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Natural and anthropogenic aerosols in the Eastern Mediterranean and Middle East: Possible impacts

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HIGHLIGHTS

- A modeling study on aerosol–cloud–precipitation interactions is presented.
- Formation of CCN and IN is examined with respect to aerosol properties.
- Semi and indirect effects of aerosols are more important than direct ones.
- Air quality model performance improved by incorporating new mechanisms.
- Integrated modeling study showed the benefits of using fully coupled atmospheric/air quality models.

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ABSTRACT

The physical and chemical properties of airborne particles have significant implications on the microphysical cloud processes. Maritime clouds have different properties than polluted ones and the final amounts and types of precipitation are different. Mixed phase aerosols that contain soluble matter are efficient cloud condensation nuclei (CCN) and enhance the liquid condensate spectrum in warm and mixed phase clouds. Insoluble particles such as mineral dust and black carbon are also important because of their ability to act as efficient ice nuclei (IN) through heterogeneous ice nucleation mechanisms. The relative contribution of aerosol concentrations, size distributions and chemical compositions on cloud structure and precipitation is discussed in the framework of RAMS/ICLAMS model. Analysis of model results and comparison with measurements reveals the complexity of the above links. Taking into account anthropogenic emissions and all available aerosol–cloud interactions the model precipitation bias was reduced by 50% for a storm simulation over eastern Mediterranean.

1. Introduction

The Mediterranean region and Middle East are well known places of high aerosol and ozone concentrations. The characteristic paths and scales of transport and the changes in the physical and chemical properties of the aerosols along the followed paths have been the subject of several past studies (e.g. Kallos et al., 1998, 2007). Aerosol levels have several impacts on various gaseous pollutants but the most important effects are associated with radiation, clouds and precipitation (direct and indirect effects). Airborne particles of natural or anthropogenic origin, may act as efficient cloud condensation nuclei (CCN), depending on their concentrations, their size distributions and their chemical composition. Some particles — such as mineral dust and soot — may also act as ice nuclei (IN) and contribute in the formation of ice particles in high clouds. For example, sea salt, sulphates, and nitrates (soluble aerosols), are responsible for the formation of cloud droplets as the air rises and the relative humidity increases to slightly above saturation near the cloud base (Levin and Cotton, 2009). Moreover, the role of natural or anthropogenic particles, such as mineral dust and black carbon, as IN has been demonstrated in several studies (e.g. Pruppacher and Klett, 1997; Liu et al., 2009). There is increased evidence that the suspension of sulphate and soot particles produced during fossil fuel use and biomass burning increases the ice number concentrations through ice nucleation mechanisms (Penner et al., 2009). On the other hand, formation of secondary particles and atmospheric aging of aerosol lead to particles with substantially different properties than those at source regions. For example, desert dust particles being initially not very soluble and ineffective CCN can become coated with soluble material turning them into effective gigantic CCN (GCCN) (Levin et al., 2005). The physical processes and interactions that define the above links can be addressed within the framework of an integrated atmospheric and air pollution model. In this study, several experimental runs were

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performed with an enhanced version of the RAMS/ICLAMS model initially described in Solomos et al. (2011) focusing mostly on the amounts and chemical compositions of the available airborne particles that could be activated as CCN or IN for each particular case. A quick description of this modeling capabilities and configuration is provided in Sections 2.1 and 3.1. The effects on precipitation and the microphysical structure inside the clouds are discussed with regard to the various types of air masses and aerosol mixtures.

2. Effects of air quality on cloud development

2.1. Model configuration

The model includes an advanced microphysical scheme (Meyers et al., 1997) with eight categories of water (vapor, cloud droplets, rain droplets, pristine ice, snow, aggregates, graupel and hail) and also an interactive mineral dust and sea salt cycle, biogenic and anthropogenic pollutant emission/transport/depletion processes, gas and aerosol chemical reactions. The radiative transfer scheme in the model (RRTM — Mlawer et al., 1997; Iacono et al., 2000) includes the aerosol feedbacks on radiation fluxes. Activation of CCN into cloud droplets is explicitly computed with the scheme of Fountoukis and Nenes (2005) based on the properties of airborne particles. The formation of IN is also calculated online with the scheme of Barahona and Nenes (2009) based on the modeled air quality properties.

Initially the model is set up in a 2-D configuration assuming flat terrain so that the effects of microphysics in cloud processes can be isolated from the possible topographic forcing. A single sounding that is representative of unstable atmospheric conditions is used to initialize the runs and a warm and moist bulb is applied in the center of the domain in order to trigger convection. Full-scale 3-D runs have been performed and discussed in Section 3.

Conceptual 2-D model runs for twelve different mixtures of aerosol particles are implemented, as seen in Table 1. Each run lasts for 6 h. The distribution of the particles in the model is represented by a three-modal lognormal distribution (fine–accumulated–coarse) with constant geometric dispersion ($\sigma = 2$) and modal diameters varying according to the needs of each experiment. The chemical composition and the aerosol concentrations also vary for each model run (see Table 1). The airborne particles are assumed to be chemically-inert during these simulations.

