

Microphysical Characterization of Aerosol Signatures over Greece

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Abstract

A detailed microphysical characterization is presented for aerosol spectra over three major areas over Greece, namely- the Area West of Crete (AWC), the Greater Thessaloniki area (GTA) and the Greater Athens Area (GAA) based on measurements made by Varotsos (2005). Aerosol size distributions along with cloud droplet concentrations were gathered from data collected from the Forward Scattering Spectrometer Probe (FSSP)-300 and Passive Cavity Aerosol Spectrometer Probe (PCASP)-100X flown on a Falcon aircraft over Greece (Varotsos, 2005). However, the microphysical and dynamical characterisation of the observations were left largely unexplored (for this later paper), particularly with regard to their growth and activation into cloud condensation nuclei. A suite of models involving Large Eddy Simulations and an Adiabatic Parcel Model were used to achieve this. The UK Met Office large eddy model (LEM) reveals that mild to moderate updraughts in the range of 0.5 ms^{-1} to 2 ms^{-1} were present over Athens during June 1997, over the regions containing the aerosol bands. The sophisticated parcel model was then been applied to this data to study the growth of aerosol in the updraughts present and further, to explore the possibility of rainfall. It is found that the grown droplets have radii spanning a range of $6 \mu\text{m}$ to $100 \mu\text{m}$ indicating a hundredfold increase over initial size and the region spanning height from 2700 to 3000 meters contains precipitable water.

Keywords: urban aerosol; maritime aerosol; cloud droplets; cloud dynamics; cloud micro-physics

1. Introduction

In a paper by Varotsos (2005), on the aerosol characterization and solar irradiance over the entire Greek area, airborne measurements were recorded using the aforementioned instrumentation. Much useful cloud microphysical data was garnered through a series of aircraft borne measurements. However, the subsequent cloud processing through condensational growth followed by stochastic collision-coalescence was not undertaken. In this paper, a complete picture is presented by a complementary microphysical modelling to ascertain the fate of aerosol particles over Greece. It is anticipated that this detailed characterization of the grown droplets through the Adiabatic Parcel Model (APM) will assist in a thorough radiative transfer calculation in climate models (Toon and Pollack, 1976; Tegen and Lacis, 1996; Ghosh *et al.*, 2007; Varotsos and Zellner, 2010) because

aerosols do not just stay aloft- they also grow with altered microphysics. It is this altered signature that we endeavor to establish in this paper.

We first give details of the instrument flown on the Falcon aircraft over Greece followed by modelled aerosol distribution and large eddy simulations (LES) over Greece and subsequently, dynamical characterization of aerosol growth. For measurements undertaken from 7-14 June, 1997 under the framework of the experimental campaign of the radiation field in the troposphere (RAFT) - a subproject of the scientific training and access to aircraft for atmospheric research throughout Europe (STAAARTE), it should be noted that during June, weak synoptic flows over Athens, Thessaloniki and Crete correspond to days of high pollution. When pollution concentrations are high, the resultant droplet sizes are small and aerosol particles do not cross the threshold of $20 \mu\text{m}$ to grow through stochastic coalescence into rain drops (Ghosh

et al., 2005; Varun Raj *et al.*, 2009). However, clouds were observed to be present over these areas. With the measured values of relative humidity (Varotsos, 2005), the aerosol particles are grown through condensation using Köhler theory (Köhler, 1936; Seinfeld and Pandis, 1998). In order to be able to do the latter, LES was used to determine the parcel ascent (Gary *et al.*, 2001).

