeled interannual variability was also noted in the European RegCM4-CHEM study (Shalaby et al. 2012) and attributed to the lack of interannual variability in the lateral and top boundary conditions. Prior studies show that tropospheric O3 in the Europe and the Middle East is affected by the top boundary condition and transport from the stratosphere (Katragkou et al. 2010), although this was held constant in these simulations.

We also evaluate the vertical profile of O3 as simulated by the model to investigate the role of transport from high altitudes, which has been shown to produce an O3 hotspot in the Middle East (Lelieveld et al. 2002). Using the online coupled chemistry-climate model, we can investigate this feature and the role of local O3 production versus O3 transported from the stratosphere. The vertical distribution of O3 indicates high concentrations of up to 90 ppbv near the model top (approximately 100 mb) due to the top model layer incorporating part of the stratosphere (Fig. 9A). Some of this stratospheric O3 is mixed down into the model troposphere up to about 500 hPa, although we note that other studies indicate that this mixing may occur down into the free troposphere (Tyrilis et al. 2013) and this downward mixing appears to be underestimated by the model. From 20° to 50° E, regional surface O3 production contributes to higher concentrations of O3 in the lower troposphere. This local O3 production coincides with urban areas, such as the GC region (visible at 30° E), producing local O3 concentrations of ~65 ppbv in the model. This suggests that if the emission sources are being modeled correctly, some of the O3 hotspot noted in the Middle East may be partly due to local surface O3 production, adding to the penetration of stratospheric O3 in the region.

As noted in Section 3.0, the RegCM-CHEM predicts a slight cool bias in the in the winter and a warm bias in the summer in the GC region (Table 2). Typically, O3 tends to increase with increasing temperatures (Steiner et al. 2010). If the model’s warm bias were affecting O3 production, we would expect that the model would also over-predict summer O3 formation. However, the model under-predicts O3 concentrations in the summer and it is therefore unlikely that the O3 bias is due to the temperature bias. Therefore, the warm bias may allow more O3 to be produced in the model given the limited NOx. Overall, this analysis indicates that the missing anthropogenic NOx in the model is the driver of an underestimation of local O3 production.

4.2. Aerosol

4.2.1. Sulfate

The RegCM4-CHEM simulates sulfate aerosol based on a simple sulfate conversion scheme that converts SO2 to sulfate (SO4^{2-}) through (1) gas-phase
oxidation of SO₂ by OH, and (2) aqueous formation though the formation of HSO₄⁻ and SO₃²⁻ ions that then react with H₂O₂ to form sulfate (Qian et al. 2001, Solmon et al. 2006). This oxidation scheme under-predicts sulfate over the USA (Owen & Steiner 2012) but still provides a computationally efficient method for estimation of sulfate aerosol. Over the model domain, sulfate formation is linked to SO₂ emissions (Fig. 1C), which are predominantly near urban areas, shipping lanes and sites of fossil fuel combustion. Because Egypt is located beneath a subsidence region, there is also transported sulfate from Europe, in addition to local production.

Modeled sulfate concentrations are highest in JJA and SON, with concentrations >10 µg m⁻³ over the Mediterranean Red Sea and the Arabian Peninsula (Fig. 10A–D). Over the GC region, modeled sulfate concentrations are the dominant species in the anthropogenic aerosol contribution, compared to relatively low concentrations of BC and OC (Fig. 11). While SO₂ emissions peak in DJF, sulfate is highest in JJA and SON (Fig. 11B) when warmer temperatures increase OH concentrations and atmospheric SO₂ oxidation peaks. Modeled sulfate also exhibits seasonal interannual variability, with some summers showing lower concentrations than others (e.g. 2000 and 2003; Fig. 11A). We also compare modeled sulfate concentrations (10 µg m⁻³; Fig. 11A,B) with aerosol speciation data (Fig. 11A–D) from the CACHE field experiment at the Giza CU site from November 2004 to May 2006, which coincides with our modeling time period (Favez et al. 2008a). The observed CACHE sulfate concentrations range from 10 to 20 µg m⁻³ (Fig. 11A,B), yielding a model underestimate of 2 to 10 µg m⁻³. Additionally, the observations show little seasonal variability in sulfate concentrations over the 3 yr of measurements, unlike the model that shows an increase in summer concentrations.