2.2. Formation of CCN from airborne particles

Activation of the different aerosol types as CCN results in significant variation in the total accumulated precipitation as indicated in Fig. 1. The aerosols that consist of dust particles and externally coated with a soluble material produce similar precipitation amounts — within the range of 300–350 mm. These results seem not to be affected by the chemical composition of the soluble fraction as seen for Cases 1–3,

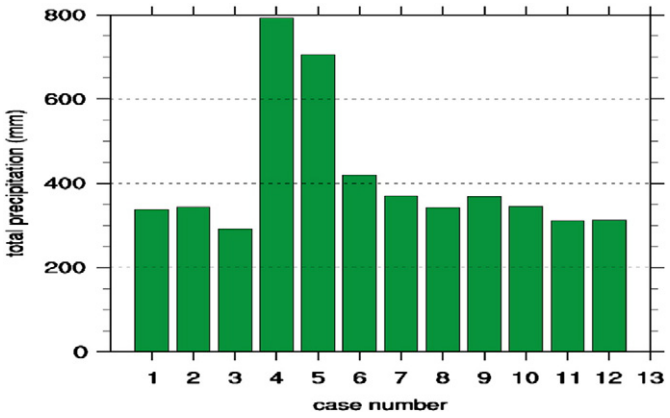

Fig. 1. Domain total accumulated (6 h) precipitation (mm) for the twelve aerosol scenarios.

where the soluble fraction is sodium chloride (NaCl) and Cases 9–11, where the soluble material is ammonium sulphate $(NH_4)_2SO_4$. However, by increasing the hygroscopicity of the particles (the percentage of soluble material), the accumulated precipitation is reduced (i.e. Case 3, Case 11). This is due to the increased number of cloud droplets resulting in slower auto-conversion rates of cloud to rain sizes. In Cases 6–8, the particles are assumed to be completely soluble (NaCl) with different size distribution characteristics (see Table 1). The results indicate a slight reduction in total precipitation for the aerosols with the greater particle diameters per size mode that is possibly related to the formation of bigger cloud and rain droplets and the suppression of the precipitation from the ice phase of the cloud.

An interesting result is the almost 100% increase in total precipitation that is found in Cases 4–5 (completely soluble particles) compared to Cases 1–3 (partially soluble particles). In both Case 4 and Case 5 runs, the precipitation rates remain relatively high even during the latest stages of cloud development. This is an indication of significant contribution of ice processes to the overall precipitable water. In order to illustrate the differences that are attributed solely to chemical composition we selected to compare the cloud properties between Case 4 and Case 12 as depicted in Fig. 2. These cases are representative of totally soluble aerosol particles with the same size and concentration properties but with different chemical characteristics, namely NaCl (Case 4) and $(NH_4)_2SO_4$ (Case 12). As seen in Fig. 2a, after the first modeling hour the precipitation rate for Case 12 falls below 4 mm/h while after 3 h the rainfall has been ended. In contrary, the rate remains very high for Case 4 (blue line in Fig. 2a). These results illustrate the crucial role of the chemical properties (Table 2) in the temporal evolution of the microphysical processes. For example, the effectiveness of ammonium sulphate particles to form CCN results in increased cloud droplet concentrations throughout the modeling period (Fig. 2b). This

Table 1
Aerosol characteristics for the twelve modeling scenarios.

Case	Chemical composition	Soluble fraction	Concentration (cm^{-3})	Mean diameter (fine–accumulated–coarse) (μm)
1	Dust + NaCl	0.2	1000	0.02–0.2–2
2	Dust + NaCl	0.5	1000	0.02–0.2–2
3	Dust + NaCl	0.7	1000	0.02–0.2–2
4	NaCl	1.0	1000	0.02–0.2–2
5	NaCl	1.0	2000	0.02–0.2–2
6	NaCl	1.0	1000	0.05–0.2–2
7	NaCl	1.0	1000	0.02–0.5–2
8	NaCl	1.0	1000	0.02–0.2–5
9	Dust + $(NH_4)_2SO_4$	0.2	1000	0.02–0.2–2
10	Dust + $(NH_4)_2SO_4$	0.5	1000	0.02–0.2–2
11	Dust + $(NH_4)_2SO_4$	0.7	1000	0.02–0.2–2
12	$(NH_4)_2SO_4$	1.0	1000	0.02–0.2–2

