

1 An integrated modeling study on the effects of mineral dust and sea salt  
2 particles on clouds and precipitation.

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31 **Abstract**

32 The amount of airborne particles that will nucleate and form cloud droplets under  
33 specific atmospheric conditions, depends on their number concentration, size  
34 distribution and chemical composition. Aerosol is affected by primary particle  
35 emissions, gas-phase precursors, their transformation and interaction with  
36 atmospheric constituents, clouds and dynamics. A comprehensive assessment of these  
37 interactions requires an integrated approach; most studies however decouple aerosol  
38 processes from cloud and atmospheric dynamics and cannot account for all the  
39 feedbacks involved in aerosol-cloud-climate interactions. This study addresses  
40 aerosol-cloud-climate interactions with the Integrated Community Limited Area  
41 Modeling System (ICLAMS) that includes online parameterization of the physical  
42 and chemical processes between air quality and meteorology. ICLAMS is an extended  
43 version of the Regional Atmospheric Modeling System (RAMS) and it has been  
44 designed for coupled air quality – meteorology studies. Model sensitivity tests for a  
45 single-cloud study as well as for a case study over the Eastern Mediterranean illustrate  
46 the importance of aerosol properties in cloud formation and precipitation. Mineral  
47 dust particles are often coated with soluble material such as sea-salt, thus exhibiting  
48 increased CCN efficiency. Increasing the percentage of salt-coated dust particles by  
49 15% in the model resulted in more vigorous convection and more intense updrafts.  
50 The clouds that were formed extended about three kilometres higher and the initiation  
51 of precipitation was delayed by one hour. Including on-line parameterization of the  
52 aerosol effects improved the model bias for the twenty-four hour accumulated  
53 precipitation by 7%. However, the spatial distribution and the amounts of  
54 precipitation varied greatly between the different aerosol scenarios. These results  
55 indicate the large portion of uncertainty that remains unresolved and the need for  
56 more accurate description of aerosol feedbacks in atmospheric models and climate  
57 change predictions.

58 *1. Introduction*

59

60 Aerosols are a mixture of natural and anthropogenic particles. Mineral dust, sea-salt,  
61 primary biogenic particles and volcanic ash originate from natural sources, while

62 anthropogenic particles originate from industrial activity, fossil-fuel and biomass  
63 burning. Aerosol particles directly affect the radiation budget of the atmosphere by  
64 absorbing and/or scattering radiation across the solar and long-wave radiation  
65 spectrum (Charlson et al., 1992; IPCC, 2001; Myhre et al., 2003b; Seinfeld et al.,  
66 2004; Ramanathan et al. 2007). Additionally, they influence the nutrient dynamics  
67 and biogeochemical cycling of both terrestrial and oceanic ecosystems and may have  
68 considerable impacts on human health (Dockery and Pope, 1994; Herut et al., 2002;  
69 Meskhidze et al., 2003,2005; Meskhidze and Nenes 2006; Mahowald et al., 2008;  
70 Mitsakou et al., 2008). Airborne particles serve as cloud condensation nuclei (CCN)  
71 and ice nuclei (IN); changes thereof can affect the cloud cover, radiative properties,  
72 the distribution of precipitation and the hydrological cycle in general (Twomey et al.,  
73 1977; Albrecht 1989; De Mott et al., 2003; Sassen et al., 2003; Andreae and  
74 Rosenfeld, 2008). Quantifying the number of particles that act as CCN , as well as the  
75 number of particles that can initiate heterogeneous ice formation processes (ice nuclei,  
76 IN) is essential for determining the role of aerosols in cloud and precipitation  
77 processes (e.g., Lohmann and Feichter, 2005; Levin and Cotton, 2009). Moreover,  
78 formation of secondary particles and atmospheric ageing of aerosol lead to particles  
79 with substantially different properties than those at source regions (Seinfeld and  
80 Pandis 1998; Levin et al., 1996; Wurzler at al., 2000; Jacobson 2001; Chung and  
81 Seinfeld 2002).

82 Mineral dust and sea salt are major components of particulate matter in the  
83 atmosphere. Desert dust accounts for more than 50% of the global aerosol load  
84 (Andreae et al. 1986; Zender et al., 2005) and the long range transport of dust  
85 particles can influence the composition and dynamic state of the atmosphere  
86 thousands of kilometers downwind of their source region (Kallos et al., 2007). Under  
87 favourable conditions, dust particles originating from Northern and Central Africa  
88 may get elevated and travel towards Atlantic and Caribbean (Karyampudi, 1979;  
89 Karyampudi et al., 1999; Prospero et al., 2005; Kallos et al., 2006) or cross the  
90 Mediterranean towards Europe affecting both air quality and meteorology in Southern  
91 Europe (Mitsakou et al., 2008 ; Querol et al., 2009). Dust particles are efficient ice  
92 nuclei (IN) and contribute to the formation of ice particles in high clouds (DeMott et  
93 al., 2003a; Teller and Levin 2006). Also they interact with sea salt or anthropogenic  
94 pollutants, mainly sulfates and nitrates, thus forming particles that consist of a core of

95 mineral dust with coatings of soluble material (Levin et al., 2006). The soluble  
96 coating on the dust particles converts them into efficient CCN while maintaining their  
97 ability as IN (Levin et al., 2006; Astitha and Kallos, 2008 ; Astitha et al., 2010). Sea-  
98 salt particles are also very efficient CCN and is the dominant source of particulate  
99 matter in the marine boundary layer (Gong at al., 2002 ; Pierce and Adams, 2006).

100 The amount of particles that can act as CCN and activate to cloud droplets depends on  
101 the concentration of available particles, their size distribution and their chemical  
102 composition (e.g., Köhler, 1936; Charlson et al., 2001; Nenes et al., 2002).  
103 Additionally, absorption of solar radiation by dust, results in heating of the dust layer  
104 and subsequently in modification of the thermodynamic structure of the atmosphere,  
105 thus leading to suppression or enhancement of precipitation depending on the type of  
106 the clouds (Yin and Chen, 2007). The interplay between cloud dynamics and the  
107 composition of the atmosphere may delay the initiation of precipitation or steer a  
108 storm towards a different location and precipitation amounts will vary accordingly  
109 (Lynn et al., 2005b ; van den Heever and Cotton, 2007 ; Rosenfeld at al., 2007 ;  
110 Zhang et al., 2007 ; Cotton et al., 2007).

111 By modifying the microphysical, optical and radiative properties of clouds, dust and  
112 salt particles contribute to the indirect aerosol effect and introduce significant  
113 uncertainty in assessments of anthropogenic climate change (Charlson et al., 1990;  
114 Lohmann and Feichter, 2005; IPCC, 2007; Andreae and Rosenfeld, 2008). The effects  
115 of dust and sea salt on regional climate depend also on the local topography and soil  
116 characteristics (e.g., Junkermann et al., 2009) and cloud type (Seifert and Beheng,  
117 2006). Therefore, the effects of atmospheric composition on clouds and precipitation  
118 are not monotonic and may differ from one area to another.

119 The complexity of the above processes and the possible interactions and feedbacks  
120 across all scales in the climate system, indicate the need for an integrated approach in  
121 order to examine the impacts of air quality on meteorology and vice versa (Stevens  
122 and Feingold, 2009). This study adopts such an approach to study an idealized case  
123 representative of mid-latitude marine boundary layers and a specific test case over  
124 eastern Mediterranean. A description of the new model developments is described in  
125 Section 2. Section 3 includes idealized sensitivity tests as well as the analysis of an  
126 experimental case. Finally the main results are summarized in Section 4.

## 127        2. Description of ICLAMS

128  
129        The Regional Atmospheric Modeling System (RAMSv6) (Pielke et al., 1992; Cotton  
130 et al. 2003) was the basis for developing the Integrated Community Limited Area  
131 Modeling System (ICLAMS) used in this study. This new version of the model has  
132 been designed for air pollution and climate research applications and includes several  
133 new capabilities related to physical and chemical processes in the atmosphere. The  
134 model components are summarised in Table 1; new developments include an  
135 interactive desert-dust and sea-salt cycle, biogenic and anthropogenic pollutants cycle,  
136 gas/cloud/aerosol chemistry, explicit cloud droplet nucleation scheme and an  
137 improved radiative transfer scheme. Each process is in modular form, and each  
138 component can be activated / deactivated during a simulation. The two-way  
139 interactive nesting capabilities of the model allow the use of regional scale domains  
140 together with several high resolution nested domains. This feature is important for the  
141 purpose of the present work since it allows for simultaneous description of long range  
142 transport phenomena and aerosol-cloud interactions at cloud resolving scales. The  
143 explicit two-moment cloud microphysics scheme of the model is used to describe the  
144 aerosol-cloud interactions. The dust and sea salt cycles parameterization together with  
145 the radiative transfer and cloud droplet nucleation modules of the model are described  
146 in the following sections.

### 147 148        2.1 Mineral Dust

149        The dust-cycle simulation adopts the approach of the SKIRON/Dust model (Spyrou et  
150 al., 2010) and is based on the “saltation and bombardment” mechanism (Marticorena  
151 and Bergametti, 1995 ; Lu and Shao, 1999 ; Alfaro and Gomez, 2001; Grini et al.,  
152 2002). The model contains 22 land-use categories (Table 2). All grid cells classified  
153 as “desert” or “semi-desert” are treated as potential dust sources. Saltation occurs  
154 when the friction velocity exceeds a characteristic threshold,  $u_f$ . For the mobilization  
155 of sand particles,  $u_f$  is calculated based on the friction Reynolds number (Marticorena  
156 et al., 1997b; Zender at al., 2003) and the soil wetness for each particular model grid  
157 point (Fécan et al., 1999). Upon mobilization, sand grains ( $>60 \mu\text{m}$ ) are elevated a  
158 few meters above ground; upon resettling, the grains “bombard” the soil and eject

159 secondary silt (2.5-60  $\mu\text{m}$ ) and clay (<2.5  $\mu\text{m}$ ) particles. These particles are  
 160 sufficiently small to remain suspended, get transferred by turbulence within the ABL  
 161 and then to free troposphere from where they can be transported thousands of  
 162 kilometers away from their source. The efficiency with which the mineral dust  
 163 particles are transported vertically is strongly sensitive to their size distribution. The  
 164 finest particles are small enough to be transported to long distances while larger  
 165 particles can only be transported to distances near their sources. Based on this  
 166 approach, the vertical dust flux is distributed into three lognormal source modes with  
 167 different shapes and mass fractions. The transport mode is represented inside the  
 168 model with eight discrete size bins as in Perez et al., (2006a) and Spyrou et al., (2010)  
 169 with effective radii of 0.15, 0.25, 0.45, 0.78, 1.3, 2.2, 3.8, and 7.1  $\mu\text{m}$  respectively.  
 170 Each dust bin is treated as a scalar variable for advection and diffusion purposes.  
 171 Partitioning of the dust spectrum and separate treatment of each size mode is  
 172 important for the description of size dependent processes such as dry and wet  
 173 deposition, CCN activation and radiative transfer.

174

## 175 *2.2 Sea salt spray*

176 The sea salt aerosol emission fluxes depend strongly on the meteorological conditions  
 177 close to the air-sea surface. The most prominent mechanism for the generation of sea  
 178 salt aerosols is the bursting of entrained air bubbles during whitecap formation due to  
 179 surface wind. The method follows the open sea white - cap formation as described in  
 180 Monahan et al., (1986) which gives a continuous particle-size distribution at a specific  
 181 relative humidity (RH), usually 80%.

$$182 \quad \frac{dF_{N-Open}}{dr_{80}} = 1.373 \cdot u_{10}^{3.41} \cdot r_{80}^{-A} \cdot \left(1 + 0.057 r_{80}^{3.45}\right) \cdot 10^{1.607 e^{-B^2}} \quad (1)$$

$$183 \quad A = 4.7 \cdot \left(1 + \Theta r_{80}\right)^{-0.017 r_{80}^{-1.44}}, \quad \Theta = 30 \quad (2)$$

$$184 \quad B = (0.433 - \log r_{80}) / 0.433 \quad (3)$$

185 where  $u_{10}$  is the wind speed at 10m height, and  $r_{80}$  is the particle radius at 80% RH.  
 186 This semi-empirical formulation is based on laboratory measurements for particles  
 187 with radius 0.8–8  $\mu\text{m}$ . The size range of the sea-salt source function has been

188 extended to below 0.1  $\mu\text{m}$  in radius based on the parameterization proposed by Gong,  
189 (2003). Additionally, in order to take into account the hygroscopic nature of sea salt,  
190 the size distribution of the particles is calculated as a function of local RH following  
191 Zhang et al., (2005). This method accounts for the hygroscopic uptake of water from  
192 sea salt particles for RH values between 45% and 99%.

193 Whitecaps may also occur along coastal zones at lower wind speeds than in open seas  
194 because of breaking of internal waves when they interact with the sea bottom and  
195 shore. The sea salt aerosols produced over the surf zone provide an additional surface  
196 for heterogeneous reactions and have a significant impact on PM concentrations in  
197 marine areas (Seinfeld and Pandis, 1998). Coastline sea salt flux is also parameterized  
198 in the model following the work of Leeuw et al., (2000) and Gong et al., (2002).

199 Sea salt particle spectrum is represented with a bimodal lognormal distribution  
200 assuming a mean diameter of 0.36  $\mu\text{m}$  for the first (accumulated) mode and a mean  
201 diameter of 2.85  $\mu\text{m}$  for the second (coarse) mode. Geometric dispersion is 1.80 and  
202 1.90 for the two modes respectively.