2. Aerosol measurements and modelled aerosol distribution

The research aircraft Falcon 20-E5 D-CMET of the German Aerospace Research Establishment (Deutsches Zentrum für Luft und Raumfahrt (DLR)) was equipped with the instrumentation to conduct measurements from the sea level up to 6.2 km over most of the part of the Greek area (Varotsos *et al.* 2001). Aerosol measurements were recorded with a recording frequency set to 1 s integrating interval using spectrometers FSSP-300 and PCASP-100X, mounted under the wings of the DLR research aircraft. The FSSP-300 spectrometer was used to measure cloud droplet size aerosols (diameters 3.5-45.5 μm), which were assumed to consist of mainly sea salt aerosol, whilst the PCASP-100X spectrometer was used to provide measurements of accumulation

mode aerosol particles (diameters 0.11-2.75 μm) which in turn represents the major sulphate mode aerosols. Varotsos (2005) provides greater details on the instrumentation employed for aerosol characterization, which are not included in the present text for reasons of brevity. In the figure below (Fig. 1), we show a summary of the number concentration variation with height of both of the sulphate as well as the salt modes measured respectively with PCASP and FSSP (Varotsos, 2005). It is observed that the sulphate layer is sandwiched between the two salt modes. The observations clearly show overlapping layers indicating the presence of a sandwich layer between heights 2-4 km corresponding to the sulphate mode.

The growth of the obtained aerosol spectra is fed into an adiabatic parcel model, discussed in section 4 of the text to ascertain the growth of the particles and their effect if any, on cloud formation and rainfall. The input salt and sulphate aerosol concentrations (Fig. 2) with the prevalent temperature, pressure and relative humidity for the APM for each of the three regions are summarized in Table 1. All aerosols exceeding 1 μm radius were assumed to be salt whilst those with smaller diameters were assumed to be sulphate aerosol (Ghosh *et al.*, 2005).

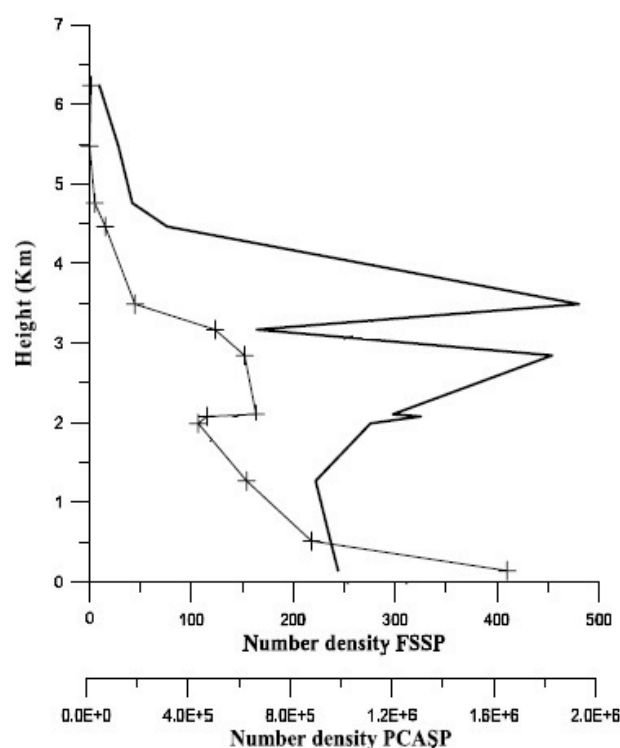


Figure 1. Distribution of the CS aerosol total number density from Forward Scattering Spectrometer Probe (FSSP) (solid line), and the PS aerosol total number density from PCASP (solid line with crosses) during June 1997.