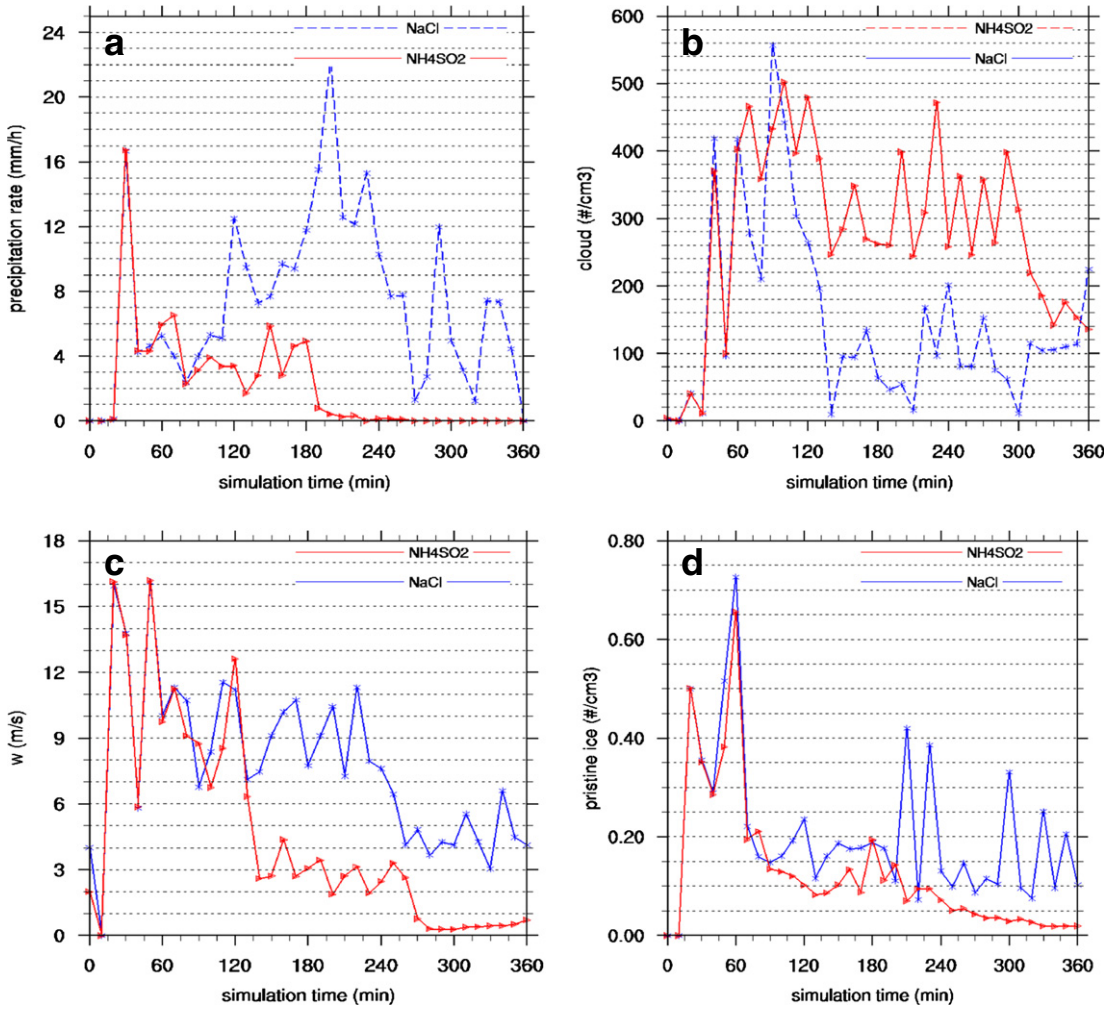

Fig. 2. a) Maximum precipitation rate (mm/h). b) Maximum cloud droplet concentration (cm^{-3}). c) Maximum updrafts (m/s). d) Maximum ice concentration (cm^{-3}) for Case 4 (blue lines) and Case 12 (red lines).

suppresses the formation of precipitable-size rain droplets due to the increased competition between smaller droplets for the available moisture.

Moreover, the most significant difference between these two runs comes from the ice stage of the cloud. As seen in Fig. 2c the cloud updrafts in Case 4 are significantly higher than in Case 12. This cloud contains more ice near the cloud top (Fig. 2d). Melting and rimming of the frozen elements invigorated the production of graupel and hail at the middle and higher cloud layers and the major part of precipitation during the 4–6 h of simulation is generated from this stage of the cloud. Additionally, one more run is performed for the same air quality properties as in Case 4. In this run the initial conditions are slightly changed and the dew point temperature is reduced by 1 °C in the initial sounding. The total accumulated precipitation in this case is more than two times lower than in Case 4. Such results imply the importance of the synergetic effects between air quality and meteorology since a minor change in any of the two can lead in significant precipitation variability.

Table 2
Chemical properties of the aerosol soluble fraction.

	Density ($kg\ m^{-3}$)	Molar mass ($kg\ mol^{-1}$)	Van't Hoff factor (ions $molec^{-1}$)
NaCl	2165	0.058	2
$(NH_4)_2SO_4$	1760	0.132	3

2.3. Formation of IN from airborne particles

The interplay between air quality and high clouds has also been tested within the framework of the model, assuming an initial sounding that is representative of a cold cloud structure. The amount of ice particles that will activate during cloud formation depends on the IN concentration, atmospheric conditions and also on the competition between homogeneous and heterogeneous ice processes. For example, by considering twelve different concentrations of dust or soot particles that can be activated as IN, the respective accumulated precipitation performs great variance as seen in Fig. 3a. In general, the precipitation remains similar for both species until an aerosol concentration of 50 $\mu g\ m^{-3}$. After this threshold, the results vary considerably. Maximum precipitation values are found for the 500 $\mu g\ m^{-3}$ and for the 1000 $\mu g\ m^{-3}$ of soot and dust particles respectively. A significant amount of precipitation for the 1000 $\mu g\ m^{-3}$ of soot scenario is hail.

Further increase of the aerosol concentrations results in less precipitation as these clouds contain great amounts of small ice elements and finally burn off before these condensates manage to grow up to precipitable sizes. The sensitivity of precipitation towards IN properties is also tested for four different IN spectra namely: MY92 (Meyers et al., 1992), PDG07 (Phillips et al., 2007), PDA08 (Phillips et al., 2008), and CNT (Pruppacher and Klett, 1997; Barahona and Nenes, 2008). These distributions have been widely used in similar studies in the past. As seen in Fig. 3b, explicitly resolving of the competition between homogeneous and heterogeneous freezing results in about 25% more precipitation

