

# Aerosol growth and activation in polluted air masses over a tropical metropolis in the Indian sub-continent

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## Abstract

Air pollution can affect cloud formation in more than one way. When the pollutant gases are condensable (e.g. oxides of sulphur), then the process of aerosol activation is eased to a certain extent aiding cloud formation. However, polluted days are often characterised by low updraught speeds which inhibit aerosol growth. In this study, we have critically examined the aerosol activation process in a polluted coastal environment where both effects are present. We have concentrated on the Chennai region (one of the largest cities in the world) of the Indian sub-continent because its pace of industrialisation is increasing rapidly, adding to increasing SO<sub>2</sub> pollution over the years. Air masses over Chennai contain a mixture of aerosol particles including NaCl, because of its proximity to the Bay of Bengal, along with ammonium sulphate. We have used observational data along with a detailed microphysical chemical parcel model to study cloud activation effects. We find that over Chennai, often the presence of the condensable pollutant vapour (SO<sub>2</sub>) more than compensates for the low updraught speeds by lowering the level of maximum super saturation significantly. This latter effect favours the activation of ammonium sulphate as well as NaCl aerosol particles. We have undertaken a systematic analysis to quantify the relative strengths of these two competing effects and find that even with low updraught speeds, oxides of sulphur can perturb the activation domain comprising a mixture of aerosol particles to such an extent that aerosol particles in polluted environments often grow efficiently. This effect is non-intuitive in the sense that one associates smaller cloud droplet sizes with polluted air masses. This is the first microphysical modelling study for the Indian sub-continent where National Environmental Engineering Research Institute (NEERI) observations have been applied to cloud microphysical processes. Copyright © 2009 Royal Meteorological Society

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## 1. Introduction

It is well known that aerosols undergo chemical and physical changes while in the atmosphere. The particles that can grow to cloud droplets in the presence of water vapour super-saturation are termed as *cloud condensation nuclei* (CCN). Although clouds have a profound effect in controlling climate, our current level of scientific understanding is low and has been testified by the recent IPCC (2007) summary report. The aerosol activation process depends sensitively on a number of factors including the composition of the air mass in which they are present. Air pollutants are known to affect cloud properties (Kulmala *et al.*, 1993; Ghosh *et al.*, 2000) and when another condensable pollutant vapour is present along with water vapour, cloud formation mechanisms are radically altered (Kulmala *et al.*, 1993). It is not entirely clear to what extent the resultant cloudiness changes. The way in which aerosols are processed by clouds depends on the type, abundance and the mixing state of the aerosols concerned.

Ghosh *et al.* (2005a) used a detailed microphysical chemical parcel model to quantify the extent of cumulus–stratocumulus coupling during the European

Aerosol Characterisation Experiment-2ACE-2 campaign. However, there are no aerosol and cloud microphysical modelling studies for urban conurbations over the Indian sub-continent – this is a first systematic modelling study where pollution effects have been linked to cloud droplet spectral development.

We have undertaken a modelling study with a particular reference to cloud formation over a coastal city. Our proximity to Chennai and the fact that it is a polluted city makes it our natural choice. We have also contrasted the polluted case studies with ‘clean’ test cases so that pollutant effects are singled out conspicuously. The unique location of Chennai along the east coast of peninsular India ensures that the city receives a mixture of aerosol particles from the natural as well as anthropogenic sources. Being a rapidly growing industrial city, gaseous air pollutants such as SO<sub>2</sub> are also present (Section 2).

## 2. Chennai’s pollution profile and meteorology

### 2.1. Aerosol characterisation over Chennai

Chennai has one of the longest beaches in the world and much of the city’s aerosol loading comprises of

both film- and jet-mode sea-salt particles. Sea salt aerosol has a large influence on the atmospheric sulphur cycle (Chameides and Stelson, 1992), and it influences the climate effects of natural and anthropogenic sulphate aerosol (O'Dowd *et al.*, 1999a). O'Dowd *et al.* (1997) has given a short review of the marine aerosol size distribution and the contribution of sea salt to this distribution. The potential role of sea salt in the marine boundary layer sulphur cycle has been highlighted in that paper. Apart from sea spray, ammonium sulphate particles are also abundantly present in the atmosphere of Chennai. The ammonium sulphate particles span a size range of 0.1  $\mu\text{m}$  to 1.0  $\mu\text{m}$ , which essentially covers an important aerosol mode called the *accumulation mode*. Fine crushed mineral (sand) particles from the arid and semi-arid regions, soot (carbon) particles from automobiles, industries, biomass burning, and sulphates from fossil fuel burning are important components of aerosol found over the continent including Chennai city. Aerosols are produced in varying proportions with large spatial and temporal variability, and reside in the atmosphere, mostly in the first few kilometres, from few hours to few days depending on their size (Jayaraman, 2001).