203

### 204 *2.3 Dry deposition*

205 Dry deposition for dust and salt particles is treated as a first order removal process,  
206 equal to the concentration multiplied by a mass transfer coefficient,  $V_d$  (termed  
207 “deposition velocity”; Seinfeld and Pandis, 1998; Slinn and Slinn, 1980).  $V_d$  accounts  
208 for the effects of reactivity, hygroscopic water uptake, size distribution of particles,  
209 meteorological conditions and surface characteristics. Wesely (1989) proposed a  
210 resistance model to account for all the elements described above; dry deposition  
211 fluxes are controlled by gravitational settling, turbulent mixing, and Brownian  
212 diffusion across two virtual layers. In the layer adjacent to the surface, Brownian  
213 diffusion (for small particles) and gravitational settling (for large particles) are the  
214 main deposition processes. In the second layer, called the “constant flux layer”,  
215 turbulent mixing and gravitational settling dominate deposition.  $V_d$  of a particle with a  
216 given diameter is then parameterized using a set of mass transfer resistances  
217 associated with the combined effects of these processes in both layers (Wesely, 1989;  
218 Seinfeld and Pandis, 1998) :

219 
$$r_a = \frac{1}{ku_*} \left[ \ln \left( \frac{1}{z_0} \right) - \phi_h \right] \quad (4)$$

220 
$$r_b = \frac{1}{u_* \left( S_c^{-2/3} + 10^{-3/S_t} \right)} \quad (5)$$

221 
$$V_d = V_{sed} + \frac{1}{r_a + r_b + r_a r_b V_{sed}} \quad (6)$$

222 where  $r_a$  is the aerodynamic resistance,  $r_b$  is the boundary resistance,  $k$  is the von  
 223 Karman constant,  $z_0$  is the surface roughness length,  $\phi_h$  a stability correction term,  $S_c$   
 224 is the Schmidt number,  $S_t$  is the Stokes number and  $V_{sed}$  is the gravitational settling  
 225 velocity.

226

227 *2.4 Wet deposition*

228

229 Proper treatment of the wet removal process is essential for a realistic aerosol  
 230 simulation, since it is the predominant removal process for atmospheric particles away  
 231 from their sources. The amount of particles removed at each model timestep from in-  
 232 cloud and below-cloud scavenging is expressed as:

233 
$$\frac{\partial C}{\partial t} = -\Lambda C \quad (7)$$

234 where  $\Lambda$  is the “scavenging coefficient” of the aerosols. For in-cloud removal,  $\Lambda$  is  
 235 calculated from the droplet-aerosol collection efficiency ( $E$ ), the precipitation rate ( $P$ )  
 236 and the radius of the scavenging droplet ( $r_d$ ) following the formulation of Seinfeld and  
 237 Pandis (1998):

238 
$$\Lambda = \frac{3}{4} \frac{EP}{r_d} \quad (8)$$

239 The collection efficiency ( $E$ ) for a particle of radius ( $r_p$ ) is calculated from the  
 240 contribution of Brownian diffusion, turbulent diffusion, interception, inertial  
 241 impaction and electric forces (Slinn, 1984; Seinfeld and Pandis, 1998):

242 
$$E(r_p) = \frac{4}{\text{Re} S_c} \left( 1 + 0.4 \text{Re}^{1/2} S_c^{1/3} + 0.16 \text{Re}^{1/2} S_c^{1/2} \right) +$$

243 
$$+ 4\phi \left[ \frac{\mu}{\mu_w} + \phi(1 + \text{Re}^{1/2}) \right] + \left[ \frac{S_t - S^*}{S_t - S^* + 2/3} \right]^{3/2} \quad (9)$$

244 where Re is the droplet Reynolds number,  $S_c$  is the particle Schmidt number,  $r_d$  is the  
 245 droplet radius,  $\phi = \frac{r_p}{r_d}$ ,  $\mu$ ,  $\mu_w$  are the kinematic viscosities of the air and liquid water,  
 246 respectively,  $S_t$  is the particle Stokes number and

247 
$$S^* = \frac{1.2 + \ln(1 + \text{Re})/12}{1 + \ln(1 + \text{Re})} \quad (10)$$

248

## 249 *2.5 ICLAMS Radiation scheme*

250 The basic options for shortwave/longwave radiative transfer in RAMS include the  
 251 Chen and Cotton (1983) and the Harrington (1997) schemes. The former treats all  
 252 hydrometeors as liquid-phase; the latter scheme includes three shortwave and five  
 253 infrared bands interacting with ice and liquid condensates and with model gases.  
 254 Radiation transfer calculations options in ICLAMS have been extended with the  
 255 implementation of the Rapid Radiative Transfer Model (RRTM) for both SW and LW  
 256 bands (Mlawer et al., 1997; Iacono et al., 2000). RRTM is a spectral-band radiative  
 257 transfer scheme based on the correlated- $k$  method (Lacis and Oinas, 1991; Fu and  
 258 Liou, 1992). Pre-calculated look-up tables are used to simulate the impact of clouds  
 259 and the impact of various atmospheric gases and aerosols in the distribution of the  
 260 radiation along the atmosphere. For both Harrington and RRTM radiation options, the  
 261 aerosol optical depth of prognostic dust has been also added to the calculation of the  
 262 total optical depth to account for its direct radiative forcing and photochemical  
 263 impacts.

264

## 265 *2.6 Cloud droplet nucleation parameterization*

266 RAMS has been widely used for cloud research during the last two decades (Krichak  
 267 and Levin, 2000 ; Mavromatidis and Kallos, 2003 ; Saleeby and Cotton, 2004 ; Van

268 den Heever et al., 2006 ; Van den Heever and Cotton, 2007 ; Mavromatidis et al.,  
269 2007 ; Zhang et al., 2007 among others). The model is able to explicitly resolve a  
270 complete set of atmospheric processes at resolutions ranging from tens of kilometers  
271 down to a few meters. The nesting capabilities of the model allow for sufficient  
272 representation of microphysical processes at cloud scales. RAMS includes eight  
273 categories of hydrometeors (vapor, cloud droplets, rain droplets, pristine ice, snow,  
274 aggregates, graupel and hail). The two-moment microphysics parameterization  
275 scheme treats both the mixing ratio and number concentration of each hydrometeor  
276 (Meyers et al., 1997). Prediction of cloud droplet number concentration is originally  
277 based on air temperature, vertical wind component and on a constant amount of  
278 available CCN. A lookup table has been constructed offline from a detailed bin-parcel  
279 model and the number of activated CCN is calculated from this table. The size and  
280 chemical properties of the CCN are not taken into consideration. This approach has  
281 been altered in the new version of the model with the addition of an explicit cloud  
282 droplet nucleation parameterization scheme (Nenes and Seinfeld, 2003; Fountoukis  
283 and Nenes, 2005). This scheme (referred to as FNS), provides a comprehensive  
284 microphysical link between aerosols and clouds. FNS computes droplet number based  
285 on the parcel framework, and solves for the maximum supersaturation,  $s_{max}$ , that  
286 develops given a set of cloud-scale dynamics (temperature, pressure and vertical wind  
287 component) and aerosol properties (number concentration, size distribution and  
288 chemical composition). The droplet number is then equal to the number of CCN with  
289 critical supersaturation less or equal to  $s_{max}$  (Nenes and Seinfeld, 2003). The water  
290 vapour uptake coefficient, used in calculating the mass transfer coefficient of water  
291 vapour to growing droplets (Fountoukis and Nenes, 2005), is set to 0.06 based on in-  
292 situ cloud droplet closure experiments (Meskhidze et al., 2005; Fountoukis et al.,  
293 2007).

294 Soil dust, sea salt spray and secondary pollutants contribute to the CCN population.  
295 Dust particles are assumed to follow a lognormal size distribution at source regions.  
296 The properties of these distributions (number mean diameter and geometric  
297 dispersion) are expected to change throughout their atmospheric lifetime. These  
298 properties are explicitly calculated at every model step based on the predicted dust  
299 concentration (Shultz et al., 1998). CCN concentrations are expressed as a function of  
300 supersaturation using Köhler theory (Köhler, 1936; Nenes and Seinfeld, 2003).

301 Freshly-emitted mineral dust particles have long been known to act as effective ice  
302 nuclei (Pruppacher and Klett 1997; DeMott et al., 2003, Levin et al., 2005). Ice  
303 production is generally facilitated over regions with high mineral dust concentrations,  
304 such as over the Atlantic Ocean during African dust transportation episodes (Astitha  
305 et al., 2010). In ICLAMS, the insoluble fraction of dust contributes to the prognostic  
306 ice-forming nuclei (IFN) following the formulation of Meyers et al., (1992).

307

### 308 *3 Clouds and precipitation in an environment with natural particles*

#### 309 *3.1 Idealized simulations*

310 In order to examine the sensitivity of the new cloud nucleation scheme to aerosol  
311 properties, we performed a set of “idealized” simulations for a convective cloud  
312 system over flat terrain. The model was configured on a two-dimensional domain  
313 with horizontal uniform resolution of 300m and 35 vertical levels, starting from 50m  
314 spacing near the ground and extending up to 18km with a geometric stretching ratio of  
315 1.2. The horizontal dimension of the domain was 24km. The model was initialized  
316 from a convectively unstable sounding (Figure1) that is considered as representative  
317 of winter weather type for the eastern Mediterranean region (Yin et al., 2002; Levin et  
318 al., 2005). Initial wind conditions of 3 m s<sup>-1</sup> wind speed and a western wind direction  
319 were applied homogeneously over the domain. The FNS parameterization was  
320 invoked in every time step and grid point and the number of activated droplets was  
321 calculated from grid-cell aerosol, P, T, and updraft velocity. All tests were performed  
322 with exactly the same configuration except for the aerosol properties. Each run started  
323 at 12:00 UTC and lasted for six hours.

324 Two scenarios were considered for the initial distribution of aerosol concentration,  
325 namely the “pristine” and the “hazy” scenario as illustrated in Figure 2. The “pristine”  
326 scenario is representative of a remote area with a relatively clean atmosphere of total  
327 particle concentration 100 cm<sup>-3</sup>, while the “hazy” scenario assumes a total  
328 concentration of 1500 cm<sup>-3</sup> similar to Teller and Levin (2006). Such high aerosol  
329 concentrations can be found near urban areas or industrial zones and are also typical  
330 during intense dust episodes. The size distribution of the particles was considered to  
331 follow a bimodal lognormal distribution that does not change shape between  
332 scenarios. The geometric standard deviation equals two for both modes, while the

333 number median diameter was set at 0.2  $\mu\text{m}$  for the first mode and at 2  $\mu\text{m}$  for the  
334 second mode. The chemical composition for the soluble fraction of the particles was  
335 assumed to be ammonium sulfate and the aerosol field was applied homogeneously  
336 throughout the model domain. Further development of the cloud system and the final  
337 amount of precipitation depend on the cloud microphysical structure and on the  
338 interplay with ambient dynamics. Both runs started developing a similar cloud  
339 structure after 80 minutes of simulation time and, as seen in Figure 3, two distinctive  
340 convective areas were identified within a horizontal distance of about 15 km.  
341 However, after the initial development, the cloud properties varied significantly  
342 between the “pristine” and “hazy” scenarios. These changes are reflected in the hourly  
343 accumulated precipitation over the entire domain and in the maximum values of  
344 mixing ratios and number concentrations for cloud droplets, rain droplets and pristine  
345 ice particles that are summarized in Table 3 for each model run.

346 In the “pristine” simulation, the cloud droplets number concentration remained low  
347 (maximum of 130  $\text{cm}^{-3}$ ) throughout the simulation. Fewer CCN had to compete for  
348 the same amount of water. So, large cloud and rain droplets were allowed to develop  
349 and the collection efficiency was enhanced. This allowed for increased  
350 autoconversion rates of cloud to rain droplets and early initiation of warm rain  
351 process. Intense precipitation started 100 minutes into the simulation, with  
352 precipitation rates reaching as high as 15  $\text{mm h}^{-1}$  (Figure 4). The high rain mixing  
353 ratio peak value (0.47  $\text{g kg}^{-1}$ ) and corresponding rain drop number concentration of  
354 27.65  $\text{l}^{-1}$  indicate the dominance of collision-coalescence during the early stages of the  
355 cloud.

356 In contrary, the “hazy” clouds suppressed precipitation at the early cloud stages. The  
357 number of cloud droplets was extremely high, especially during the first two hours of  
358 cloud development and reached 2133  $\text{cm}^{-3}$  after 120 minutes of run. As a result, the  
359 conversion rates of cloud droplets to rain droplets were very low and precipitation was  
360 inhibited. Maximum precipitation rate at this stage was only 4  $\text{mm h}^{-1}$  which is about  
361 4 times less than the “pristine” scenario. However, pristine ice particles were almost  
362 double that of the “pristine” cloud and rain droplets coming from the melting of ice  
363 condensates produced a significant amount of rain between 150 and 210 minutes  
364 model time as seen in Figure 4. The accumulated precipitation over the entire domain

365 was 286 mm for the “pristine” and 215 mm for the “hazy” case. Most of this  
366 difference can be attributed to the inhibition of precipitation during the early stages of  
367 cloud development as illustrated in Figure 5. Cloud structure was also very different  
368 between the two simulations. This is clearly shown in Figure 3; two separate cloud  
369 systems were still distinct after 170 minutes of simulation for the “pristine” case while  
370 during the “hazy” case the two clouds had merged to one cell. The merged system  
371 contained increased amounts of ice elements and continued precipitating with slower  
372 rates until the end of the simulation. Melting of ice hydrometeors enhanced  
373 precipitation for the “pristine” clouds after the system is well developed (Figure 4).

374 The impact of gigantic cloud condensation nuclei (GCCN) is also important for cloud  
375 processes and precipitation (Teller and Levin, 2006; van den Heever et al., 2006).  
376 When aerosol sizes are comparable to cloud droplet size - which is often the case for  
377 dust and sea-salt, kinetic limitations are imposed on cloud nucleation processes  
378 (Barahona et al., 2010). Neglecting such effects may result in significant  
379 overestimation of activated cloud droplet number and in reduction of precipitation  
380 rates. Nucleation of GCCN is parameterized in the model according to Barahona et  
381 al., (2010). In order to examine the impact of GCCN on precipitation, we performed  
382 another couple of tests by adding a third coarser mode to the aerosol distribution. The  
383 third mode was assumed to have a median diameter of  $10\mu\text{m}$  a standard deviation of 2  
384 and a total concentration of  $5\text{ cm}^{-3}$ . Adding GCCN to a hazy environment limited the  
385 number of cloud droplets that nucleated and as seen in Figure 6b the rainfall during  
386 the early stages of cloud development was increased. In contrary, GCCN did not  
387 change significantly the warm stage precipitation for the pristine environment (Figure  
388 6a) because the clean clouds have some large CCN anyhow and the small number of  
389 CCN makes them all grow fast.