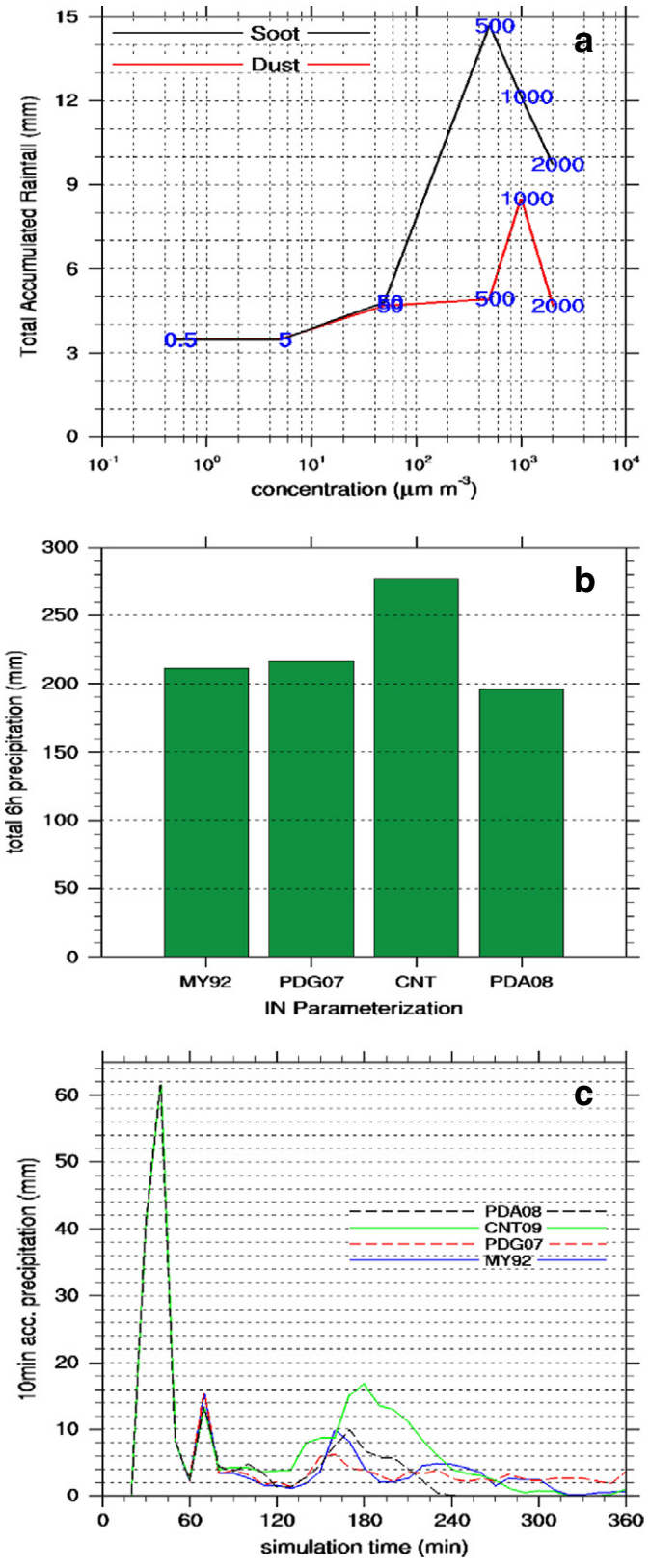

Fig. 3. a) Total accumulated precipitation (mm) for various concentrations of soot (black line in μg m^{-3}) and dust (red line in μg m^{-3}). b) Total domain precipitation (mm). c) Ten minutes of accumulated precipitation (mm).

according to these conceptual runs. The precipitation rate is similar for all IN spectrums during the first modeling hour (Fig. 3c). During this stage all of the precipitation comes from the warm phase of the cloud.

Significant variability is only evident after 2 h of run when the production of rain from ice processes becomes dominant. Similar results regarding the variability of the condensate mixing ratios and precipitation due to IN variations have also been found in earlier relevant studies (e.g. Phillips et al., 2008; Barahona and Nenes, 2009; Teller et al., 2012). In this context, it is expected that the proper knowledge and parameterization of the air quality properties in an integrated modeling study can lead to a better understanding of the aerosol indirect effect. Adaptation of the above air quality/meteorology approach for the interpretation of the physical interactions during a past event over Mediterranean is discussed in the following section.

3. Cloud–aerosol interactions over Mediterranean

3.1. Case study description and model setup

On 29 January 2003, an event of heavy precipitation took place over Eastern Mediterranean. Previous studies on this event (Levin et al., 2005; Solomos et al., 2011; Teller et al., 2012) indicated that the clouds in this area were affected by mineral dust and sea salt particles penetrating the base of the clouds (Fig. 4). A considerable percentage of these particles was activated as CCN. The selected case is representative of a weather pattern that is responsible for significant precipitation heights along the Middle East coast. Such storms are essential for the water supplies of the population at these areas. Moreover during this event the MEIDEX experiment was active providing a valuable set of aircraft and ground observations thus making this event a suitable benchmark for testing new model developments. In the present study, this event is examined taking also into consideration the anthropogenic aerosols over the region (sulfate particles) and their role on cloud and precipitation. The gas, aqueous and aerosol phase chemistry mechanisms namely SAPRC99 (Carter at al., 2003) and ISORROPIA (Nenes et al., 1998) are directly coupled with the RAMS/ICLAMS model described in Solomos et al. (2011) and are activated for these runs. The model configuration for these simulations is described in Table 3. The sulfate aerosol properties are calculated in the aqueous phase chemistry sub-module of the model and these particles are also included in the CCN activation calculations. The aerosol size-spectrum is assumed to fit in a 5-modal lognormal distribution where the first three modes represent mineral dust, accumulated and coarse sea salt particles respectively. The last two modes represent the accumulated and coarse sulphates.

The number concentrations of all five modes are prognostic model variables. The first mode (dust) is assumed to fit a distribution with constant deviation ($\sigma = 2$) and the median diameter is recalculated at every model step. The second and third modes (accumulated and coarse sea salt) have median diameters of 0.36 μm and 2.85 μm and geometric deviations of 1.80 and 1.90 respectively. Geometric standard deviation and geometric mean diameter for the accumulated sulfate are explicitly calculated. For the coarse sulfate, the geometric mean diameter is also explicitly calculated and the standard deviation is set to 2.2.