Earlier observations indicate a seasonal maximum concentration of sulfate in the winter, when relatively higher relative humidity enhances chemical conversion (Shakour & Zakey 1998). We correlate the monthly modeled sulfate anomaly (µg m⁻³, as calculated compared to the 1998 to 2009 model mean) with the modeled specific humidity (kg kg⁻¹) (Fig. 11C) to show that the sulfate anomaly is correlated with atmospheric moisture content (R² = 0.55), as sulfate growth is a strong function of relative humidity (Qian et al. 2001). While we do see evidence of increased humidity increasing sulfate concentrations in the model (Fig. 11C), this does not translate to an increase in winter sulfate concentrations. The modeled seasonal sulfate cycle is driven by atmospheric oxidation, and comparison with ground-based observations in the continental USA indicates that oxidation is underestimated in the model (Owen & Steiner 2012). The underestimation of sulfate in all seasons suggests that SO₂ emissions may be too low in this region and that the model is missing some of the atmospheric oxidation processes that may lead to underestimation in the winter.

4.2.2. Black and organic carbon

Simulated BC and OC provide relatively small contributions to the overall aerosol burden, likely due to missing anthropogenic emissions sources from inventories as discussed in Section 2.2. Anthropogenic BC and OC emissions from the GC and Nile Delta areas are evident in all seasons (Fig. 10), with greater emissions (Fig. 2) and resulting concentrations in the winter and early spring (Fig. 11B). Despite larger emissions of primary anthropogenic OC than BC (Fig. 2C,D), the modeled atmospheric concentrations of both species are similar in magnitude. Simulated BC concentrations are typically ~1 µg m⁻³, which is 5 to 12 µg m⁻³ lower than the observed measurements at Cairo University (Fig. 11A,B) (Favez et al. 2008a), with a greater bias in the winter (12 µg m⁻³) than the summer (5 to 6 µg m⁻³).

Modeled OC concentrations of 1 µg m⁻³ are even lower than those observed, with observed concentrations reaching up to 8 µg m⁻³ during the 2005 CACHE experiment in Cairo (not shown), 25 to 30 µg m⁻³ during the fall burning events (Mahmoud et al. 2008) and ~30 µg m⁻³ in other seasons (Favez et al. 2008a) (Fig. 11A,B). This discrepancy is likely due to (1) missing local emissions in the global emissions inventories that may not accurately capture anthropogenic OC emissions from industry or mobile sources, as well as (2) the lack of modeled SOA formation which is an important contributor to the OC burden. Favez et al. (2008b) suggest that SOA formation in Cairo is significant and both water-soluble and water-insoluble species are important in the region, likely deriving from a suite of mobile sources. Comparison of the model with these measurements suggests that we are underestimating the OC by up to a factor of 10 in some seasons, and this underestimate is similar to other important mobile emissions such as NO₂. This underscores the importance of secondary anthropogenic SOA formation in the region and its inclusion in regional chemistry–climate models, and highlights an area of future model development.
Fig. 10. Seasonal modeled climatological aerosol concentrations for the period 1998–2006: aerosol species (A−D) sulfate, (E−H) black carbon, (I−L) organic carbon, and (M−P) dust, as coarse particulate matter (PM₁₀).
4.2.3. Dust

Dust represents one of the most important particulate matter components in the GC region and exhibits a strong seasonal cycle, with greater dust events from the Sahara occurring in the spring (Fig. 10M–P). Observations indicate that dust in Cairo accounts for up to 65% of the total observed aerosol in the spring and 35 to 50% in other seasons (Favez et al. 2008a). In our model simulations, dust emissions are interactive with regional climate parameters and are affected by wind and soil moisture. As a result, modeled dust concentrations exhibit substantial interannual variability (Fig. 11D), with low dust years producing concentrations in Cairo of ~100 µg m\(^{-3}\) and high dust years producing concentrations up to 240 µg m\(^{-3}\). The modeled seasonal cycle produces a peak dust concentration in the spring with a minimum in October (Fig. 11E). This is consistent with observed dust occurrences over Cairo (Zakey et al. 2008, Favez et al. 2008a) which show a similar seasonal cycle. Considering the total contribution from all size bins, the total dust modeled over Cairo is slightly higher (10 µg m\(^{-3}\)) than observed, and the climatological springtime peak occurs 1 mo later than observed in 2004–2006. The size distribution of dust emissions in our simulation may also result in an overestimate of the fine dust mass fraction (Nabat et al. 2012).

Generally, dust concentrations in the GC region depend on the broader climatic conditions in northern Africa. Dust concentrations within the GC region are anti-correlated with local wind speed, which is likely a function of the aerosol burden in the atmosphere instead of wind-driven emissions in the...
source region (Fig. 11F). Therefore, the discrepancies between observed and modeled dust concentrations can be attributed to the prescription of soil aggregate distributions, which determine threshold friction velocities, sandblasting efficiency and its ratio to the effective vertical dust flux. These very sensitive parameters have been revised in the latest RegCM version, and show a substantial reduction of dust fluxes, concentrations and AOD as compared to the results presented here.