390 Cloud processes are of course sensitive to several other model parameters and there  
391 are more combinations of cases that could be performed. However, these results are  
392 always limited to calculations of single idealized clouds and do not represent real  
393 conditions. For example, by adding topographic effects in a 3-D model configuration  
394 that is equivalent to the 2-D “pristine” and “hazy” model simulations resulted in  
395 substantially different spatial distribution of precipitation as shown in Figure 7.  
396 During these simulations, all model parameters remained unchanged except the

397 surface features (topography in this case). The same initial conditions as in previous  
398 runs were used (Figure 1) and a western flow with initial wind speed of  $3 \text{ ms}^{-1}$  was  
399 considered for all runs. The impact of topography on precipitation was investigated  
400 for three cases, namely “flat terrain”, “idealized hill” and “complex hilly area”. The  
401 first case (flat terrain) considers no topographic features and uniform landscape (soil  
402 and vegetation classes). In this case, atmospheric stability and cloud microphysics are  
403 the governing factors for the evolution of the cloud system. As seen in Figures 7a, b,  
404 most of the precipitation was distributed over the western side of the domain for both  
405 “pristine” and “hazy” clouds but with different maxima (“pristine” case gave more  
406 precipitation). For the second run (“the idealized hill”) the landscape remains the  
407 same as in the previous case but a 290 m high ridge with a N-S uniform orientation is  
408 added at the center of the domain. The combination of microphysics and cloud  
409 dynamics due to mechanical elevation over the hill resulted in a substantially different  
410 precipitation pattern that is shown in Figures 7c, d. The distribution of precipitation  
411 for this case is clearly related to the location of the hill with more rain falling over the  
412 downwind area at the eastern part of the domain. Finally, the third case includes also  
413 the same landscape but the topography is representative of a complex hilly area. As  
414 illustrated in Figures 7e, f, these topographic features resulted in a completely  
415 different distribution of precipitation. Such results indicate that the synergetic effects  
416 between the microphysical and macrophysical parameters that contribute in cloud and  
417 precipitation processes should be taken into account in relevant modeling studies on a  
418 combined way. Otherwise, the results may be misleading when compared to real  
419 atmospheric conditions. For example, the “pristine” cases produced overall more  
420 precipitation than the “hazy” ones but the distribution of precipitation was found to be  
421 much more sensitive to terrain variability than to any of the variations in aerosol  
422 properties.

423 The next section will focus on the use of ICLAMS in a fully coupled mode of air  
424 quality and meteorology for a specific test case at the regime of Eastern  
425 Mediterranean. The FNS explicit cloud droplets nucleation scheme of the model is  
426 used to provide an extra link between cloud processes and prognostic airborne  
427 particles, such as mineral dust, sea-salt, sulphates and nitrates.

428

429        *3.2 Case study*

430 We focus on a case study that combines a low pressure system and a dust storm over  
431 the eastern Mediterranean. On 28 January 2003, a cold cyclone moved from Crete  
432 through Cyprus accompanied by a cold front. A second air mass transported dust  
433 particles from NE Africa towards the coast of Israel and Lebanon. The two air masses  
434 interacted over the sea, triggering deep convection as illustrated in Figure 8. These  
435 clouds moved northeasterly and on 29 January 2003, heavy rain and hail dispersed  
436 over the East Mediterranean coastline and a few kilometres inland. Flood events and  
437 agricultural disasters were reported. A detailed analysis and airborne measurements of  
438 this episode were obtained during the Mediterranean Israeli Dust Experiment  
439 (MEIDEX) as described in Levin et al., 2005. Several runs have been performed for  
440 this case. Special attention was given to the amount of available airborne particles that  
441 could act as CCN and GCCN for each particular case, examining both the effects on  
442 the precipitation reaching the ground and also the effects on the microphysical  
443 structure inside the clouds. Modelling results are compared to ground and aircraft  
444 observations and some of the findings are discussed here.

445 The model was configured with three nested grids (15km, 3km and 750m) as seen in  
446 Figure 9 and with 32 vertical levels starting from 50m above ground and stretching up  
447 to 18km with a geometric ratio of 1.2. For the initial and boundary conditions, a high  
448 resolution reanalysis dataset was used. This dataset has been prepared with the Local  
449 Analysis and Prediction System (LAPS) (Albers, 1995; Albers et al., 1996). LAPS is  
450 a fully integrated, meso- $\beta$ -scale data assimilation and analysis system designed to  
451 handle all types of meteorological observations. It uses an effective analysis scheme  
452 to harmonize data of different temporal and spatial resolutions on a regular grid.  
453 LAPS surface and upper air fields can then be used as initial and boundary conditions  
454 in local forecast models. The prepared dataset includes 24 years of reanalysis (1986-  
455 2009) with a grid resolution of 15 $\times$ 15 km and temporal intervals of 3 hours. It is based  
456 on the ECMWF operational analysis dataset (with resolution of 0.5 $\times$ 0.5 degrees as  
457 initial guess fields and the utilization of all available surface and upper air  
458 measurements. The sea surface temperature (SST) used is the NCEP 0.5 $\times$ 0.5 $^\circ$   
459 analysis. During the simulation experiments, two main dust sources were identified:  
460 One is located at North East Libya (Gulf of Sidra) and the second at North West

461 Egypt (Qattara Depression). These areas are illustrated in Figure 9. The chemical  
462 properties of dust particles are associated with their origin and with the chemical  
463 transformations that occur during their atmospheric lifetime. Aged dust clouds include  
464 particles that are coated with sea-salt or sulfates that increase their hygroscopicity and  
465 CCN efficiency (Levin et al., 1996, Bougiatioti et al., 2009).

466 Consistent with Levin et al., (2005), the aerosol particles within the lowest two  
467 kilometers of the atmosphere were a mixture of dust and sea-salt. As illustrated in the  
468 3D model plots of Figure 10, dust and sea-salt particles were present all along the  
469 frontal line, near the cloud base and the clouds that were formed in this area were  
470 highly affected by this increased aerosol concentrations. The south-to-north and west-  
471 to-east vertical cross sections of Figure 11 indicate that the dust particles did not  
472 elevate higher than two kilometers in the atmosphere and coexisted with sea-salt spray  
473 particles along their transportation path. The location of the cross sections is shown in  
474 Figure 9.

475 The number concentration of modelled dust and sea salt particles was tested against  
476 in-situ aircraft observations that were performed (between 7:30 and 9:30 UTC) at  
477 various heights inside the dust-storm area. Detailed information about the aircraft  
478 instrumentation, the sampling and averaging techniques and the variability in particle  
479 measurements is provided in Levin et al., (2005). According to their description, the  
480 aircraft was flying with a constant speed of  $70 \text{ m s}^{-1}$  and covered a distance of about  
481 125 kilometres inside the stormy area. Two optical particle counters were used for the  
482 measurements of the aerosol size distributions and concentrations. The first was used  
483 for aerosols between  $0.1\text{-}3 \mu\text{m}$  in diameter and the second for aerosols between  $2\text{-}16$   
484  $\mu\text{m}$  in diameter. Measurements were performed in an irregular manner along the  
485 aircraft path and each sampling period lasted for five minutes. For the current work,  
486 the average concentration of natural particles for each measurement point is compared  
487 towards the corresponding model results. In order to co-locate the aircraft  
488 measurements with the appropriate model grid point we considered the average  
489 location along the aircraft track. The simulated dust and salt concentrations for the  
490 corresponding location in the model is then calculated as the weighted average from  
491 the eight nearest model grid points. The concentrations of modelled particles inside  
492 the dust layer are in satisfactory agreement with airborne measurements as illustrated

493 in Figure 12, with a correlation coefficient  $R=0.89$ . These results indicate that the  
494 model is able to reproduce the horizontal and vertical structure of the dust storm  
495 qualitatively and quantitatively. The ability of the model to capture these features is  
496 important for the next steps of the experiment, since the estimated particles contribute  
497 to the cloud droplet nucleation mechanism. As seen in Figure 13, the dust storm is  
498 approaching the coast of East Mediterranean from south-west during the morning of  
499 28 January 2008. The clouds that were formed in this area were affected by the dust  
500 storm and also by the increased sea salt production due to the relatively strong winds  
501 and wind shear. The coexistence of salt and dust particles at heights below cloud base  
502 provided significant amounts of highly hygroscopic mixed particles. These clouds  
503 contained increased numbers of cloud droplets and moved north-northeast towards the  
504 Israel and Lebanon coast. Most of the precipitation from these clouds fell after they  
505 reached land and the peak rain rates were reported on the morning of 29 January 2003  
506 around Haifa and North Israel.

507 Three different scenarios related to the properties of the aerosol particles during the  
508 model runs are discussed here. All model parameters were held constant except the  
509 percentage of dust particles containing soluble material, thus becoming effective  
510 CCN. In experiment 1 (EXP1), 5% of dust particles were hygroscopic while for  
511 experiment 2 (EXP2), this percentage was increased to 20%. DeMott et al. (2003a),  
512 found that during intense dust episodes, the concentration of ice nuclei (IN) was  
513 increased by 20-100 times compared to non-dust environment. Following this  
514 approach, EXP3 incorporated 5% hygroscopic dust while the concentration of IN in  
515 the model was multiplied by a factor of ten in the presence of mineral dust. The  
516 hygroscopic dust particles were assumed to contain 33% sea-salt, as in Levin et al.,  
517 (2005) where the airborne measurements were analyzed. The aerosol spectrum was  
518 fitted in a 3-modal lognormal distribution. The first mode corresponds to particles of  
519 mineral dust origin while the other two correspond to sea-salt accumulation and  
520 coarse modes, respectively. The median diameter and standard deviation for the  
521 accumulated salt mode is  $0.36 \mu\text{m}$  and 1.8 respectively. For the coarse salt mode the  
522 median diameter is  $2.85 \mu\text{m}$  and the standard deviation is 1.9. However, dust particles  
523 may range from small submicron diameters (away from the sources and at high  
524 atmospheric levels) up to GCCN sizes (near sources and at low atmospheric levels).  
525 In order to represent this kind of inhomogeneous spatial distribution, the median

526 diameter and standard deviation for the dust mode are estimated from the relative  
527 prognostic concentrations of the eight dust bins according to Schultz et al. (1998).

528 Increasing the percentage of hygroscopic dust particles from 5% to 20% increased  
529 also the concentration of small liquid droplets inside the cloud. This resulted in lower  
530 autoconversion rates of cloud to rain droplets and significant amount of water was  
531 transferred above freezing level. The EXP2 clouds reached higher tops, included more  
532 ice water content and the initiation of rainfall was in general delayed by almost 1  
533 hour. In Figure 14, the less polluted cloud (EXP1) reached the maximum top at 9:00  
534 UTC. The EXP2 cloud extended much higher (about 3km higher than EXP1),  
535 contained more ice, and eventually produced more rain (one hour later than EXP1;  
536 10:00 UTC instead of 9:00 UTC). The EXP3 cloud also exhibited significant vertical  
537 development, with a structure and precipitation amounts similar to that of EXP2.

538 Mineral dust particles affect the microphysical structure of the clouds by acting both  
539 as CCN, GCCN and IN and also they interact with cloud dynamics. As illustrated in  
540 Figure 15a for the EXP2 case, there was significant convective activity over Haifa at  
541 8:20 UTC on 29 January and the convective available potential energy (CAPE) was  
542 1027 J. When CAPE is large enough, significant amounts of liquid condensates may  
543 thrust into the upper levels of a cloud and eventually freeze in higher altitudes. The  
544 released latent heat invigorates convection at higher levels. As seen in Figure 15b, a  
545 significant amount of liquid condensates were transported above the freezing level.  
546 The release of latent heat due to the glaciation of these supercooled droplets had as a  
547 result the increase of equivalent potential temperature. This procedure is evident at  
548 Figure 15c and is indicated with the arrow pointing the area of increased equivalent  
549 potential temperature. After 10 minutes, strong updrafts reached up to 8 kilometers  
550 height and transferred condensates to the upper cloud layers as illustrated in Figure  
551 15d. These condensates interact with the available IN in this area of the cloud for the  
552 formation of ice particles through heterogeneous icing processes. Thus, increasing the  
553 percentage of hygroscopic mineral dust or increasing the IN by an order of magnitude  
554 resulted in enhancement of ice particles formation and therefore release of latent heat  
555 at higher levels. These interactions between aerosols and cloud dynamics produce  
556 clouds with stronger updrafts that reach higher tops and finally produce heavier  
557 rainfall.

558 In order to examine the sensitivity of accumulated precipitation to aerosol properties,  
559 we performed a total of nine scenarios with the same model configuration but  
560 changing the chemical composition of airborne particles. The physio-chemical  
561 characteristics used on each run are shown in Table 4. The first run was performed  
562 with the original RAMS model and we call it “control run”. For Case2, only particles-  
563 radiation interaction was enabled. For cases three and four, the FNS cloud nucleation  
564 parameterization was enabled using the prescribed “pristine” and “hazy” air mass  
565 types as in section 3.1. For the next four runs, particle concentration was a prognostic  
566 variable and the cloud nucleation scheme was used in an explicit way. The percentage  
567 of hygroscopic dust was set to be one (1%), five (5%), ten (10%) and thirty per cent  
568 (30%) respectively. For Case 9, we considered five (5%) hygroscopic dust and also  
569 the IN concentration was increased by a factor of ten similar to Levin et al., (2005).  
570 The modelled 24-hour accumulated precipitation on 29 January 2003 for all nine  
571 cases was tested against ground measurements from 86 measuring stations over North  
572 Israel.