The Israeli Mediterranean coast is one of the areas with significant concentrations of sulfate aerosols due to major sulfur emission sources (Luria et al., 1996). Other sources of anthropogenic pollutants in the Eastern Mediterranean are shipping and major urban conglomerate upwind from the area under consideration. These sources are included in the emission inventory used in this study. The emission inventory used has been prepared at the framework of the EU-funded Framework Program 6 project CIRCE (http://www.circeproject.eu). Measurements of fine aerosol particles (diameter < 1 μm) during this event (Levin et al., 2005) showed that, below 1000 m, anthropogenic sulfate represented 11.6% of the total sample. For measurements above 1000 m the respective ratio reached 35%. The average ratio of the modeled sulfate aerosols to the total particles is 11.9% below 1000 m and 39.6% above 1000 m indicating a good agreement with the observations. As seen also in Fig. 5 comparison of modeled PM10 with ground measurements at the stations of Afule ($R = 0.866$), Modiin ($R = 0.907$) and Beer Sheva

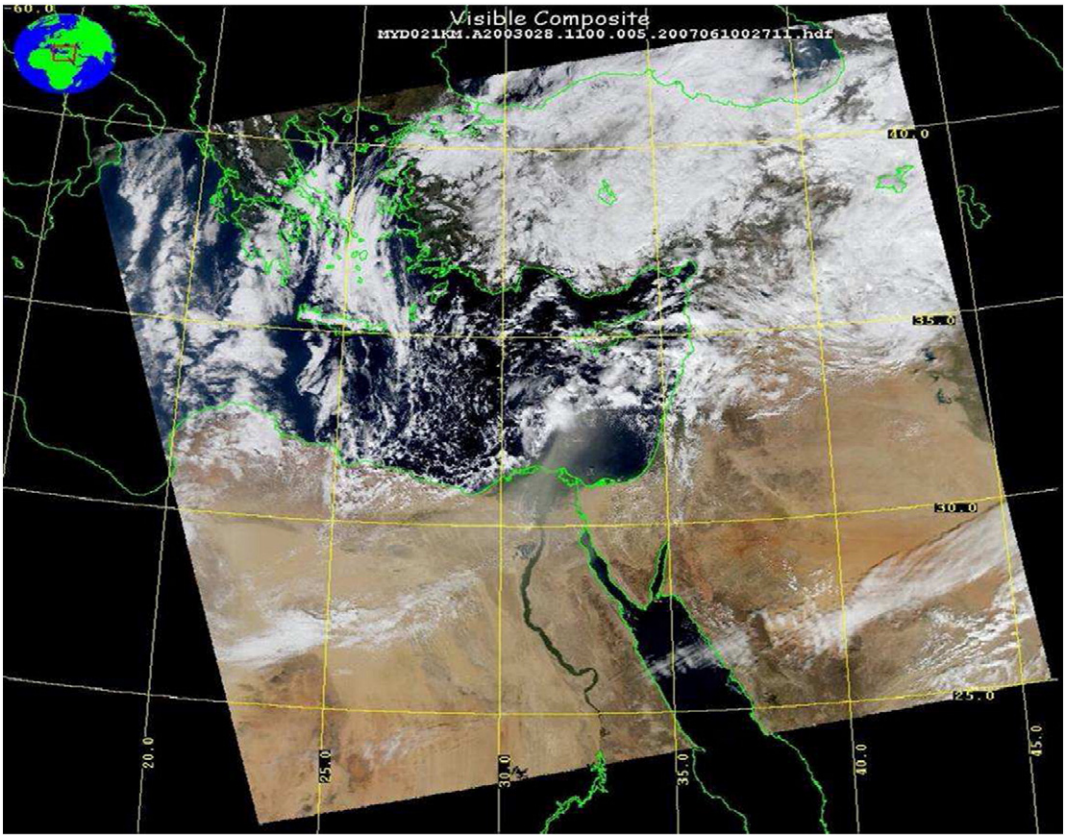

Fig. 4. Satellite image over Eastern Mediterranean on 28 January 2003, at 11:00 UTC (MODIS-Aqua). Colocation of dust and clouds is evident over SE Mediterranean (http://modis.gsfc.nasa.gov/).

($R = 0.895$) denotes the abrupt increase of particulate matter concentrations in the area during 28–29 January 2003. This increase is mostly related to the dust event.

3.2. CCN activation of natural and anthropogenic aerosols

Three sensitivity studies were performed for this event: SIM0 — reference simulation with no impact of aerosols on clouds, SIM1 — simulation with impact of natural aerosols (mineral dust and sea salt) and SIM2 — simulation with impact of natural and anthropogenic (sulfate) aerosols. The averaged values of hourly precipitation during 29 January 2003 show similar variation for SIM1 and SIM2. As shown in Fig. 6a, slightly higher precipitation rates are found in the SIM2 case, however the average 24 h accumulated precipitations do not differ significantly (SIM1: 60.69 mm, SIM2: 63.81 mm).

The anthropogenic aerosol forcing is more noticeable in the hourly precipitation maxima and timing (Fig. 6b) and in the dislocation of the rainfall areas. As shown in Fig. 7a–b including the sulfate aerosols in the computation of the cloud droplet activation results in the suppression of cloud development over the sea. The clouds in this case exhibit more vigorous development inland towards the eastern part of the domain. In the next hour of the simulation the cloud formation is horizontally constrained and vertically enhanced over the coastal zone while ice formation is less favored compared to the SIM1 cloud structure (Fig. 7c–d). Despite the differences in the horizontal and vertical structures the two cloud systems reached the same top height (10 km) at 10:00 UTC.