4.2.4. Aerosol optical depth

Despite the lack of consistent aerosol speciation data at ground-based sites, the model’s ability to simulate the climatology of total AOD can be evaluated by comparing with satellite-derived observations. The annual cycle of modeled AOD peaks at 0.7 in spring (MAM) due to dust, decreasing in JJA to 0.2 and increasing slightly in the fall to 0.35 due to secondary sulfate and dust contributions (Fig. 12A–D). On a seasonal basis, the MODIS ‘Dark Product’ Terra and Aqua satellite retrievals show a similar pattern as modeled, with a springtime peak typically attributed to dust that is slightly lower (AOD = 0.5) than modeled and a second peak in September (AOD = 0.55) associated with biomass burning that is higher than modeled (Fig. 12E,F). However the Aqua and Terra ‘Deep Blue’ retrievals exhibit a different seasonal cycle that does not exhibit the fall peak of biomass burning. Additionally, the Terra ‘Deep Blue’ retrieval observes a different seasonal cycle than the other 3 products, with a JJA maximum and the lack of a spring dust peak. With the exception of the Terra ‘Deep Blue’ retrieval, these comparisons with the remotely sensed AOD data suggest that the dust model is producing too much particulate matter from the Sahara in the spring. Ground-based AERONET data (not shown) support the ‘Dark Product’ MODIS retrievals, as they typically show a fall peak in the 500 nm portion of the spectrum (El-Metwally et al. 2010, Marey et al. 2011).

5. DISCUSSION AND CONCLUSIONS

We used the model RegCM4-CHEM to conduct a decadal scale simulation of coupled chemistry and climate over northern Africa and the Mediterranean, with a specific focus on the GC region. Because this region is undergoing massive transformation in terms of its social, political and economic climate, this work provides a baseline for understanding the interaction between atmospheric chemistry and climate in the region. Additionally, this study presents the first climatological simulations of tropospheric O3 and aerosols in the GC region to assess the ability of models to simulate air quality. Because this study was designed to provide a broad evaluation of the model in the northern Africa region, we selected a relatively coarse grid cell resolution (60 km) in order to capture all emissions that may affect this broad area. Our results indicate that this resolution is too coarse to fully understand the air quality factors in the urban core of Cairo; however, they can provide insight into future air quality modeling efforts for the GC region.

We find that the global emissions inventories used over the GC region tend to under-predict key anthropogenic species, such as NOx and anthropogenic black and organic carbon. The model greatly underestimates average July daily maximum NO2 concentrations by 15 ppbv (Fig. 6D), and this could be attributed to missing local mobile emissions, or the relatively coarse model grid cell resolution (60 km) and its reduced ability to capture boundary layer dynamics related to urban air quality. Because similar resolution simulations in Europe did not indicate this NO2 discrepancy, this suggests that the underestimate is due to the local GC emissions inventory. For atmospheric aerosols, our model simulations generally under-predict the fall burning events that are important for air quality in this season. Most major global inventories does not include the fall agricultural biomass burning that occurs in the Nile Delta region, and even new, satellite-derived fire products may not be able to detect these low-temperature burns due to the nature of the burning itself as well as the satellite overpass time in the Nile Delta. Additionally, our urban aerosol analysis is limited by the lack of secondary organic aerosol (SOA) formation in the current model, as prior studies and this model comparison indicate that this is an important component of the total aerosol concentration in all seasons (Favez et al. 2008a). Because biogenic volatilic organic compounds (VOC) emissions are low in the GC region, understanding organic aerosol and the role of SOA formation from anthropogenic emissions in Cairo will be necessary for the accurate simulation of aerosols in the region, and future model simulations will improve upon these known weaknesses. Finally, ground-level concentrations in highly polluted areas are affected by other sources emitting pollutants at the surface, including the cement industry, metal smelters, brick factories, aluminum, petrochemical and chemical production, and sugar fac-
The simulation of tropospheric \( \text{O}_3 \) in the region remains a challenge. The regional chemistry–climate model captures the general seasonal cycle of \( \text{O}_3 \), but the model underestimates the observed July summer maxima by 10 ppbv in the GC region. Previous studies have suggested that this may be due to transport from other regions but our evaluation of local \( \text{NO}_2 \) observations versus modeled values indicate that local \( \text{O}_3 \) production is under-predicted in the model and can be regionally important. Future work will investigate the role of model resolution, emissions inventories, and photochemical processing to reduce this bias.

Overall, this study highlights the need to improve inventories in the region to account for unique local
sources of biomass burning and regional atmospheric chemistry. Despite the poor air quality of Cairo, air quality studies in the region have historically been very limited, in part due to the lack of complete and robust air quality observations. We hope that this foundational work will motivate an increase in the number and diversity of air quality measurements in this region. Future work at the Egyptian Meteorological Authority will use this framework to continue to evaluate and assess air quality over the GC region and Nile Delta.

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LITERATURE CITED


Lamarque JF, Bond TC, Eyring V, Granier C and others


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