Jayaraman (2001) illustrated the aerosol-radiation-cloud interactions over the tropical Indian Ocean prior to the onset of the summer monsoon. From December to April every year, the trade wind blowing from the northeast brings polluted continental air over the Arabian Sea and the tropical Indian Ocean. Likewise, the northeast monsoon winds bring in polluted air over the east coast of India. A variety of fine suspended particles, majority of them manmade, get accumulated over the ocean region till the summer monsoon sets in and cleans the atmosphere. The aerosol–cloud interaction could be playing a crucial role for the onset of monsoon over different regions and the spatial distribution of rain over India. Recent observations over the Arabian Sea and the tropical Indian Ocean, conducted as part of the Indian Ocean Experiment,

have revealed the presence of large amount of aerosols over these regions. Jayaraman (2001) has illustrated that the aerosol transport in the horizontal direction is controlled by winds and is a mesoscale process. The vertical transport and scavenging is determined by the convective motions and takes place on convective scales. The aerosol particles caught in the convective updrafts serve as CCN which eventually grow into cloud droplets. The efficiency of an aerosol particle to serve as a CCN depends on its size and the amount of water adsorbing material in it. He has also shown that aerosols play a major role in modifying the earth's radiation budget over the polluted region. The estimated population increase in India and China will make the region more susceptible to the associated effects of pollution.

## 2.2. Air pollution meteorology over Chennai

Chennai is located on the southeast coast (Coromandel Coast) of India at an average altitude of 6 m above the sea level. The latitude and longitude of this city are 13.04°N 80.17°E, respectively. It covers a total area of 174 sq km spread irregularly in the northeast corner of Tamil Nadu (Figure 1). With an estimated population of over 7 million (2007), it is the fourth largest metropolitan city in India and one of the largest metropolitan areas in the world. The industrial hub of South India, Chennai, has a diversified economic base. The city's economy largely depends on trade and industry. The main industries are automobile, software services and hardware manufacturing, including financial services. In Chennai, oxides of sulphur are produced mainly from the combustion of fossil fuels that contain sulphur, such as coal and oil (for example, coal being burnt for electricity generation or fuel used in diesel-powered vehicles). Other industrial sources include iron and steel mills, petroleum refineries, and pulp and paper mills. Ammonia emission sources include motor vehicles, through their exhaust, intensive livestock production, and emissions from humans



**Figure 1.** Map of India showing the location of Chennai. Note that Chennai is a coastal city on the Bay of Bengal with substantial sea salt aerosol loading. The box with the red star symbol is the region of interest. This region is shown as an enlarged map in the adjoining expanded figure on the right. The sites marked in red receive more urban air pollutants when the prevailing wind direction is southwest (SW) during the pre-monsoon season.