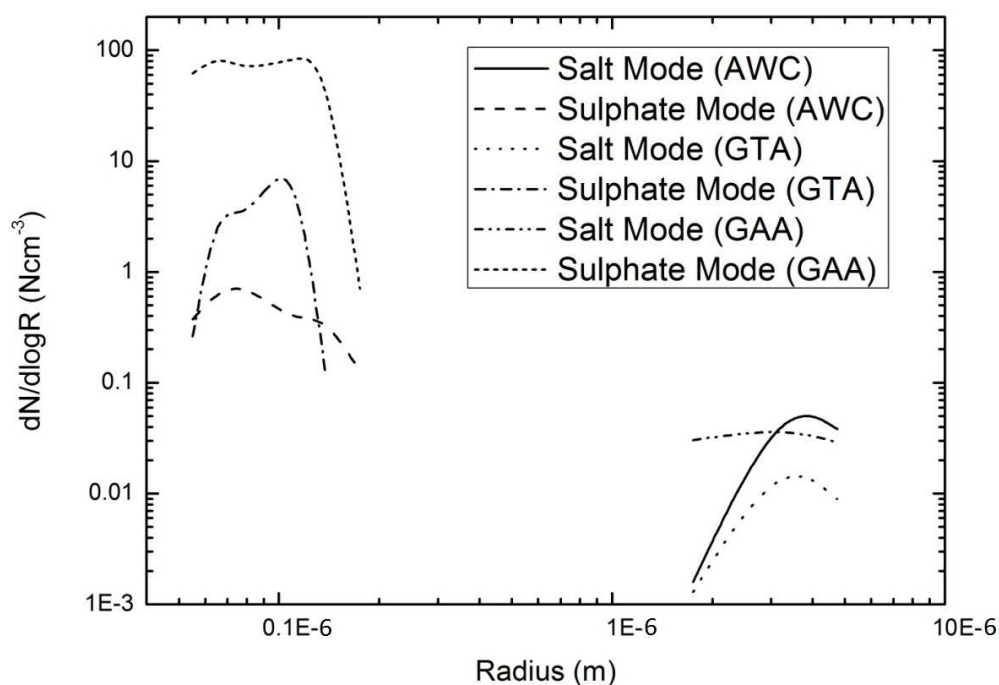


Figure 2. Aerosol spectra over Western Crete (AWC), Greater Thessaloniki (GTA) and Greater Athens (GAA). Note the high concentration of Sulphate aerosol over the highly populated and landlocked GAA.

Table 1. Input parameters for the APM, all concentrations are assumed to be at a reference height of 1800 m for AWC and 2000 m for GTA and GAA.

S. No.	Date/Area	Pressure [mbar]	Temperature [K]	Relative Humidity [%]	Salt		Sulphate	
					Radius [μm]	Concentration [cm^{-3}]	Radius [μm]	Concentration [cm^{-3}]
1	7 th July 1997/AWC	830	280	65	1.75	1.60E-03	0.055	3.74E-01
							0.065	6.62E-01
					3.25	7.76E-02	0.0775	7.52E-01
							0.0925	5.36E-01
					4.75	3.82E-02	0.1125	3.69E-01
2	9 th July 1997/GTA	780	276	80			0.1375	3.95E-01
							0.175	1.27E-01
					1.75	1.30E-03	0.055	2.62E-01
							0.065	4.0
					3.25	2.40E-02	0.0775	3.0
3	13 th July 1997/GAA	840	287	50			0.1125	6.0
							0.1375	10.0
					4.75	8.90E-03	0.175	1.27E-01
							0.055	62.0
					3.25	3.94E-02	0.065	88.0
							0.0775	70.0
							0.0925	74.0
					4.75	2.91E-02	0.1125	84.0
							0.1375	90.0
							0.175	0.70

Sourced from Varotsos (2005)

3. Large Eddy Simulation (LES) over Greece

For a definitive microphysical characterisation, a dynamical characterization of the cloudy region in question is required, in particular, the strengths and positioning of the up and down draughts effecting the vertical mixing of air masses. This is achieved through a Large Eddy Simulation. LES over Greece were performed to obtain the vertical velocity perturbations in order to determine the input updraught velocities to be supplied in the APM for subsequent aerosol growth characterization. Corresponding to the date of the actual experiment, a June 1997 run was performed over the greater Athens area (GAA), for a period of 6 hours at a height of 3 km. The vertical velocity perturbations over Athens are shown in Fig. 3(a) where a succession of sparsely spread peaks of strong up and downdraughts reaching a maximum of 2 ms^{-1} are observed. The mean vertical velocity perturbation is found to be 0.4 ms^{-1} . The corresponding 2-D contours of vertical velocity perturbations are shown in Fig. 3(b). Due to the relatively small size of the Grecian peninsula, the updraughts over AWC and GTA were assumed to be similar to that of GAA. The results obtained from the dynamical characterization at the cloud level using LES are coupled with APM to predict the cloud droplet spectral development.