Table 3
Model configuration.

Grid number	Grid 1/grid 2/grid 3
Horizontal spacing	12 km/4 km/1 km
Horizontal X grid points	Nx: 188/182/154
Horizontal Y grid points	Ny: 116/146/106
Domain center	(33° N, 28° E)/(33° N, 33° 30′ E)/(33° N, 35° 10′ E)
Statistics time period	29 January 2003
Cloud microphysics	SIM0: no impact of aerosols on clouds SIM1: impact of natural aerosols (mineral dust and sea salt) on clouds SIM2: impact of natural and anthropogenic (sulfate) aerosols on clouds

3.3. Comparison with measurements

In order to assess the model performance for the different aerosol scenarios, the 24-hour accumulated precipitation is compared with observations from 86 stations located over Northern Israel (Z. Levin, pers. comm., 2010). The bias analysis of precipitation is based on the behavior of the modeled exceedances versus observational exceedances. The model bias is calculated as:

$$\mathrm{bias} = \frac{a+b}{a+c}$$

where, a is the number of exceedances that was correctly modeled, b is the number of modeled exceedances that was not observed and c is the number of the observed exceedances missed by the model. Bias greater than one indicates overprediction while bias less than one indicates underprediction. Unbiased model results have bias equal to 1. The bias scores are calculated for all three cases for 29 January 2003 (Fig. 8). In general, the passive tracer experiment (SIM0) underestimates precipitation while the two feedback runs (SIM1 and SIM2) overestimate the precipitation at high thresholds. In order to extract a single metric for the model performance at each aerosol scenario the bias performance

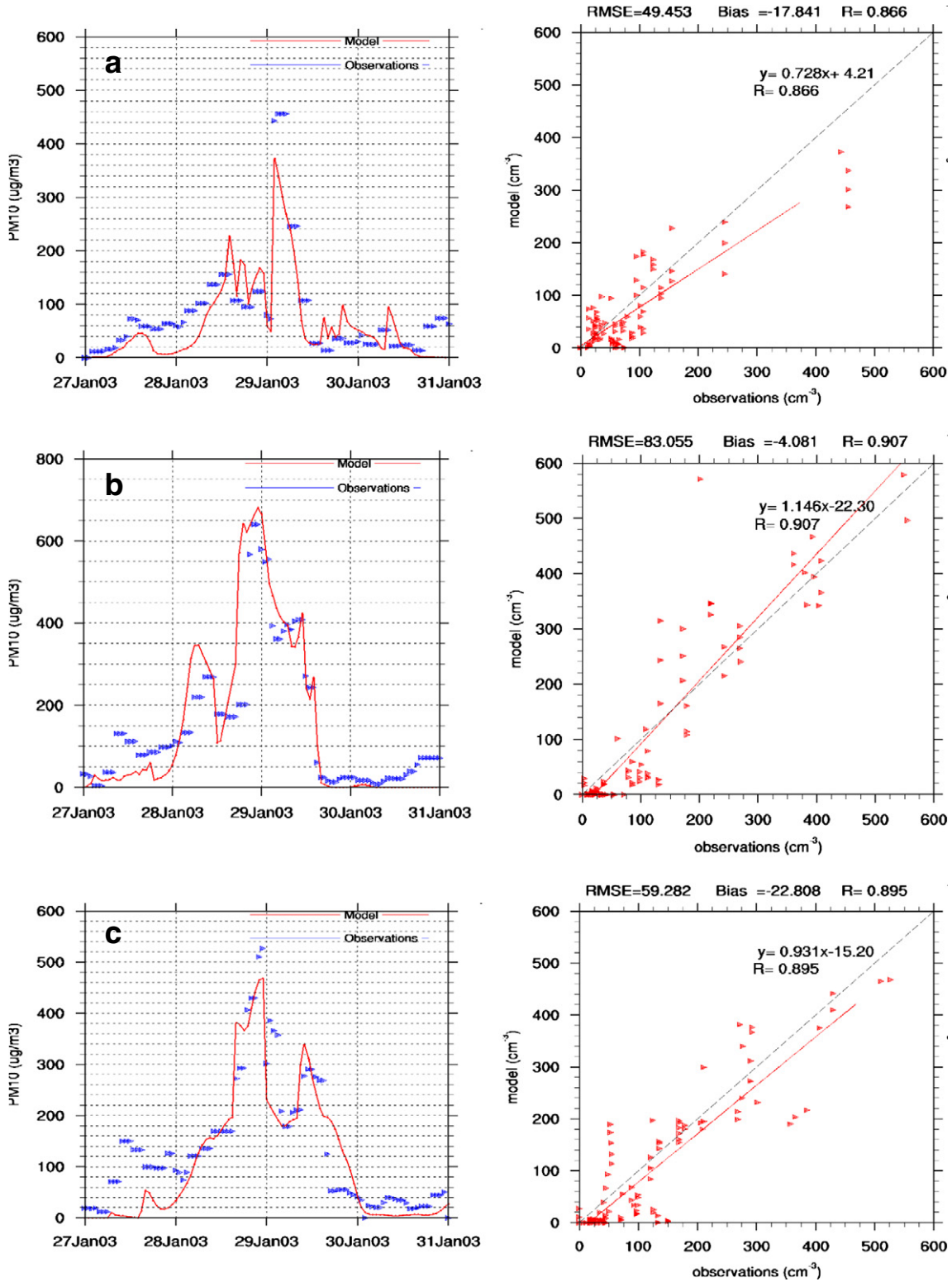

Fig. 5. Comparison of PM10 (μg m^{-3}) measurements (blue dots) and modeled (red lines) at the stations of: a) Afule, b) Modiin and c) Beer Sheva during 27–31 January 2003.