and pets. Other sources of ammonia emission are from the manufacturing industries producing basic chemicals, metals, leather products, cement, lime, plaster and concrete products, glass products, ceramics, beverages, cars, car parts, textile products, paper and paper products. Ammonia is also produced from mining, electricity supply and petroleum refining activities. It is expected that the aerosols age faster in cities due to the pollutants in the atmosphere such as oxides of nitrogen and sulphur. The combustion of fossil fuels has greatly increased the concentrations of various oxide forms of nitrogen and sulphur, especially in major cities of India including Chennai (AQS, 1994), to the point where they have started to adversely affect important biotic components of ecosystems (Odum, 1983). Sulphates have been found to be the major contributors to precipitation acidity especially in the Indian context (AQS, 1994; Khemani *et al.*, 1994; Ravichandran and Padmanabhamurty, 1994). They are formed because of the atmospheric oxidation of sulphur dioxide (SO<sub>2</sub>) to ammonium sulphate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>) or sulphuric acid (H<sub>2</sub>SO<sub>4</sub>). As far as ammonia emission is concerned, combustion, bacterial decomposition of animal excreta and emanation from soils are its main sources. Reported NH<sub>3</sub>-emission factors are: 1 g kg<sup>-1</sup> for coal, 0.12 kg m<sup>-3</sup> for fuel oil, 10 mg m<sup>-3</sup> for natural gas, 1.2 g kg<sup>-1</sup> for wood and 0.15 g kg<sup>-1</sup> for forest fires. Present mainly as organic nitrogen, the nitrogen content of coal is 1–2% by mass, of which a large fraction is released as NH<sub>3</sub> upon heating (Warneck, 1988; Erisman *et al.*, 1995). The oxidation of sulphur dioxide produces a variety of secondary acidic sulphate compounds (Sirois, 1993; Cyrus *et al.*, 1995), which are products of either homogeneous (gas or aqueous phase) or heterogeneous (gas-particle phase) phase oxidation. In addition, strong acidic aerosols are also emitted (primary acid aerosols) by coal and oil burning power plants and various other industrial activities. Acidic sulphate aerosols are formed through both gaseous and aqueous phase oxidation of SO<sub>4</sub>. These aerosols, after rapidly reacting with gaseous ammonia (NH<sub>3</sub>), produce various molecular forms of SO<sub>2</sub><sup>-4</sup>, *viz* NH<sub>4</sub>HSO<sub>4</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>.

The ambient air quality standard according to the Central Pollution Control Board (CPCB) for SO<sub>2</sub> pollution for residential areas is 80 µm<sup>-3</sup> for an averaging time of 24 h. Pollution levels are monitored in the Chennai region by the National Environmental Engineering Research Institute (NEERI, 2006, 2007) at two spots (marked in red in Figure 1) for an

industrialised area (Thiruvottiyur) and a residential area (Mylapore).

The prevailing winds in Chennai are mainly south-westerly between May and September and northeasterly during the rest of the year. The city experiences its highest levels of pollution during the pre-NE monsoon months i.e. July and August, when the prevailing SW winds brings in polluted air from the inland regions (Table I – highest recorded SO<sub>2</sub> level in July 2007). This is in contrast to the other metropolitan cities of India (Delhi, Mumbai and Kolkata) where July and August are clean months since these cities receive much precipitation from the more common SW monsoon.

The average annual rainfall is about 1300 mm (51 in.). The city gets most of its seasonal rainfall from the northeast monsoon winds, from mid-September to mid-December. The extra-tropical low pressure systems passing through the northern part of the country from west to east, mainly during November to April, are termed as western disturbances. These systems pass through Chennai every year and cleanse the city from pollution (Table I – low pollution level during November).

Pollution data for Chennai from NEERI were scrutinized and dates with cumulus cloud formation as well as high pollution were chosen. For the control runs, days with cloud formation and low pollution were selected. Initial gas concentrations of SO<sub>2</sub> and NH<sub>3</sub> from NEERI data were used to initialise the model runs (Table I). From Table I we used a value of 30/3 µg m<sup>-3</sup> as the input SO<sub>2</sub> concentration for all the polluted/clean runs (July 16–17 2007/November 16–17 2006). The corresponding Ammonium concentrations for these days were also used in the initialisation of the model runs. It is observed that the ammonium concentrations are lower when SO<sub>2</sub> pollution levels are high indicating that some of the ammonia is neutralised by oxides of sulphur (NEERI, 2007). Our clean runs have SO<sub>2</sub> concentrations an order of magnitude lower than the polluted runs as is observed from the NEERI measurements. This observational trend supports our modelling rationale – we wanted to contrast the polluted days from the clean days so that extremes were accounted for suitably with the expectation that the other possible cases would fit in within the observed range of variability. We find that SO<sub>2</sub> concentrations are higher in industrial areas (Thiruvottiyur) as compared to residential areas (Mylapore). However it is important to note that even on a polluted day the concentrations are well below the

**Table I.** Pollution data from NEERI. Note that only cloudy days have been selected. The measured SO<sub>2</sub> concentrations are the highest in July (pre-SW monsoon) and are the lowest during November [northeast (NE) monsoon season].