573 The model bias score (see Appendix) was calculated for nine thresholds of  
574 accumulated precipitation, namely 0.5 mm, 2 mm, 4 mm, 6 mm, 10 mm, 16 mm, 24  
575 mm, 36 mm and 54 mm. The results for each case and each precipitation threshold are  
576 shown in Figure 16. Biases equal to one mean that the particular precipitation  
577 threshold was simulated as often as observed. Bias below unity indicates model  
578 underprediction and bias over one indicates overprediction. The limited time period of  
579 the study and the relevant small number of measuring stations (especially at high  
580 thresholds) does not allow extracting robust statistical results. However, as seen in  
581 Figure 16, the accumulated precipitation was found to be very sensitive to variations  
582 of the percentage of dust particles that can be activated as CCN and IN. These results  
583 indicate the need for a proper treatment of the links and feedbacks between cloud and  
584 aerosol processes. Model results with prognostic aerosol treatment were in general  
585 closer to the observations than those of the control run and the model bias for these  
586 cases was improved by almost 40% for some thresholds especially at medium and  
587 high precipitation heights. Also, assuming a constant prescribed air mass type as in  
588 cases three and four, did not improve the model results. The average bias for all  
589 thresholds was calculated for each one of the nine cases (see Appendix for definition)  
590 and is illustrated in Figure 17. The significant variability in model results that is

591 related to aerosol properties is indicative of their role in atmospheric processes. Cases  
592 one to four exhibited more or less the same statistical performance that is probably  
593 explained from the use of constant prescribed air mass properties for these runs.  
594 However, including the radiative dust effects (Case2) slightly improved the model  
595 bias. During the eighth case, the accumulated precipitation field was clearly  
596 underestimated due to the increased concentration of hygroscopic particles for this  
597 case. Increasing the number of CCN delayed the initiation of precipitation and  
598 resulted in the enhancement of ice concentrations. These ice crystals did not grow  
599 much because of the lack of water drops at higher levels. Most of these clouds  
600 evaporated before they managed to precipitate and the accumulated precipitation was  
601 underestimated. The model results were significantly improved for the remaining  
602 prognostic aerosol cases (five, six, seven and nine) with average biases of 0.84, 0.84,  
603 0.96 and 0.94 respectively. These findings imply that a more detailed representation  
604 of the atmospheric composition and of the aerosol - cloud – radiation feedbacks can  
605 provide some insight in the processes involved in the formation of clouds and  
606 precipitation and also improve the model performance.

607

#### 608 *4 Concluding remarks*

609 Aerosol partitioning (anthropogenic/natural) and perturbations on it, such as aging  
610 particles, have significant impacts on cloud structure and spatial and temporal  
611 distribution of precipitation. Therefore, there is a need for quantification of this  
612 forcing at the regional scale since the impacts mentioned above have significant  
613 feedbacks with net results not easily quantified. There is still significant amount of  
614 uncertainty regarding the aerosol – cloud interaction mechanisms and especially the  
615 formation of IN. Therefore there is a need for extensive regional, mesoscale and  
616 microscale model simulations with detailed physical and chemical parameterization  
617 together with detailed cloud microphysics in order to understand some of the links  
618 and feedbacks between air quality and meteorology.

619 Several sensitivity tests were performed with an integrated atmospheric model that  
620 includes online parameterization of aerosol processes, aerosol-radiation interaction,  
621 explicit cloud droplet activation scheme and a complete microphysics package. Two-  
622 dimensional tests for an idealized case of cloud development indicated a significant

623 response of cloud processes and precipitation to the variations of aerosol number  
624 concentration and also to the size distribution of the particles. “Hazy” clouds  
625 suspended precipitation while “pristine” clouds precipitated faster and produced more  
626 rain that is in agreement with earlier publications. However, in order to simulate the  
627 aerosol effects in an approach that is closer to real atmospheric conditions, it is  
628 necessary to take also into account the synergetic effects between the various  
629 microphysical and macrophysical processes. For example, the distribution of  
630 accumulated precipitation was found to be much more sensitive to topographic  
631 variations than to aerosol number concentration and/or composition.

632 A second application for a specific event of dust transportation and convective  
633 activity over Eastern Mediterranean, illustrated that this kind of regional modeling  
634 approach can be very useful in reproducing many of the important features of aerosol  
635 and cloud processes. These findings can be summarized as follows:

- 636 1. The meteorological conditions during this particular event and the aerosol  
637 field properties were reproduced in the model in satisfactory agreement with  
638 observations. This is also indicated by the correlation factor of 0.89 between  
639 modelled aerosol concentrations and airborne measurements.
- 640 2. An increase of 15% in the concentration of soluble dust particles produced  
641 clouds that extended about three kilometres higher and the initiation of  
642 precipitation was delayed by almost one hour.
- 643 3. Variations between 1-30% in the amount of dust particles that were assumed  
644 to contain soluble material resulted in significant changes in cloud properties.  
645 The associated variations in the precipitation bias score were up to 80% for  
646 some thresholds.
- 647 4. In general, online treatment of dust and salt particles as prognostic CCN,  
648 GCCN and IN improved the average bias score for the 24h accumulated  
649 precipitation by almost 7% in comparison to the runs considering a uniform  
650 atmospheric composition all over the domain.

651 These results illustrate the highly non-linear response of precipitation to aerosol  
652 properties and indicate that a large portion of uncertainty remains unresolved. This  
653 study focuses mostly on investigating the mechanisms that are associated with the

654 aerosol cloud interactions for a specific event. Therefore it is not possible to extract  
655 generic results. Nevertheless, this work represents one of the first limited area  
656 modelling studies for aerosol-cloud-radiation effects at the area of Eastern  
657 Mediterranean and could be used as a base for future improvements and longer term  
658 studies. The role of dust for the weather in Mediterranean is important since dust  
659 particles are almost always present at the area and also interact with other natural or  
660 anthropogenic pollutants. Especially the role of dust in the distribution of precipitation  
661 is more important over areas that suffer from long drought periods such as the Middle  
662 East. More intense combined modeling and observational surveys on the interactions  
663 between airborne particles and cloud processes at regional and local scale are  
664 necessary in order to improve our knowledge on the interactions between atmospheric  
665 chemistry and meteorology.

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670 APPENDIX

671 For each model scenario (Cases 1-9) and for each precipitation threshold a  
 672 contingency table is constructed as follows (Wilks 2006):

673

674

675

676

677

678

679

		Observed	
		Yes	No
Modeled	Yes	a	b
	No	c	d

680 The *a* model-observation pairs mean that both model and observation are over the  
 681 specific threshold and are usually called hits. Similarly, *b* pairs mean that the model is  
 682 over the threshold but observation is below it and are called false alarms; *c* pairs mean  
 683 that the observation is over the threshold but the model is below it and are called  
 684 misses and *d* pairs mean that both model and observation are below the threshold for a  
 685 station and are called correct rejections.

686

687 The total number of hits (*a*), false alarms (*b*) and misses (*c*) for each threshold are  
 688 then used to calculate the MODEL BIAS (B):

689

$$B = \frac{a + b}{a + c}$$

690

691 Unbiased forecasts exhibit bias=1, while bias greater than one indicates  
 692 overprediction and bias less than one indicates underprediction.

693

694 The AVERAGE BIAS ( $\bar{B}$ ) for all precipitation thresholds is calculated as:

695

$$\bar{B} = \frac{1}{N} \sum_{i=1}^{i=N} B(i),$$

696

697 Where  $B(i)$  is the bias for each specific threshold and N is is the total number of  
 precipitation thresholds.

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978 doi:10.1029/2007GL029876, 2007.



Table 1. ICLAMS configuration options. New capabilities (compared to RAMS) are shown in bold.

Basic Equation	<ul style="list-style-type: none"> <li>• Non hydrostatic time split compressible</li> </ul>
Dimensionality	<ul style="list-style-type: none"> <li>• 2 dimensional</li> <li>• 3 dimensional</li> </ul>
Vertical Coordinate	<ul style="list-style-type: none"> <li>• Standard Cartesian coordinate</li> <li>• Terrain following height coordinate</li> </ul>
Horizontal Coordinate	<ul style="list-style-type: none"> <li>• Rotated polar-stereographic transformation</li> <li>• Lambert conformal transformation</li> </ul>
Grid Structure	<ul style="list-style-type: none"> <li>• Arakawa-C grid stagger</li> <li>• Unlimited number of nested grids</li> <li>• User specified space and time step nesting ratios</li> <li>• Ability to add and subtract nests</li> </ul>
Time differencing	<ul style="list-style-type: none"> <li>• Hybrid combination of leapfrog and forward in time</li> </ul>
Turbulence closure	<ul style="list-style-type: none"> <li>• Smagorinsky (1963) deformation K closure scheme with stability modifications made by Lilly (1962) and Hill (1974)</li> <li>• Deardorff level 2.5 scheme – eddy viscosity as a function of TKE</li> <li>• Mellor-Yamada level 2.5 scheme – ensemble averaged TKE (Mellor and Yamada 1982)</li> <li>• Isotropic TKE parameterizations for high resolution simulations</li> </ul>
<b>Cloud microphysics</b>	<ul style="list-style-type: none"> <li>• Warm rain processes</li> <li>• Five ice condensate species</li> <li>• Two-moment bulk scheme (Walko et al., 1995; Meyers et al., 1997).</li> <li>• <b>Cloud droplet activation scheme (Nenes and Seinfeld, 2003; Fountoukis and Nenes, 2005)</b></li> </ul>
Convective Parameterization	<ul style="list-style-type: none"> <li>• Modified Kuo – Tremback (1990)</li> <li>• Kain-Fritsch cumulus parameterization</li> </ul>
<b>Radiation</b>	<ul style="list-style-type: none"> <li>• Chen and Cotton (1983) long/shortwave model – cloud processes considering all condensate as liquid</li> <li>• Harrington (1997) long/shortwave model – two stream scheme interacts with liquid and ice hydrometeor size spectra and <b>with dust particles</b></li> <li>• <b>Rapid Radiative Transfer Model (RRTM) (Mlawer et al. 1997, Iacono et al. 2000) with aerosol radiative effects</b></li> </ul>
<b>Aerosol parameterization</b>	<ul style="list-style-type: none"> <li>• <b>Mineral Dust</b></li> <li>• <b>Sea salt spray</b></li> <li>• <b>Anthropogenic aerosols (primary emissions and chemical formation)</b></li> <li>• <b>Dry deposition</b></li> <li>• <b>Wet deposition</b></li> </ul>
<b>Emissions</b>	<ul style="list-style-type: none"> <li>• <b>Anthropogenic emissions (JRC 0.1°x 0.1° global emissions of CO<sub>2</sub>, NH<sub>3</sub>, CH<sub>4</sub>, SO<sub>2</sub>, NO<sub>x</sub>, CO, N<sub>2</sub>O, VOCs, OC &amp; BC)</b></li> <li>• <b>Biogenic emissions (Gunther et al. 1995)</b></li> <li>• <b>Any other emission inventory or combinations of more than one.</b></li> </ul>
<b>Chemistry parameterization</b>	<ul style="list-style-type: none"> <li>• <b>Online calculation of photodissociation rates</b></li> <li>• <b>Online gas, aqueous and aerosol phase chemistry</b></li> </ul>
Lower boundary	<ul style="list-style-type: none"> <li>• Soil – vegetation – snow parameterization (LEAF-3) (Walko et al, 2000)</li> <li>• Urban canopy scheme – 3D field of drag coefficients based on building characteristics</li> </ul>
Boundary conditions	<ul style="list-style-type: none"> <li>• Klemp and Wilhelmson (1978) radiative condition</li> <li>• Large-scale nudging boundary conditions Davies (1983)</li> <li>• Cyclic or periodic boundaries</li> </ul>
<b>Initialization</b>	<ul style="list-style-type: none"> <li>• Horizontally homogeneous from a single sounding</li> <li>• RAMS/ISAN package – hybrid isentropic terrain following analysis using gridded larger scale model data (ECMWF, NCEP) combined with a variety of observed data types Tremback (1990)</li> <li>• <b>LAPS 3-D data assimilation pre-processing system</b></li> </ul>
Data Assimilation	<ul style="list-style-type: none"> <li>• 4-D analysis nudging to data analysis</li> <li>• Observational data nudging scheme based on ‘direct’ nudging to the observations</li> </ul>

Table 2. Land cover and vegetation type as categorized in LEAF3 scheme

Category number	Vegetation type
0	Ocean
1	Lakes, rivers, streams
2	Ice cap/glacier
3	Desert, bare soil
4	Evergreen needleleaf tree
5	Deciduous needleleaf tree
6	Deciduous broadleaf tree
7	Evergreen broadleaf tree
8	Short grass
9	Tall grass
10	Semi-desert
11	Tundra
12	Evergreen shrub
13	Deciduous shrub
14	Mixed woodland
15	Crop/mixed farming, grassland
16	Irrigated crop
17	Bog or marsh
18	Wooded grassland
19	Urban and built up
20	Wetland evergreen broadleaf tree
21	Very urban

Table 3. Hourly accumulated precipitation over all the domain and maximum values for the number concentration and mixing ratio of cloud, rain and pristine-ice condensates, for two air mass type scenarios.

Time after model start (h)	Air mass type scenario	Total accumulated precipitation (mm)	Cloud No concentration [ $\text{cm}^{-3}$ ]	Cloud mixing ratio [ $\text{g kg}^{-1}$ ]	Rain No concentration [ $\text{L}^{-1}$ ]	Rain mixing ratio [ $\text{g kg}^{-1}$ ]	Pristine-ice No concentration [ $\text{L}^{-1}$ ]	Pristine-ice mixing ratio [ $\text{g kg}^{-1}$ ]
2	PRISTINE	84	130	0.76	27.65	0.47	207	0.13
	HAZY	26	2133	0.48	2.20	0.37	444	0.19
3	PRISTINE	74	99	0.08	3.19	0.22	89	0.05
	HAZY	101	1363	0.39	2.47	0.33	115	0.05
4	PRISTINE	98	22	0.05	1.28	0.14	88	0.05
	HAZY	67	592	0.13	2.98	0.09	97	0.06
5	PRISTINE	23	111	0.08	1.85	0.12	81	0.05
	HAZY	20	377	0.44	2.24	0.09	94	0.05
6	PRISTINE	7	97	0.31	2.97	0.08	109	0.06
	HAZY	3	231	0.12	2.04	0.03	74	0.04

Table 4. Model characteristics for nine aerosol scenarios.

<b>Aerosol Cases</b>	<b>Aerosol-cloud interaction</b>	<b>Aerosol-radiation interaction</b>
<b>Case1</b> (control run)	NO	NO
<b>Case2</b> (only radiation interaction)	NO	YES
<b>Case3</b> (constant air mass – “pristine”)	YES	NO
<b>Case4</b> (constant air mass – “hazy”)	YES	NO
<b>Case5</b> (prognostic air mass - 1% hygroscopic dust)	YES	YES
<b>Case6</b> (prognostic air mass - 5% hygroscopic dust)	YES	YES
<b>Case7</b> (prognostic air mass - 10% hygroscopic dust)	YES	YES
<b>Case8</b> (prognostic air mass - 30% hygroscopic dust)	YES	YES
<b>Case9</b> (prognostic air mass - 5% hygroscopic dust + INx10)	YES	YES

## Figure captions

Figure 1: Initial conditions for the thermodynamic profile of the atmosphere.

Figure 2: Distribution of the available aerosol particles.