index (BPI) is calculated as the sum of the absolute differences of each thresholds biases from unity:

$$\mathrm{BPI} = \sum_{i=1}^{N} |\mathrm{bias}_i - 1|,$$

where i is the number of the precipitation thresholds. The closer the BPI value is to zero the greater the agreement between the model and the observed values of precipitation (threshold biases closer to 1). The respective BPI value for each case is shown in parentheses next to the legend labels in Fig. 8. Values closer to zero indicate better agreement between model results and observations. The improvement of about

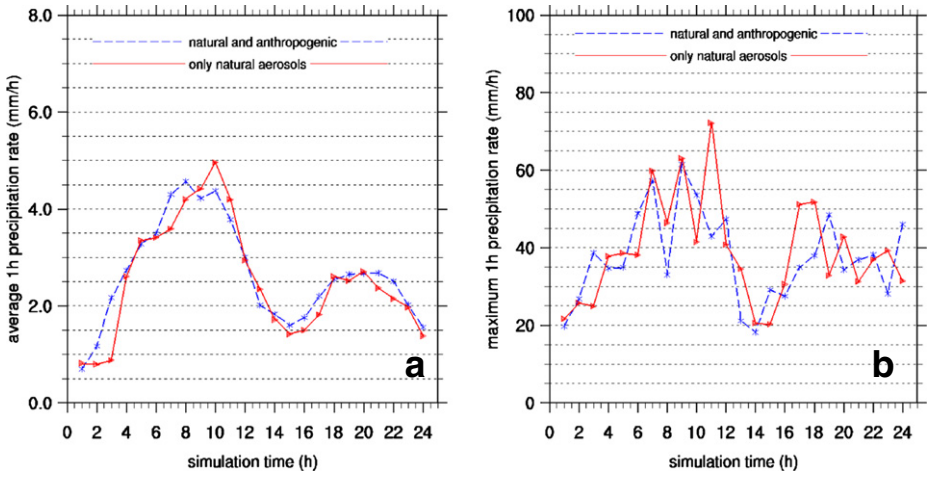

Fig. 6. Time series of the hourly precipitation rate in mm/h. a) Averaged over North Israel and b) maximum values on January 29th, 2003 with natural aerosols (red line) and with natural and anthropogenic aerosols (blue dashed line).