Date	Sampling site description	Averaging time	Pollution/Cloudiness status	SO <sub>2</sub> concentration (µg/m <sup>3</sup> )	NH <sub>3</sub> concentration (µg/m <sup>3</sup> )
16–17 July 2007	Industrial (Thiruvottiyur)	4 h max.	High pollution, cloudy	30	5
16–17 Nov 2006	Residential (Mylapore)	24 h max.	Low pollution, cloudy	3	9

prescribed Pollution Control Board limits of  $80 \mu\text{g}/\text{m}^3$  (for an averaging time of 24 h). As we shall see later (Section 4) even with the modest levels of Chennai's pollution our model runs show a great deal of sensitivity to ambient pollution levels as well as aerosol loading levels. It is anticipated that in the years to come the perturbing effects of  $\text{SO}_2$  pollution on cloud micro physical developments will be even greater – it is expected that  $\text{SO}_2$  pollution will increase over Chennai at least for the next decade. Concentrations of the other gases,  $\text{H}_2\text{O}_2$ ,  $\text{O}_3$ ,  $\text{CO}_2$  and  $\text{HCl}$ , were not measured by NEERI and for these gases the model was initialised with conservative estimates and was held fixed for all runs. The concentration of  $\text{H}_2\text{O}_2$ ,  $\text{O}_3$ ,  $\text{CO}_2$  and  $\text{HCl}$  used in the model runs were 1 ppb, 30 ppb, 350 ppm and  $10^{-5}$  ppt, respectively.

### 3. Model description

A detailed microphysical parcel model was the main tool used (O'Dowd *et al.*, 1999b) and is, in essence, a Lagrangian parcel model with explicit micro-physics utilising the dynamic growth equation (Pruppacher and Klett, 1997) to model the growth of aerosol solution droplets by condensation of water vapour on to a size-resolved droplet spectrum. Rather than being fixed, the boundaries of these size channels move as the solution droplets grow. The growth law includes curvature and solution effects, and is corrected for the breakdown of the continuum approximation close to the droplet surface (Pruppacher and Klett, 1997). Mass-transport limitations based on Schwartz (1986) have also been included. The aqueous phase chemical aspects of the model consist of (1) an equilibrium module; (2) a kinetic reaction module; and (3) the non-ideal behaviour correction module (Pitzer, 1991). The equilibrium module is used to calculate the amount of soluble gaseous species dissolved in the aerosol, and to apportion the aqueous aerosol species between the various associated as well as dissociated forms, which is achieved by solving the charge balance equation for each droplet. Although there are a number of similar models in existence, the present Pitzer model is one of the most advanced of its kind. Apart from including all the standard features of droplet growth considered by all microphysical models, it also takes account of high ionic strength, non-ideal solution effects. The kinetic reaction module is used to compute the rate of heterogeneous sulphate production in the solution droplets and sulphate production is allowed to occur by  $\text{SO}_2$  oxidation using dissolved  $\text{H}_2\text{O}_2$  and/or  $\text{O}_3$  as the oxidant. The reaction rates and their temperature dependence are taken from Ayers and Larson (1990). Some of the nucleation-mode particles coagulate on pre-existing background aerosol to form accumulation-mode particles so important to the aerosol indirect effect. The second contribution to accumulation-mode sulphate comes from  $\text{SO}_2$  oxidation in clouds followed by evaporation, and recent

sensitivity tests have shown this latter sulphate component to be more effective than the former as far as the second indirect effect is concerned (Seland and Iversen, 1999). For these reasons, it was very important to include the kinetic reaction module in our model simulations. Further details of the model can be found in O'Dowd *et al.* (1999b). The model does not include entrainment effects. However, entrainment dilution effects are directly proportional to the updraught speeds and inversely to the radius of the air parcel (Mason and Jonas, 1974). From Table II we can see that the maximum updraught speeds encountered are of the order of  $0.5 \text{ m s}^{-1}$  and very often, particularly for the calm wind conditions they were as low as  $0.25 \text{ m s}^{-1}$ . In view of this observation we can state that entrainment-induced spectral changes are not large. Further, we wanted to single out the pollution signature on the resultant spectra without introducing added dynamical perturbations such as those induced by turbulence and entrainment.