Figure 3: Total condensates mixing ratio ( $\text{g kg}^{-1}$ ) for the “pristine” (left column) and the “hazy” (right column) scenarios.

Figure 4: Maximum precipitation rate ( $\text{mm h}^{-1}$ ) for the “pristine” and “hazy” air mass scenarios. Values are taken every 10 minutes.

Figure 5: Hourly accumulated precipitation (mm) over the domain, for the “pristine” and “hazy” CCN scenarios.

Figure 6: a) Maximum precipitation rate ( $\text{mm h}^{-1}$ ) for the “pristine” and “pristine+GCCN” air mass scenarios. b) Maximum precipitation rate ( $\text{mm h}^{-1}$ ) for the “hazy” and “hazy+GCCN” air mass scenarios.

Figure 7: 4h accumulated precipitation (colour palette in mm) and 50m topographic line contours. 1st row: “pristine” aerosol. 2nd row: “hazy” aerosol. 1st column: No topography (flat terrain). 2nd column: artificial obstacle vertical to the general flow. 3rd column: complex topography.

Figure 8: a) Cloud cover percentage (greyscale), near surface streamlines (green contours) and dust - load (red contours in  $\text{mg m}^{-2}$ ). ICLAMS valid 28 January 2003, 1100 UTC. b) MODIS-Aqua visible channel, on 28 January 2003 1100, UTC.

Figure 9: Dust flux in  $\mu\text{g m}^{-2}$  on 27 January 2003 09:00 UTC. Dashed rectangles indicate the location of the nested domains. Solid lines indicate the locations of the cross sections of Figure 11.

Figure 10: a) Isosurface of 90% relative humidity (blue surface) and dust concentration ( $\mu\text{g m}^{-3}$ ) over this surface (colour palette), 28 January 2003 12:00 UTC. b) Isosurface of  $5 \mu\text{g m}^{-3}$  sea-salt concentration (blue) coloured with dust concentration over it (colour palette in  $\mu\text{g m}^{-3}$ ), 28 January 2003 12:00 UTC.

Figure 11: Vertical cross sections of dust concentration (color palette in  $\mu\text{g m}^{-3}$ ) and sea salt concentration (red line contours in  $\mu\text{g m}^{-3}$ ).

Figure 12: Comparison of aircraft measurements of natural particles with modeled dust and salt concentrations inside the dust layer (below 2km). The red line indicates the linear regression line while the dotted line indicates the  $y=x$  line.

Figure 13: Modeled dust number concentration ( $\text{cm}^{-3}$ ) at 538 m height on 28 January 2003, 09:20 UTC. Dots indicate the locations of the aircraft measurements.

Figure 14: West to East cross-section of rain mixing ratio (color palette in  $\text{g kg}^{-1}$ ) and ice mixing ratio (red line contours in  $\text{g kg}^{-1}$ ) at the time of highest cloud top over

Haifa. a) 9 UTC 29 January 2003 assuming 5% hygroscopic dust (EXP1). b) 10 UTC 29 January 2003 assuming 20% hygroscopic dust (EXP2). c) 9 UTC 29 January 2003 assuming 5% hygroscopic dust and INx10 (EXP3).

Figure 15: a) Modelled thermodynamic profile of the atmosphere over Haifa at 08:20 UTC, 29 January 2003. b) Liquid water mixing ratio (colour palette in  $\text{g kg}^{-1}$ ) and ambient temperature (red contours in  $^{\circ}\text{C}$ ) (W-E cross-section over Haifa at 08:20 UTC, 29 January 2003). c) Equivalent potential temperature (colour palette in K) and updrafts (black contours in  $\text{m s}^{-1}$ ) (W-E cross section over Haifa at 08:20 UTC, 29 January 2003). d) Equivalent potential temperature (colour palette in K) and updrafts (black contours in  $\text{m s}^{-1}$ ) (W-E cross section over Haifa at 08:30 UTC, 29 January 2003).

Figure 16: Bias of the 24 hours accumulated precipitation for 86 stations and for nine scenarios of aerosol composition.

Figure 17: Average bias of the 24 hours accumulated precipitation for nine scenarios of aerosol composition.