4. Microphysical characterization–adiabatic parcel model

An adiabatic parcel model (APM) (O'Dowd *et al.* 1999) that employs the use of dynamical growth equations to predict the growth of aerosol solution droplets by the condensation of water vapour in an updraught, on a size resolved droplet spectrum is used in the present study. The input aerosol spectrum, relative humidity and temperature were derived from Varotsos (2005). The updraught velocities over the target areas for the requisite days were calculated using the UK Met office Large Eddy Model.

The reference heights for the AWC, GTA and GAA are assumed to be 1800 m, 2000 m and 2000 m respectively (Varotsos, 2005). This reference height is always below the cloud base which is set at $Z=0$ (where Z is the vertical distance) in the APM. All heights mentioned hereafter are above the reference height, unless mentioned otherwise. The growth curves for the three cases reveal that for the AWC and GTA, all aerosol modes exhibit rapid condensational growth at a height up to a height of 1000 m (Fig. 4(a) and (b)), breaching the critical radius of $20 \mu\text{m}$ (Ghosh *et al.*, 2005) required for stochastic coalescence to set in, thereby invoking the

possibility of rainfall over these areas. Over the GAA, only a few of the larger sulphate modes breach the $20 \mu\text{m}$ barrier (Fig. 4(c)). It is interesting to note here that the height at which this growth occurs coincides with the upper layer of the aerosol sandwich observed by Varotsos (2005), i.e. about 3 km from the mean sea level (MSL). The aerosol sandwich, which consists of two cloud droplet size aerosol distribution maxima (sizes in the range of $3.5\text{--}12.5 \mu\text{m}$) at a height of about 2.2 and 3.2 km respectively along with a middle layer of aerosol particles, all three coexist with the maximum of relative humidity (75%). This opens the door to the possibility that the upper layer of this aerosol sandwich may have formed in part, due to the condensational growth of the aerosols in the lower layers as they are transported upwards by the eddies whose mean updraught velocities lie close to 0.4 ms^{-1} , as ascertained from the LEM run. The supersaturation profile (Fig. 5) for the AWC reveals a clear cloud base forming at 1000 m, which is the same height at which maximum aerosol growth is observed. The profiles over the GTA and GAA also show the formation of a cloud base at 500 m and 1200 m respectively, but the value of supersaturation is considerably diminished as compared to the AWC.

5. Conclusions

The aerosol spectra over populous cities of today are a complex mixture of natural and anthropogenic aerosols (Kavouras *et al.* 2013; Varotsos *et al.* 2012). The anthropogenic aerosols comprising small sulphate particles result in the suppression of cloud and rain formation by virtue of their small size and large number concentrations (Ghan *et al.*, 1998). In contrast, larger sized sea salt aerosol in moderate concentrations has been shown to accelerate rainfall (Jung *et al.*, 2015; Kogan *et al.*, 2012; Yin *et al.*, 2000). The microphysical characterization of aerosol measurements over Greece reveals that in the coastal regions, i.e. GTA and AWC, the possible modulating effect of the sea salt results in favourable growth of all aerosol modes to well above the critical radius of $20 \mu\text{m}$, leading to the possibility of rainfall during the period of study-June 1997. Over the GAA, anthropogenic contribution from pollution results in a lot of fine mode sulphate particles which overwhelms the salt number concentrations. Only a few sulphate modes breach the critical size required for the onset of stochastic coalescence.