### 4. Results and discussion

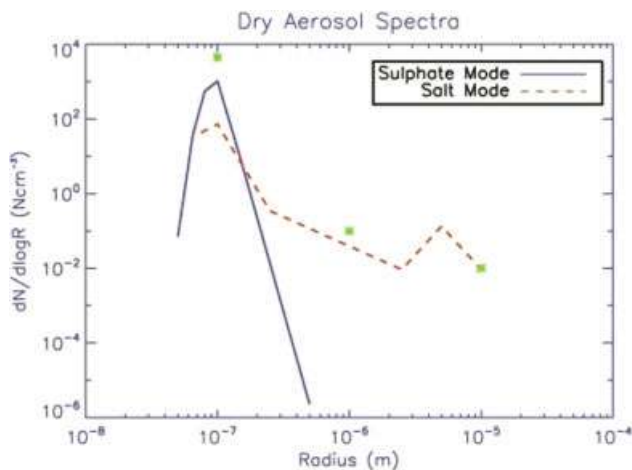
The model described in Section 3 was updated to include a multi-mode log-normal aerosol spectrum with prescribed mean and standard deviation based on globally averaged values (Ghosh *et al.*, 2007). See Figure 2. Although we have used globally averaged values to characterize the spectral size distributions shown in Figure 2, these values conform also to those of aerosol size distributions over the Indian sub-continent (Jayaraman, 2001). We generated six model runs with varying number densities for the sulphate mode (Table III). We have left the salt modes unchanged in all the six runs because it is expected

**Table II.** Dynamical inputs at the cloud base for the microphysical model.

Parameter	Value
Temperature	30 °C
Updraught speed	0.25 $\text{m s}^{-1}$ , 0.5 $\text{m s}^{-1}$
Relative humidity	99.0%
Pressure	941.0 mb

**Table III.** Summary of model runs.

Run No	Clean/Polluted	Run details	
		Dry sulphate aerosol number loading	Updraught speed ( $\text{m s}^{-1}$ )
1.	Polluted	Normal	0.25
2.	Clean	Normal	0.5
3.	Polluted	Halved	0.25
4.	Clean	Halved	0.5
5.	Polluted	Doubled	0.25
6.	Clean	Doubled	0.5



**Figure 2.** Multi-mode log-normal aerosol spectrum with prescribed mean and standard deviation. The asterisks in green are observations from Jayaraman (2001).

that pollution induced perturbations are more likely to affect the sub-micron fine mode. Further the range of variability of the fine-mode number concentration is within a factor of 4 – this trend is also supported by recent measurements. Chennai is an urban, industrial and a well-populated city, and is a constant source of aerosol particles – it is not unnatural for them to vary by a factor of 4. Measurements over the Indian Ocean have indicated the variation of the fine-mode number concentration could be within a factor of 6 (Murugavel *et al.*, 2001). Monthly variations in the 24-hourly average values of suspended particulate matter (SPM) concentration in Chennai also show a range of variability of the order of 4 or more (NEERI, 2006). We accounted for the observed range of variability of the aerosol fine-mode distribution by doubling and halving the number concentrations shown in the standard run corresponding to the distribution shown in Figure 2. It is expected that the aerosol distribution patterns covering Runs 1 through 6 are a reasonable proxy for the actual aerosol distributions over Chennai.

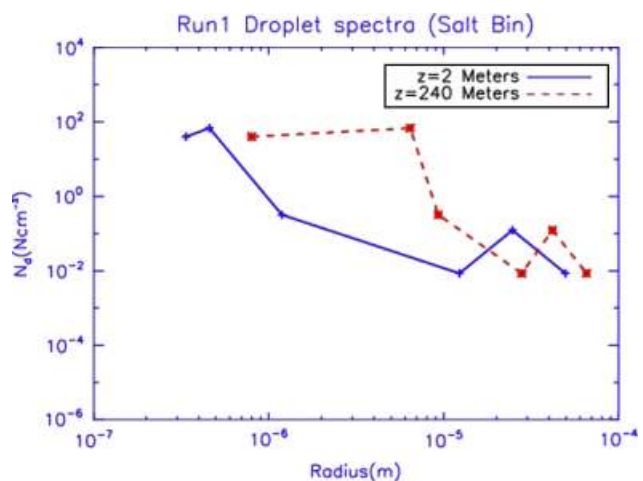
Three distinct modes are clearly visible in the input spectra shown in Figure 2. The first represents the sulphate mode and covers much of the accumulation-mode aerosol. In addition, two salt modes are also clearly visible, corresponding to the fine film mode and the coarse jet mode. The existence of these salt modes is wind speed dependent (O’Dowd *et al.*, 1999a).