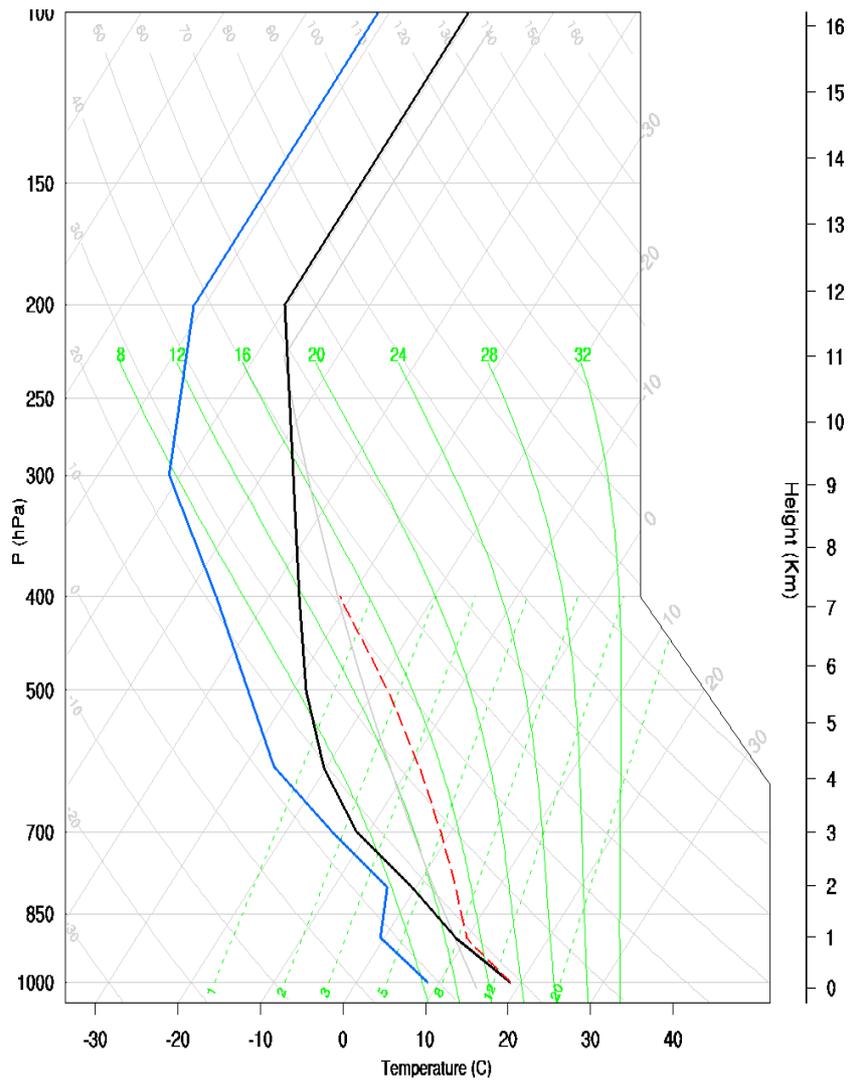


Figure 1

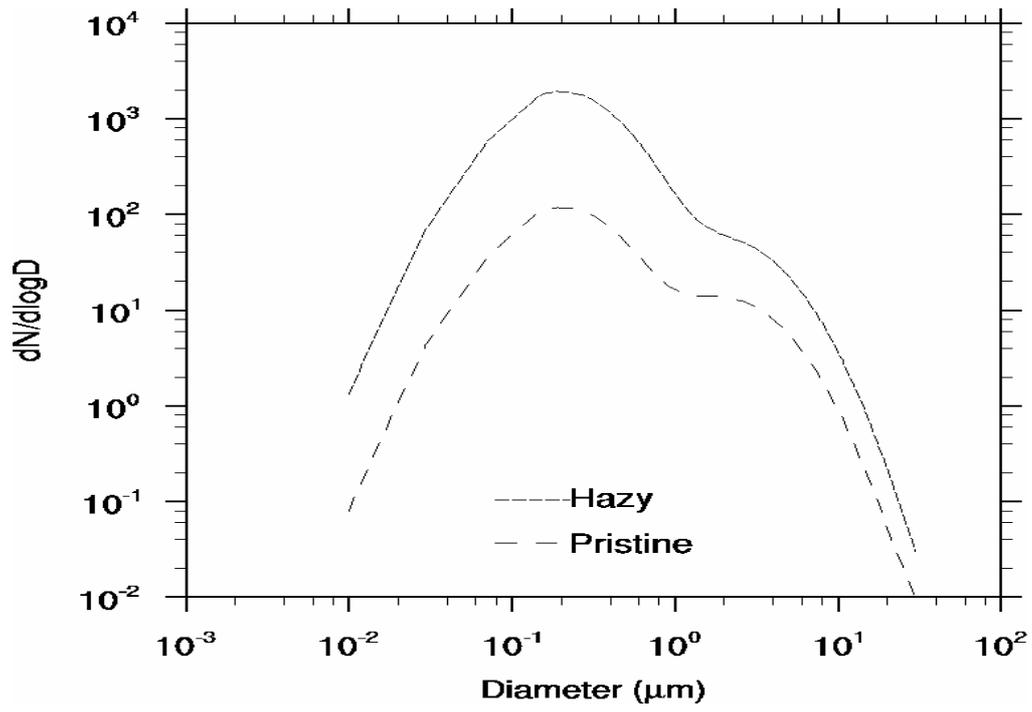


Figure 2

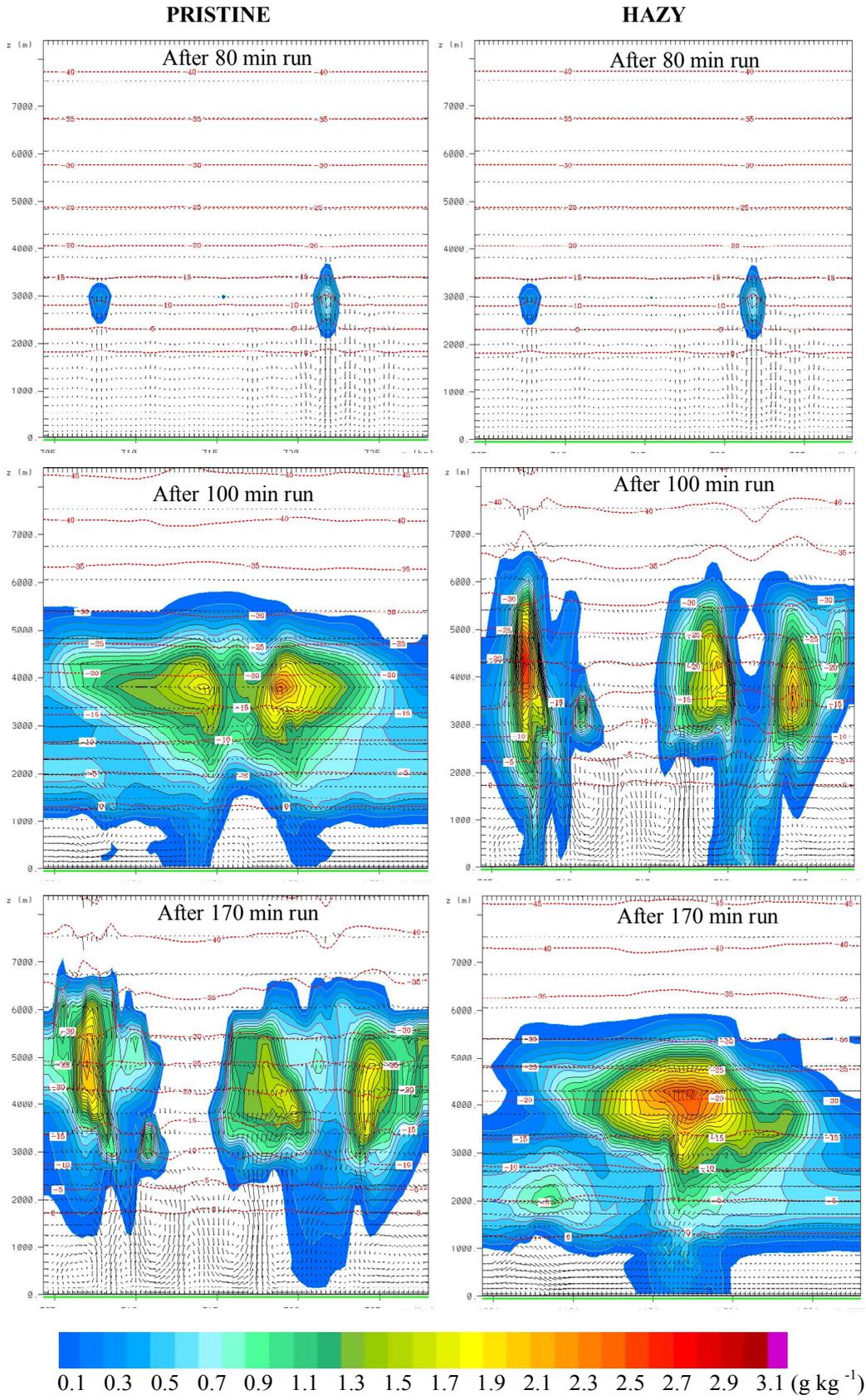


Figure 3

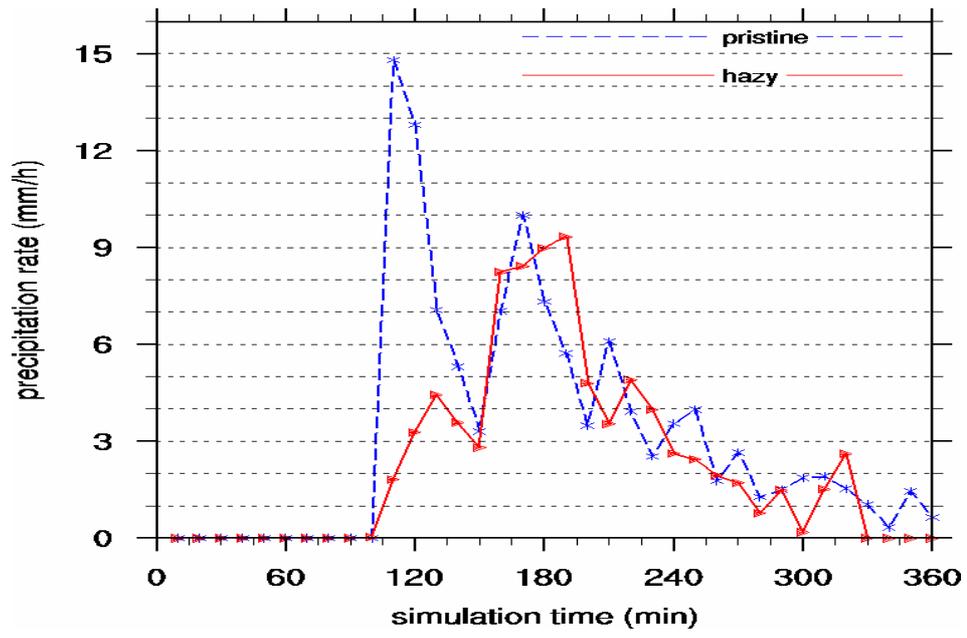


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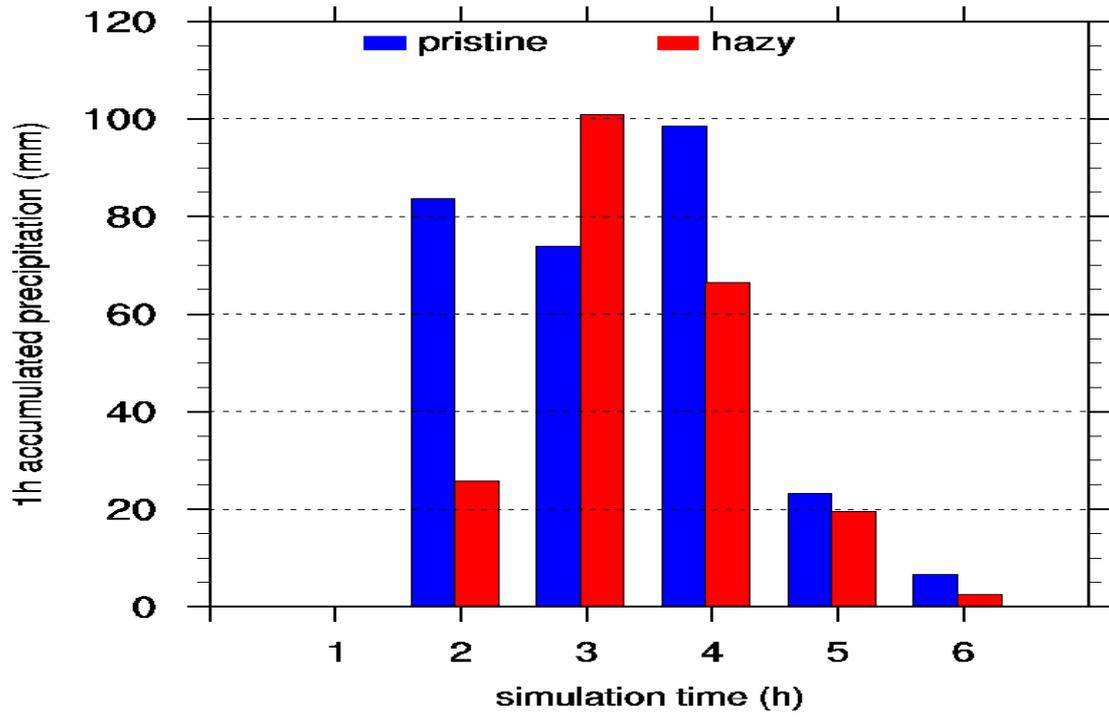


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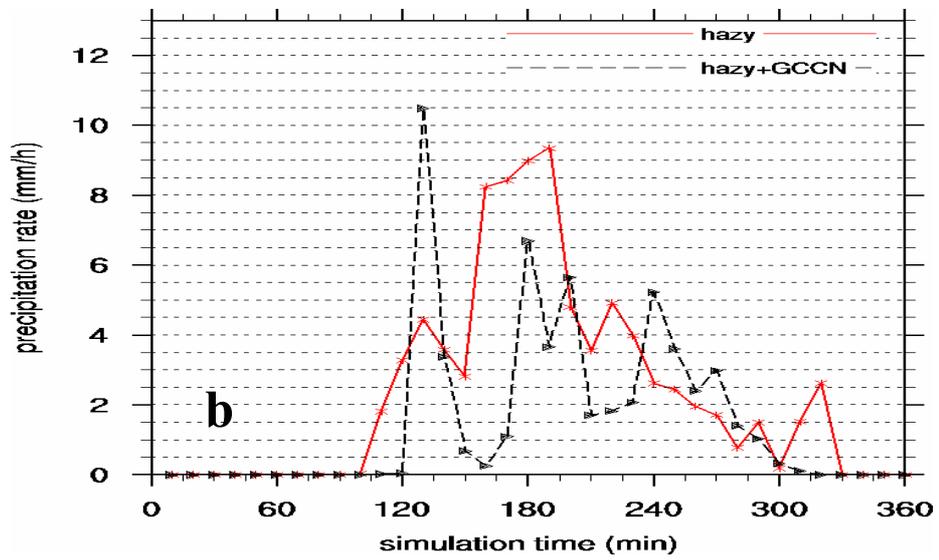
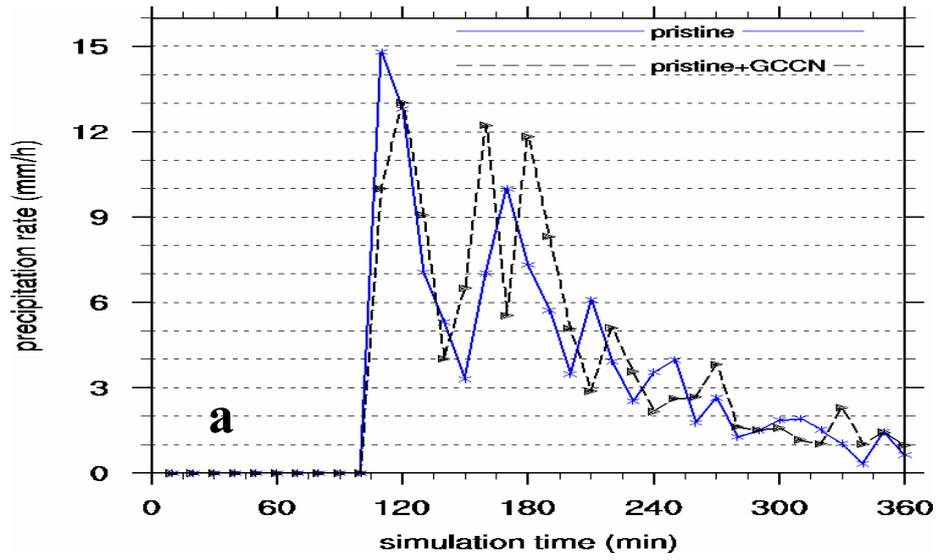


Figure 6

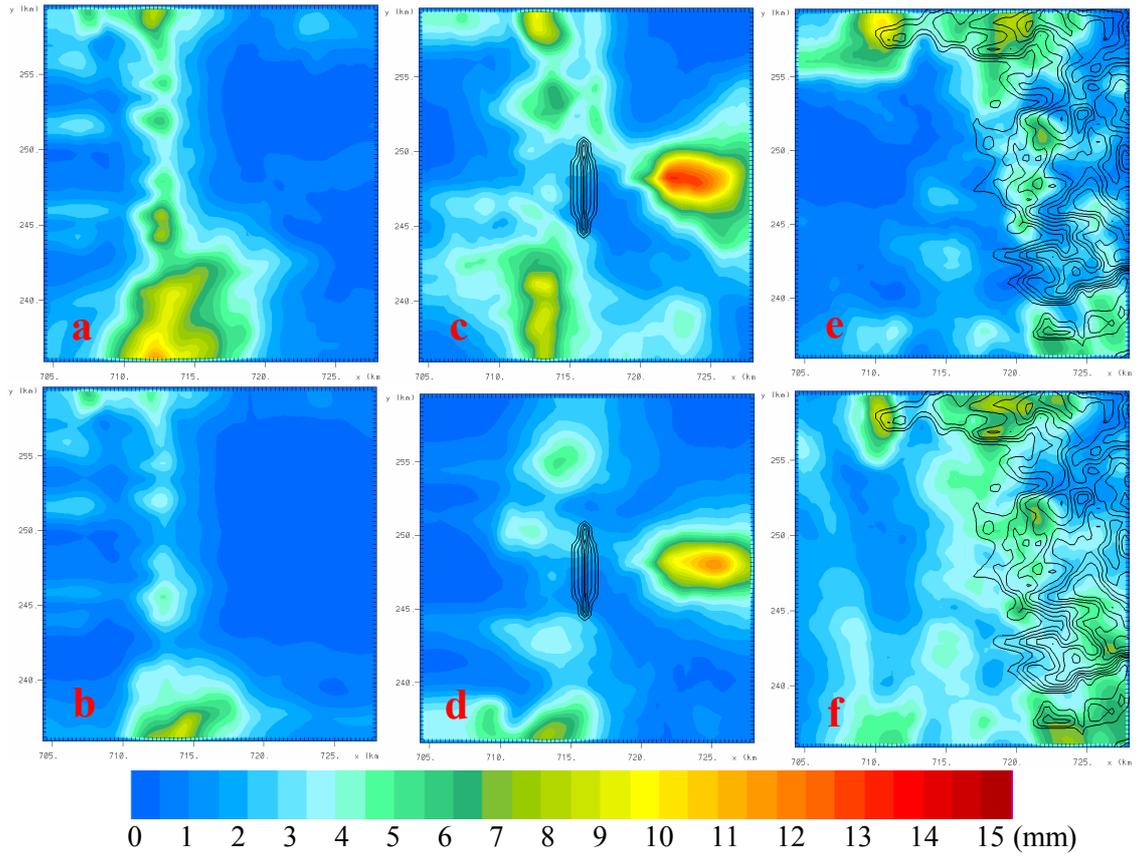


Figure 7

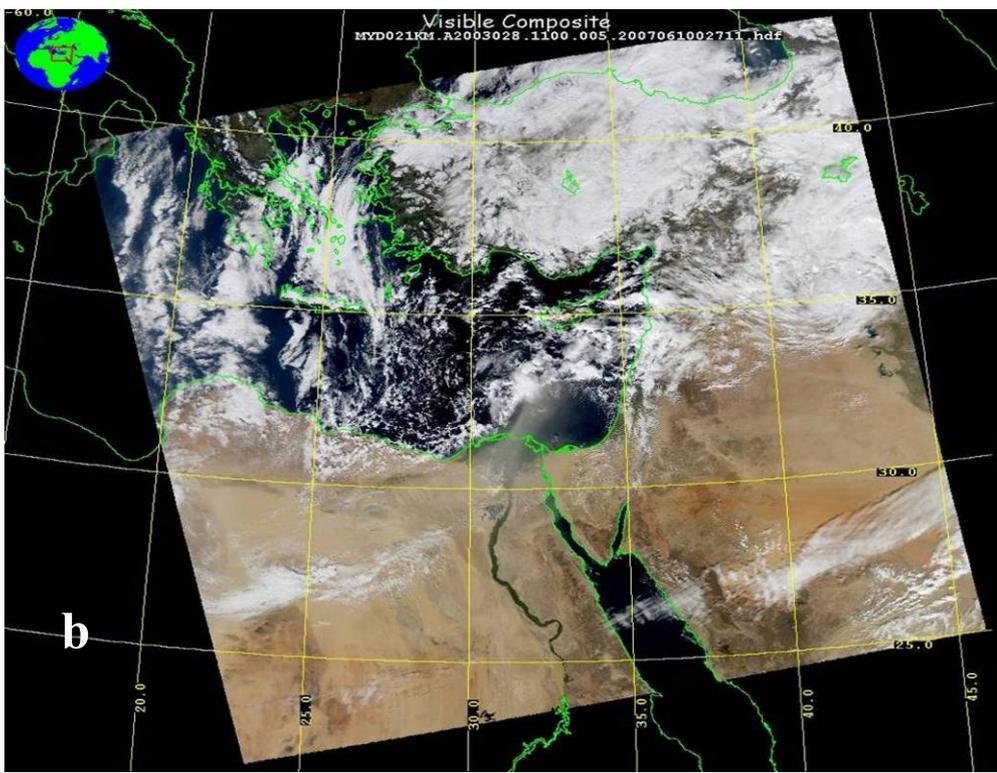
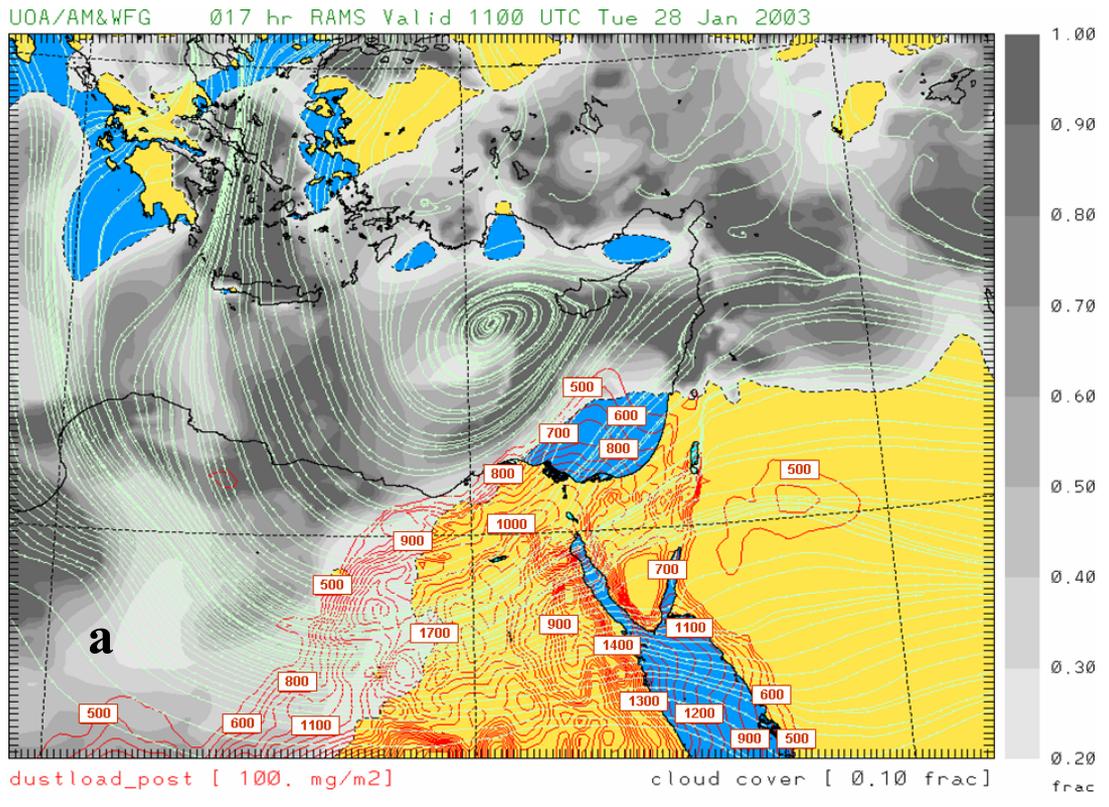