Another feature revealed by the microphysical characterization is the possible explanation of the bi-layered aerosol sandwich noticed over most of

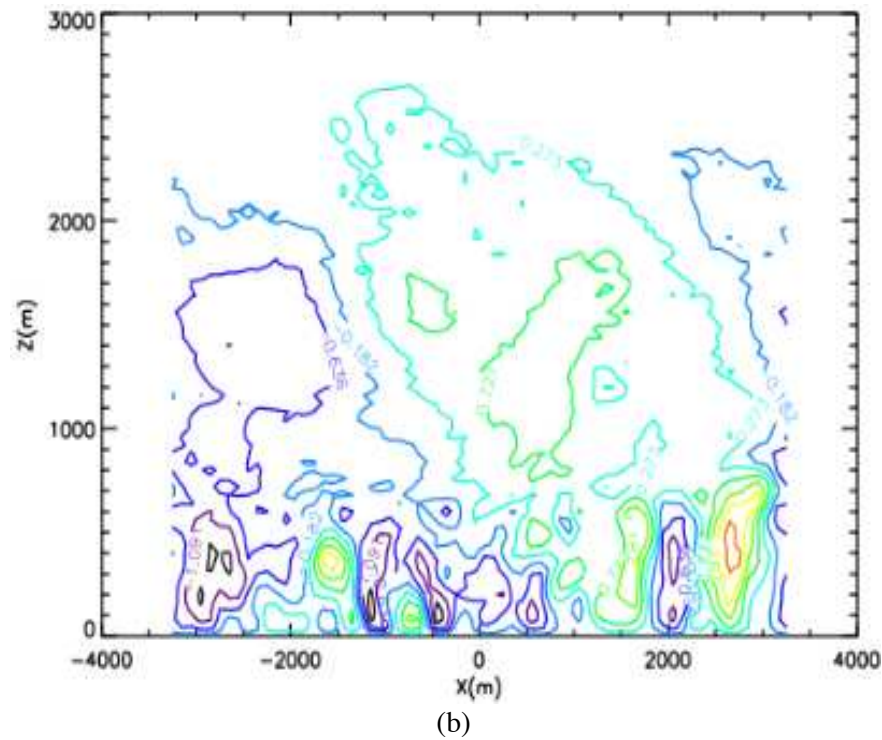
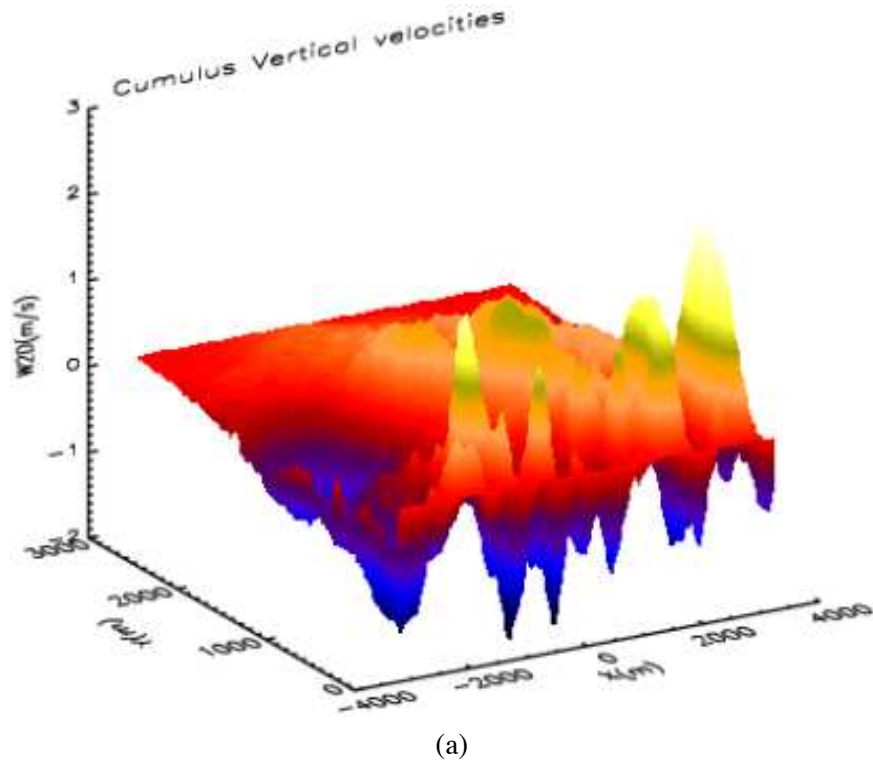