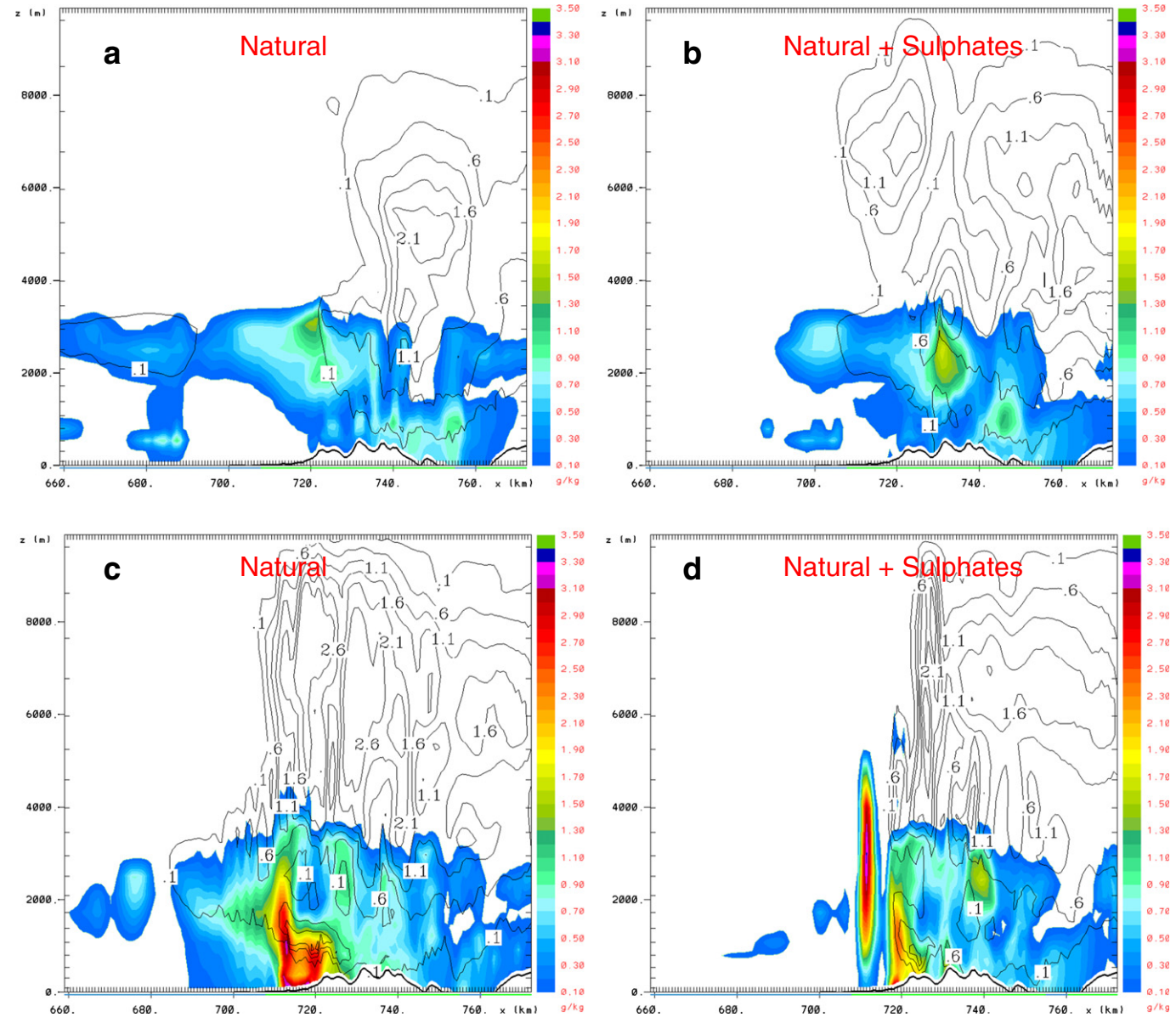

Fig. 7. West–East cross section of liquid water mixing ratio (color scale in g kg^{-1}) and ice mixing ratio (black line in g kg^{-1}) over Haifa (latitude = 32.81, longitude = 34.99) on 29 January 2003 at a) 09:00 UTC with natural aerosols, b) 09:00 UTC with natural and anthropogenic aerosols, c) 10:00 UTC with natural aerosols and d) 10:00 UTC with natural and anthropogenic aerosols.

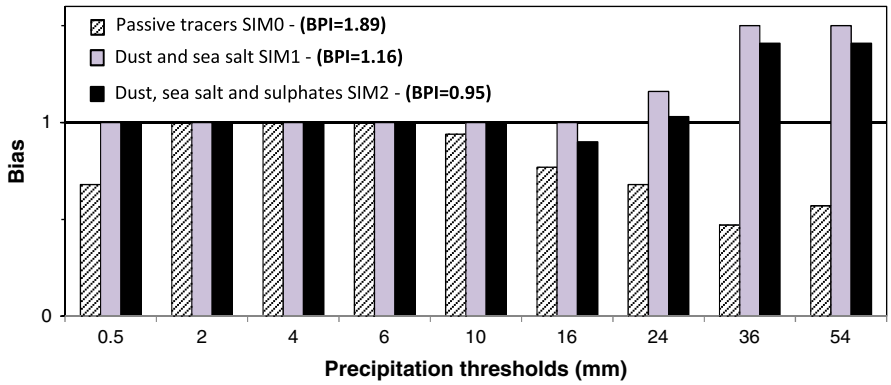

Fig. 8. Bias of the 24-hour accumulated precipitation for 86 stations for each threshold and for three cases of aerosol composition. The number of stations exceeding each threshold is denoted in parenthesis: 0.5 (86), 2 (86), 4 (86), 6 (86), 10 (84), 16 (81), 24 (57), 36 (34) and 54 (14) mm h^{-1}. The BPI for each case is specified within parenthesis after the legend label.

18% that is found for SIM2 (BPI = 0.95) compared to SIM1 (BPI = 1.16), is possibly attributed to the more detailed representation of the aerosol field due to the inclusion of the anthropogenic aerosol. Compared to the reference run the model performance is improved by 39% in SIM1 (only natural sources) and by 50% in SIM2 (natural and anthropogenic sources).

4. Conclusions

Based on the results from the conceptual model runs and the real event analysis we can derive the following concluding remarks: Cloud and precipitation processes are found to be very sensitive to variations in both concentrations and chemical compositions of the airborne particles. These feedbacks are attributed to the complex role of the aerosols in cloud microphysics for both the warm and cold stages of the clouds. Model results indicate that even minor changes in the aerosol field can result in considerable changes in cloud structure and precipitation. In addition, the efficiency of several aerosols to form IN depends on their composition. Similar concentrations and size distributions of mineral dust and anthropogenic soot modify the cloud structure in different ways, thus adding to the complexity of the system.

The aerosol forcing is more evident in the spatial and temporal distribution of clouds and precipitation and less in the total amount of rainfall. In general, the amount of precipitation is not significantly affected by the air quality properties since convective and stratiform precipitations are mostly governed by atmospheric dynamics. However, the temporal evolution of a cloud system and the corresponding time and type of precipitation (rain, snow, hail) are found to be highly dependent on the available CCN and IN. Certain combinations of atmospheric composition (mainly aerosols) and atmospheric conditions are likely to trigger flood events or, in contrary, to suppress rainfall. For example, when the ice processes are favored in the presence of dust or soot IN, the accumulated precipitation was in some cases found to increase up to five times. The sensitivity tests reveal a clear link between aerosol physical and chemical properties and clouds but this link is not always towards the same direction while the intensity of the associated phenomena varies significantly.

The detailed analysis of a characteristic case in the Eastern Mediterranean where both anthropogenic and natural aerosol sources were considered resulted in a model precipitation bias that is half the bias of the non-interactive model. Moreover, the bias is improved by 18% between the only natural and the natural and anthropogenic runs. The CCN and IN activation schemes used in this study are based on fundamental physical principles. Thus the continuous improvement of the results with the inclusion of the additional information is an indication that a number of physical processes related to the indirect effect are now covered on a satisfactory way. Following this approach, more work needs to be done in order to a) further improve our interpretation of similar phenomena and b) investigate other interactions between aerosols and clouds (for example the role of bacteria in IN that seems to be the next important step in cloud microphysics). Additional model runs utilizing also in situ and laboratory experimental measurements are currently underway in order to improve our understanding on the indirect effect.

Acknowledgments

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