Figure 8

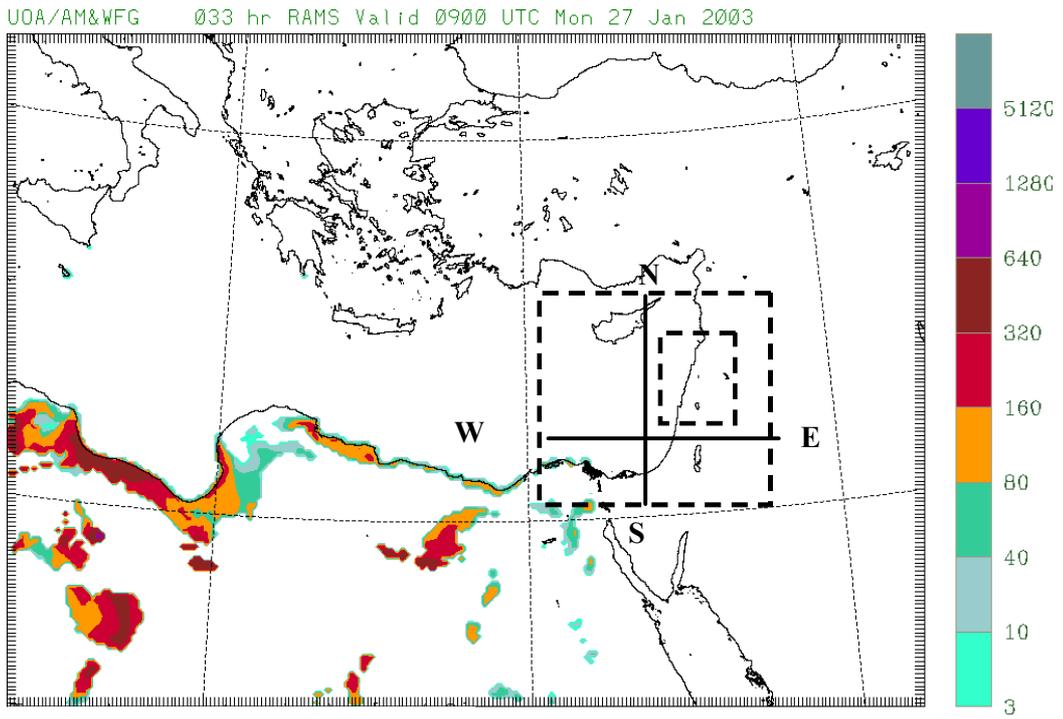


Figure 9

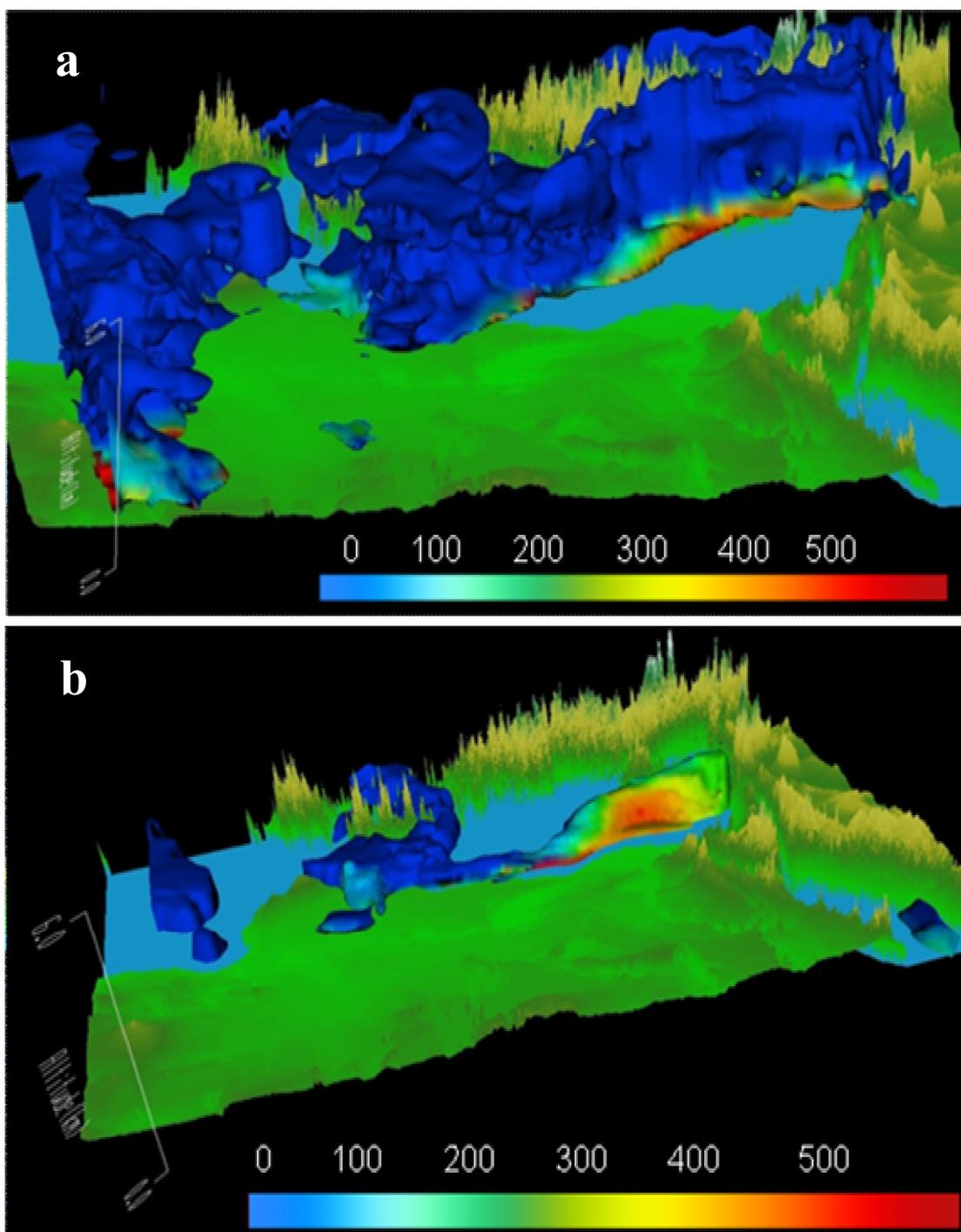


Figure 10

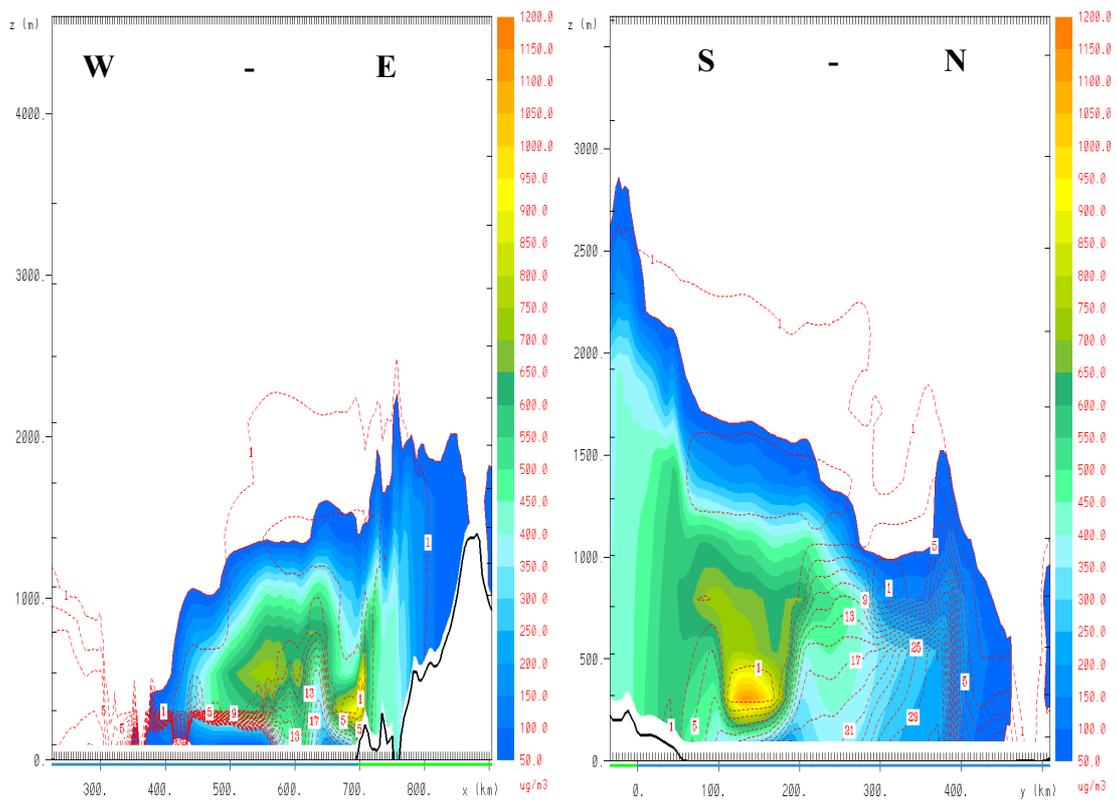


Figure 11

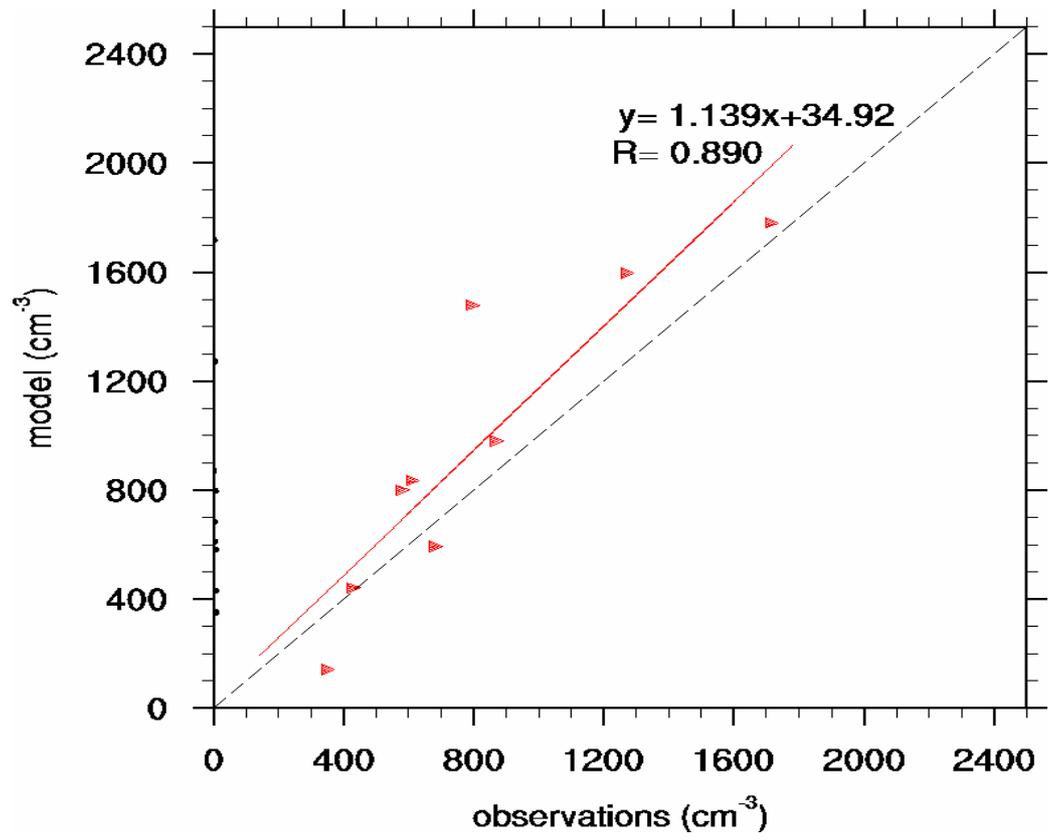


Figure 12