Figure 3. LES Simulations over Athens. (a) shows vertical velocity perturbations and (b) shows contours of vertical velocity perturbation over Athens during June, 1997. Note that the updraught velocities reach a maximum of 2 ms⁻¹, while presence of significant downdraughts yields a mean updraught velocities of 0.4 ms⁻¹. It is also clear from (b) that the smaller eddies are concentrated at the ground level whilst the larger eddies are prevalent at higher altitudes.

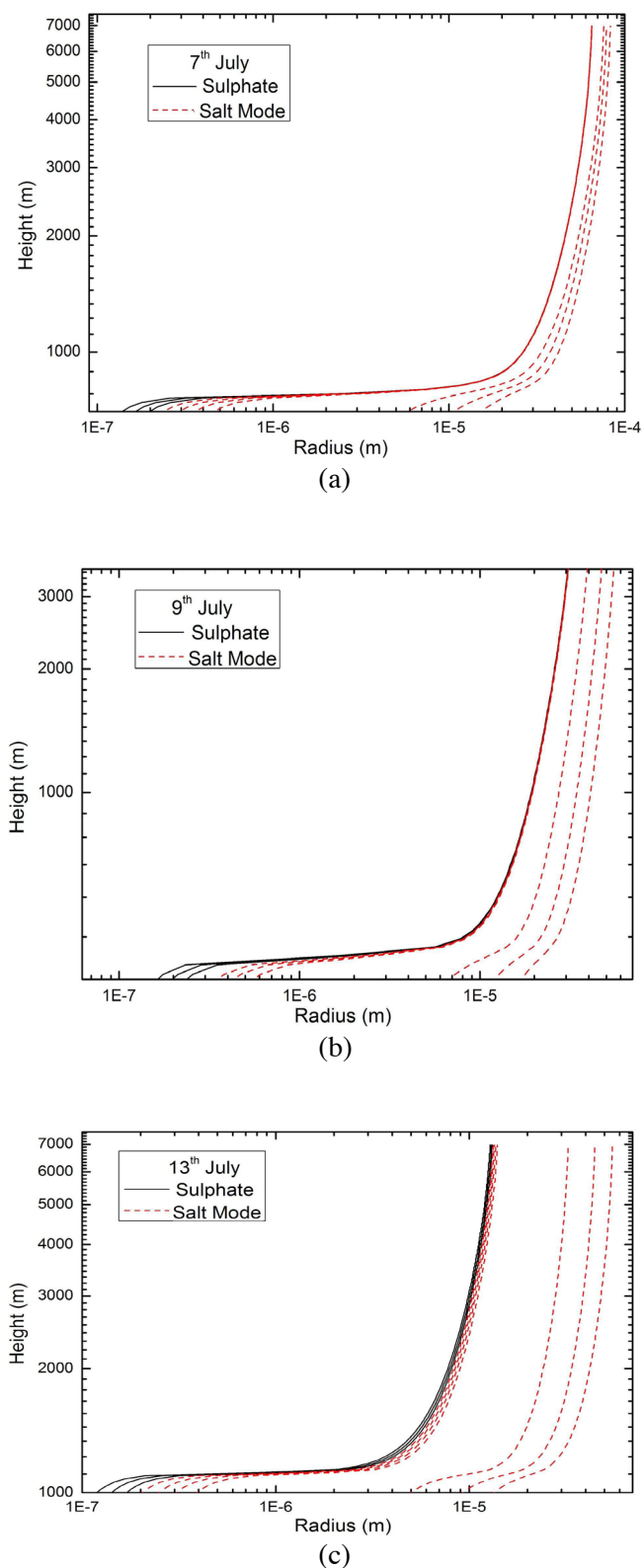


Figure 4. Growth profiles for salt and sulphate modes over the (a) AWC, (b) GTA and (c) GAA. It is worth noting that all aerosol modes seen over AWC and GTA grow above the 20 micron critical radius at around 1000 m above the reference height, whereas only a few sulphate modes achieve this over the GAA in the given experiments

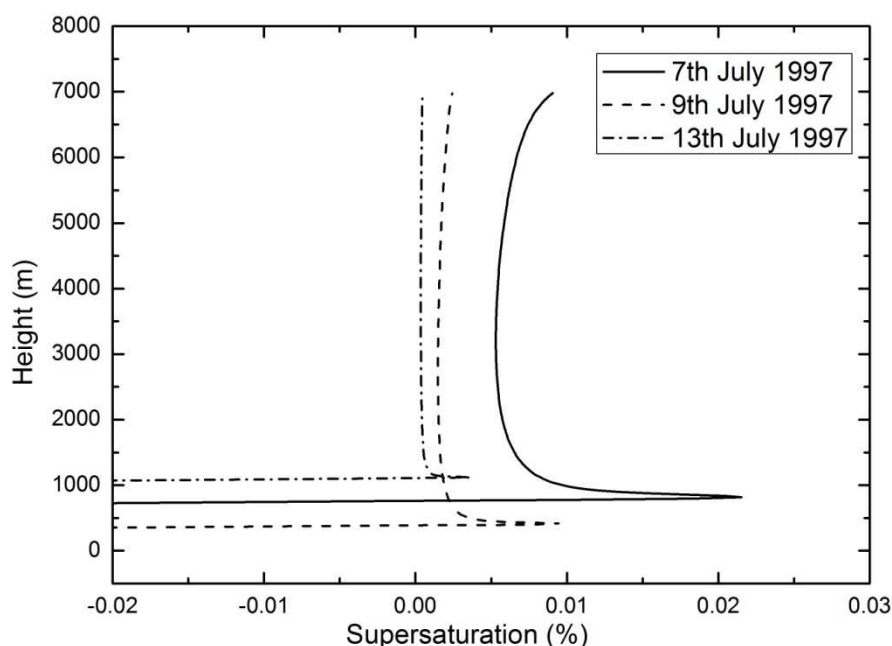


Figure 5. Supersaturation profiles on 7th July (AWC), 9th July (GTA) and 13th July (GAA). In the AWC, a clearly defined cloud base is seen at a height of 1000m above reference height.

the area under study. The growth model shows that the growth of the bottom layer (0.7 km to 1 km) may itself be wholly or in part responsible for the formation of the upper layer (3 km to 3.2 km). As observed in Fig. 3, preponderance of significant updraught and downdraught velocities forces the mixing of the aerosols present in the two layers. A net positive updraught velocity of 0.4 ms^{-1} ensures the vertical transportation of the aerosols from the bottom layer up to the upper levels - which then grow by condensation growth, owing to an adequate amount of relative humidity confirmed from the supersaturation curves (Fig. 5). In addition, the larger size of the aerosols forming the upper layer also increases the possibility of stochastic coalescence thereby causing rainfall.

References

- Gary M, Petch J, Derbyshire SH, Brow AR, Lock AP, Swann HA. Version 2.3 of the met office large eddy model. Turbulence and Diffusion Notes 2001, Met Office, Exeter, UK. 2001; 275-77.
- Ghan SJ, Guzman G, Abdul-Razzak H. Competition between sea salt and sulfate particles as cloud condensation nuclei. *Journal of the atmospheric sciences* 1998; 55(22): 3340-47.
- Ghosh S, Davila J, Hunt JCR, Srdic A, Fernando HJS, Jonas PR. How turbulence enhances coalescence of settling particles with applications to rain in clouds. *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences* 2005; 461(2062): 3059-88.
- Ghosh S, Osborne S, Smith MH. On the importance of cumulus penetration on the microphysical and optical properties of stratocumulus clouds. *Atmospheric Chemistry and Physics* 2005; 5: 755-65.
- Ghosh S, Smith MH, Rap A. Integrating biomass, sulphate and sea-salt aerosol responses into a microphysical chemical parcel model: implications for climate studies. *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences* 2007; 365(1860): 2659-74.
- Jung E, Albrecht BA, Jonsson HH, Chen YC, Seinfeld JH, Sorooshian A, Metcalf AR, Song S, Fang M, Russell LM. Precipitation effects of giant cloud condensation nuclei artificially introduced into stratocumulus clouds. *Atmospheric Chemistry and Physics* 2015; 15(10): 5645-58.
- Kavouras IG, Lianou M, Chalbot MC, Vei IC, Kotronarou A, Hoek G, Hameri K, Harrison RM. Quantitative determination of regional contributions to fine and coarse particle mass in urban receptor sites. *Environmental Pollution* 2013; 176: 1-9.
- Kogan YL, Mechem DB, Choi K. Effects of sea-salt aerosols on precipitation in simulations of shallow cumulus. *Journal of the Atmospheric Sciences* 2012; 69(2): 463-83.
- Köhler H. The nucleus in and the growth of hygroscopic droplets. *Transactions of the Faraday Society* 1936; 32(2): 1152-61.
- O'Dowd CD, Lowe JA, Smith MH. Observations and modeling of aerosol growth in marine stratocumulus-case study. *Atmospheric Environment* 1999; 33(18): 3053-62.

- Seinfeld JH, Pandis SN. Atmospheric chemistry and physics: from air pollution to climate change. Cloud Physics, 1998: 777-840.
- Tegen I, Lacis AA. Modeling of particle size distribution and its influence on the radiative properties of mineral dust aerosol. Journal of Geophysical Research 1996; 101(D14): 19237-44.
- Toon OB, Pollack JB. A global average model of atmospheric aerosols for radiative transfer calculations. Journal of Applied Meteorology 1976; 15(3): 225-46.
- Varotsos C. Airborne measurements of aerosol, ozone, and solar ultraviolet irradiance in the troposphere. Journal of Geophysical Research 2005; 110(D9): 1-10.
- Varotsos C, Alexandris D, Chronopoulos G, Tzanis C. Aircraft observations of the solar ultraviolet irradiance throughout the troposphere. Journal of Geophysical Research 2001; 106(D14): 14843-54.
- Varotsos C, Ondov J, Efstathiou M. Scaling properties of air pollution in Athens, Greece and Baltimore, Maryland. Atmospheric Environment 2005; 39(22): 4041-47.
- Varotsos C, Ondov J, Tzanis C, Öztürk F, Nelson M, Ke H, Christodoulakis J. An observational study of the atmospheric ultra-fine particle dynamics. Atmospheric Environment 2012; 59: 312-19.
- Varotsos CA, Zellner R. A new modeling tool for the diffusion of gases in ice or amorphous binary mixture in the polar stratosphere and the upper troposphere. Atmospheric Chemistry and Physics 2010; 10(6): 3099-105.
- Varun Raj S, Ghosh S, Sivakumar B. Aerosol growth and activation in polluted air masses over a tropical metropolis in the Indian sub-continent. Atmospheric Science Letters 2009; 10(2): 66-74.
- Yin Y, Levin Z, Reisin T, Tzivion S. Seeding convective clouds with hygroscopic flares: numerical simulations using a cloud model with detailed microphysics. Journal of Applied Meteorology 2000; 39(9): 1460-72.

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