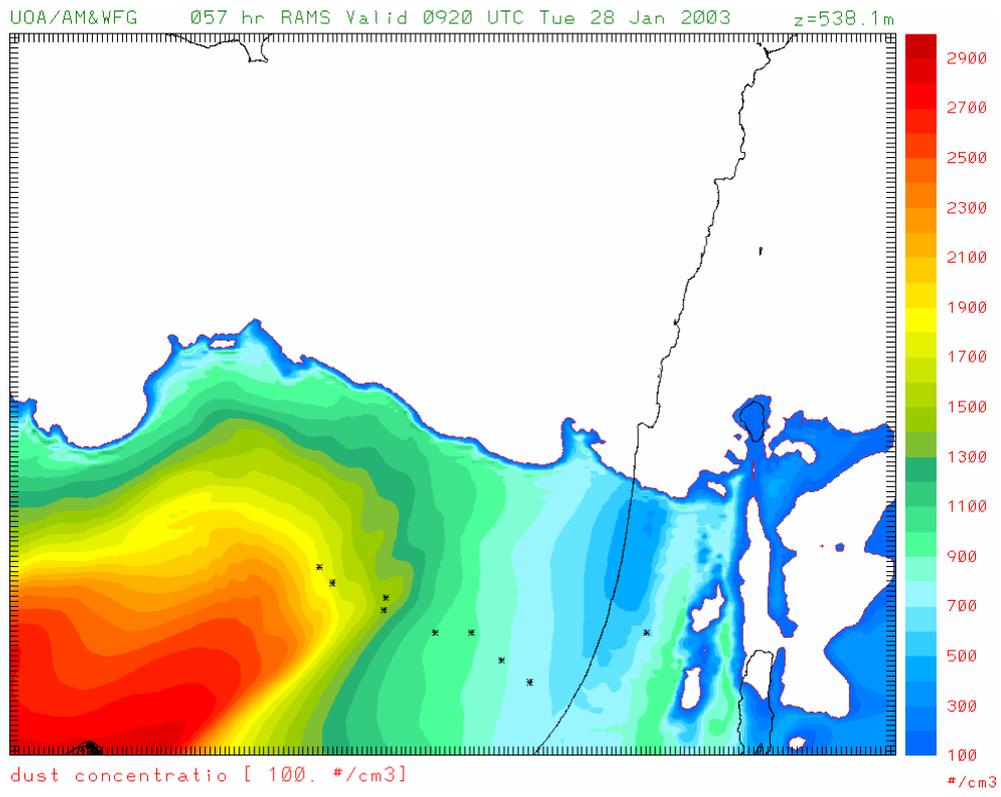


Figure 13

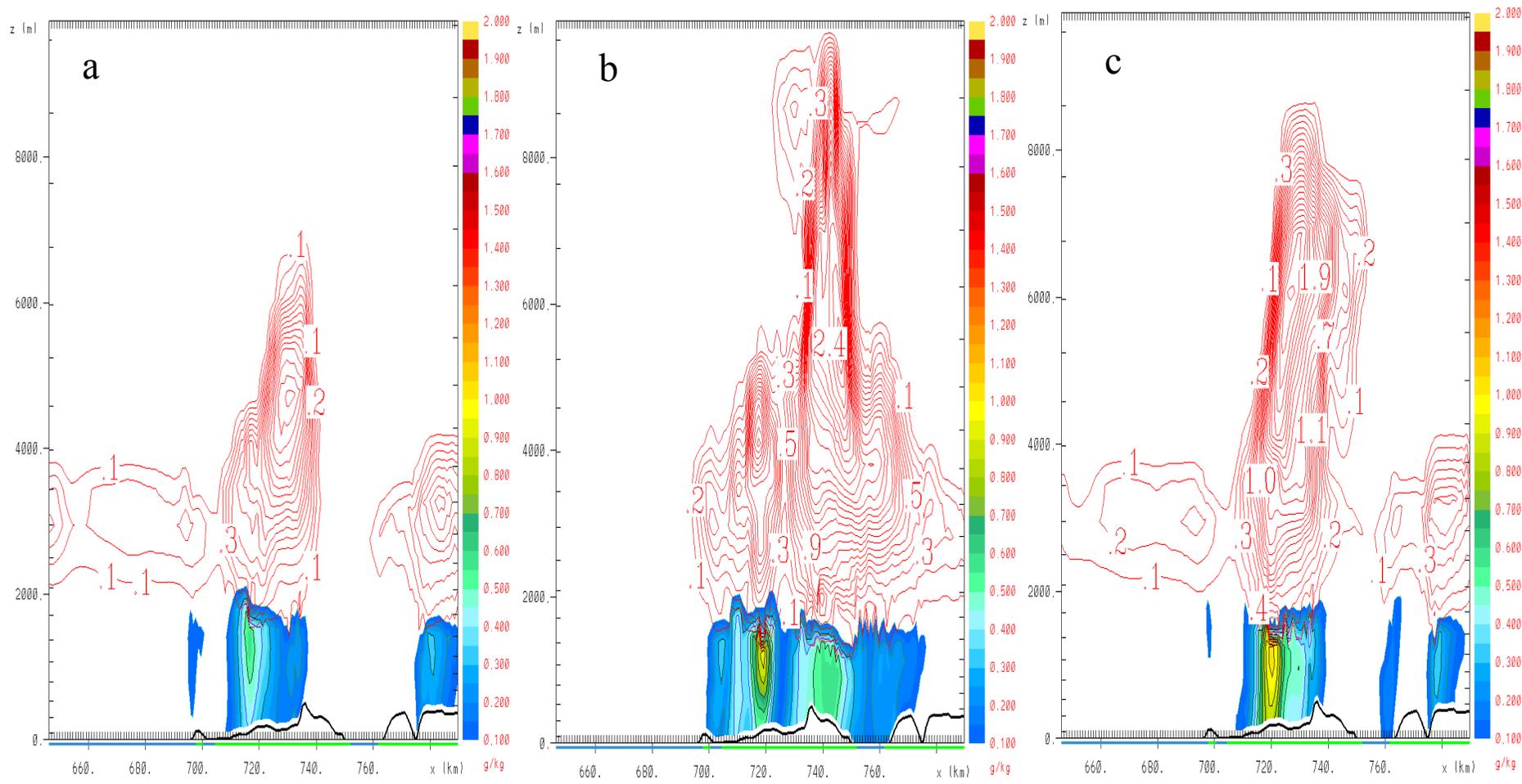


Figure 14

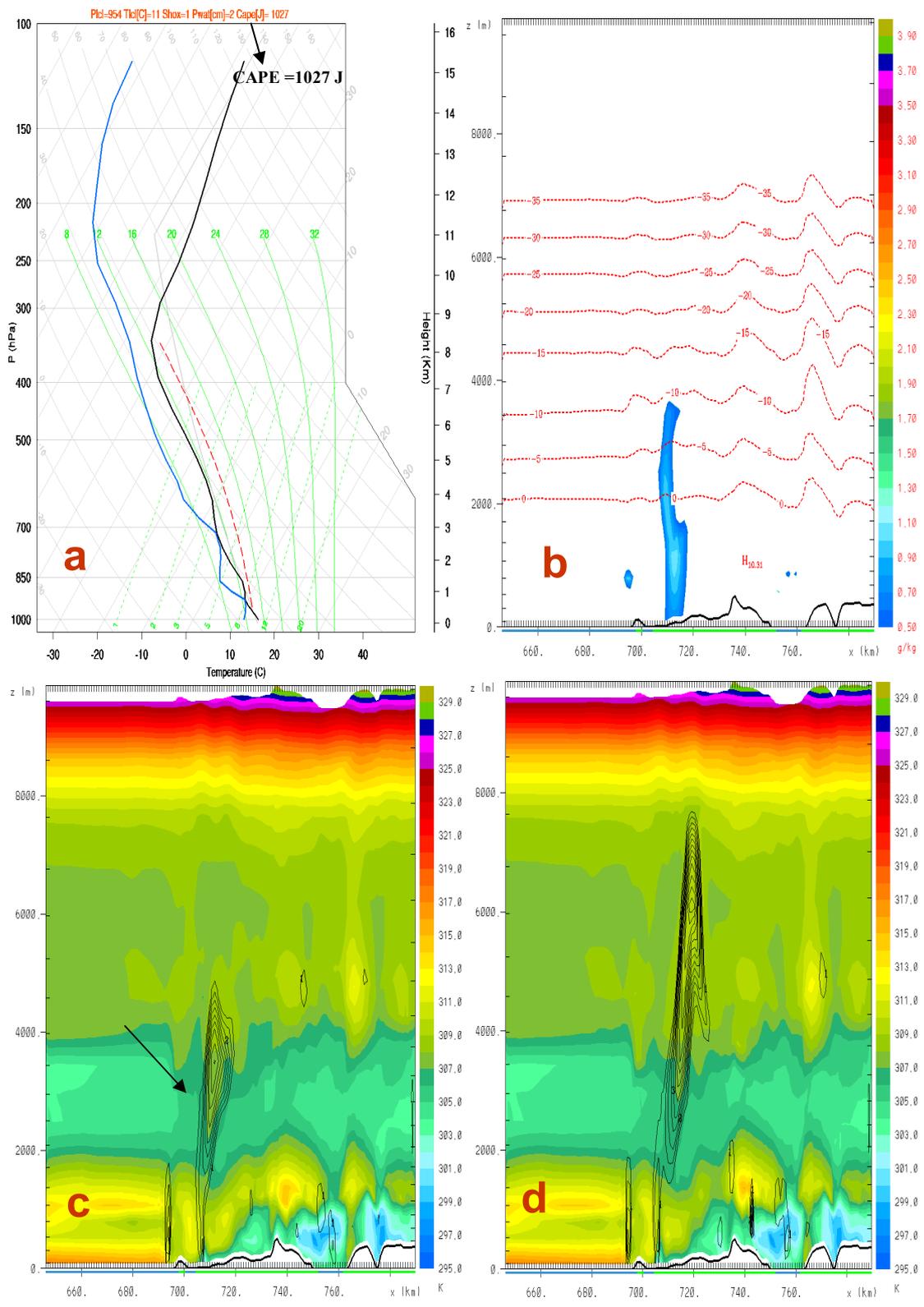


Figure 15

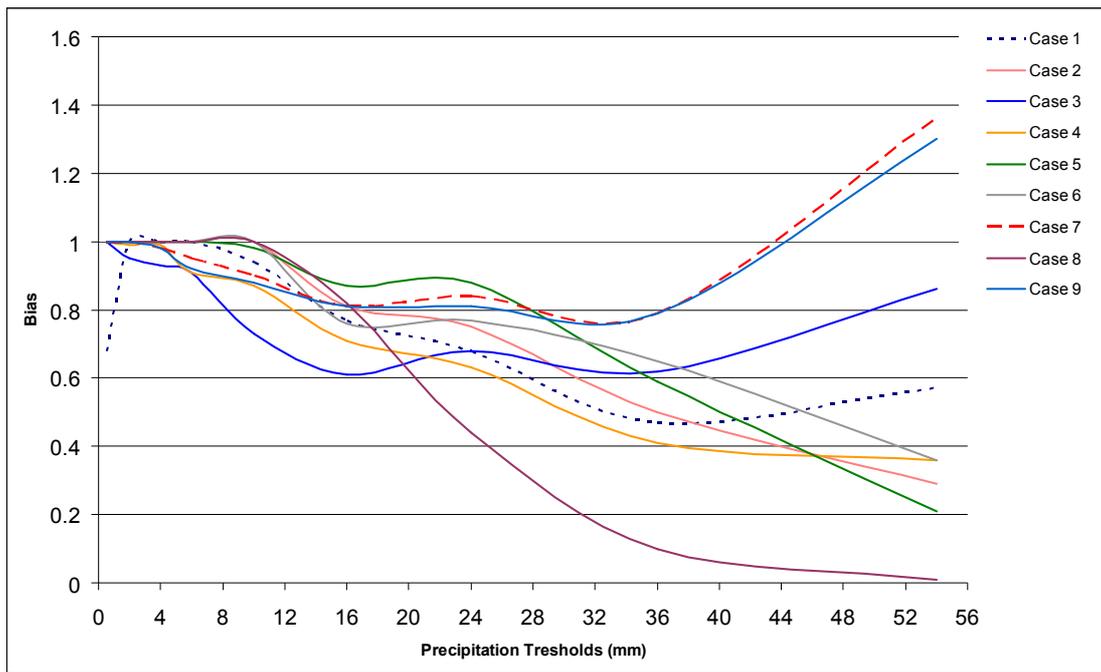


Figure 16

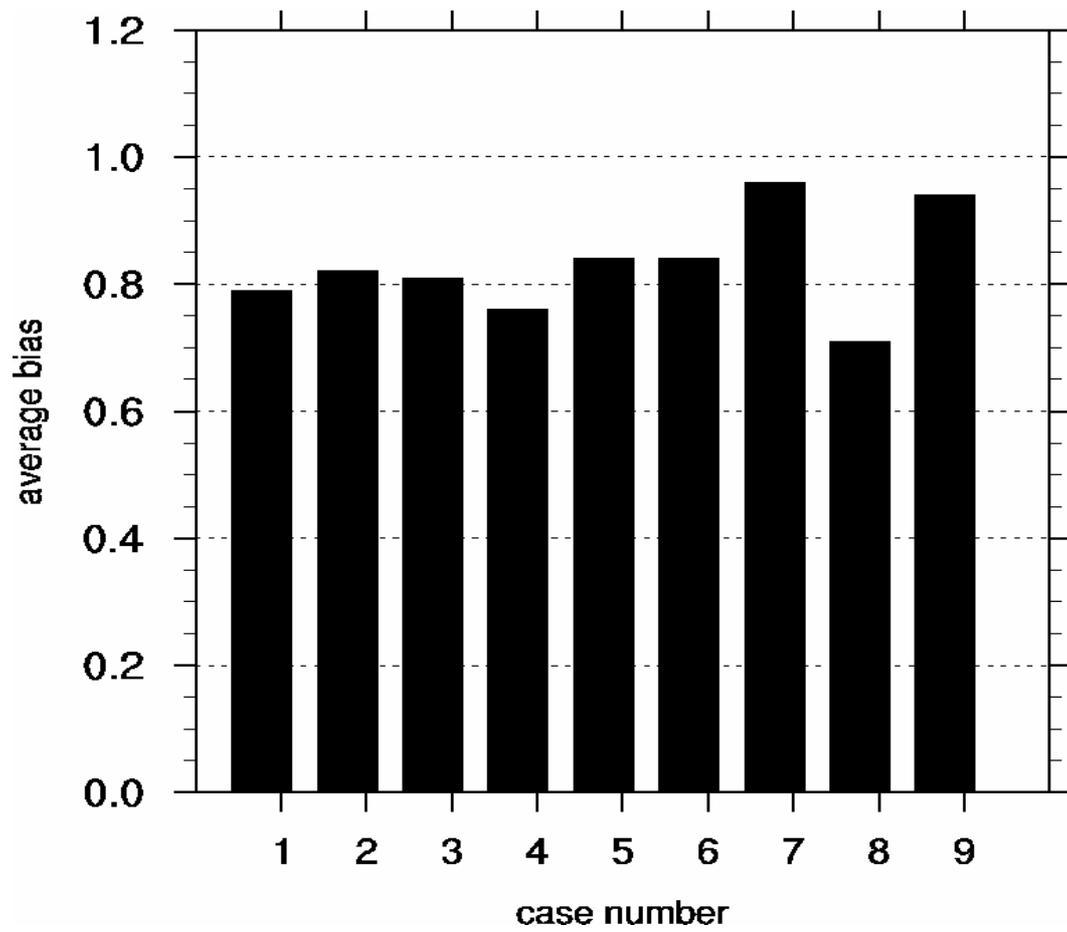


Figure 17