Dynamical input parameters like the relative humidity, pressure and temperature at cloud base height were also used (Table II) and were based on radiosonde data (Srinivas *et al.*, 2007). The updraught speed was determined by the NCAR Mesoscale Model MM5. The details of the initialisation and the model are given in Srinivas *et al.*, 2007. Being a tropical city near the equator, large seasonal variations are not as severe as in higher northern latitudes. Further, the main aim was to isolate cloud activation effects, so the dynamical inputs had to be held constant for all the runs except for the updraughts, which were different for clean

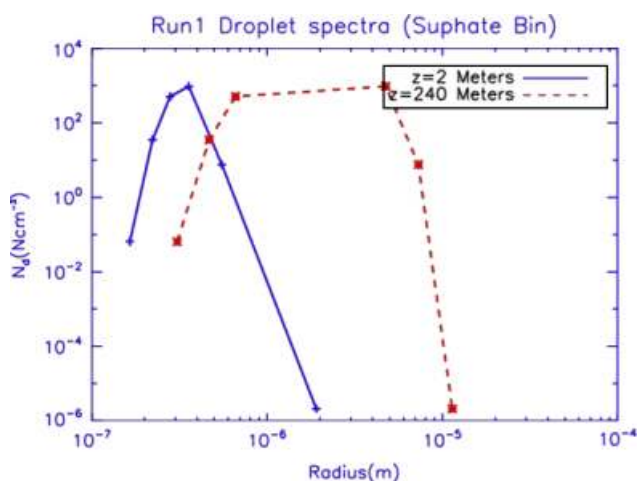
and polluted days. The NEERI (2007) observations showed that polluted days were characterised by lower updraught speeds, often about 50% lower than the measured values on days with low pollution – based on this fact, and for keeping the run numbers tractable for this study, we used only two representative values for the updraught speed i.e.  $0.25 \text{ ms}^{-1}$  and  $0.5 \text{ ms}^{-1}$  respectively for polluted and clean days.

Six runs were undertaken to study the nature and the characteristics of pollutant impacts on clouds (Table III). The classification is based on the variation of dry sulphate aerosol number loading—the levels shown in Figure 2 correspond to Runs 1 and 2. In these two runs only the concentrations of  $\text{SO}_2$  and  $\text{NH}_3$  are different and every other parameter is identical. In Runs 3 and 4, the sulphate aerosol number concentrations have been halved whilst in Runs 5 and 6, they have been doubled. The fine and coarse-mode salt concentrations have been left unchanged in all the six runs. These sensitivity tests were necessary in order to cover a wide range of possibilities. Runs 1 and 2 are typical of most days and correspond to observed aerosol data, whilst days with aerosol concentration numbers doubled/halved correspond to highly polluted/very clean conditions. As stated earlier, field measurements have indicated the variation of the fine-mode number concentration could be within a factor of 6 (Murugavel *et al.*, 2001). Although the model runs were initialised with dynamical and microphysical characterisations for specific days, these should be fairly representative of Chennai’s current pollution profile.

With the dry input aerosol distribution shown in Figure 2 we now show how these aerosol particles grow from a level close to the cloud base to a level deep within the cloud— we have shown these separately for the polluted Run 1 for the salt and the sulphate bins (Figure 3 and Figure 4). We see that of the six sulphate and the six salt bins the smallest bins in each mode remain non-activated whilst the larger ones grow significantly.



**Figure 3.** Run 1 Droplet spectra (salt bin) close to the cloud base and at a level deep inside the cloud. Note that the smallest bin hardly grows, while the subsequent bins grow significantly.



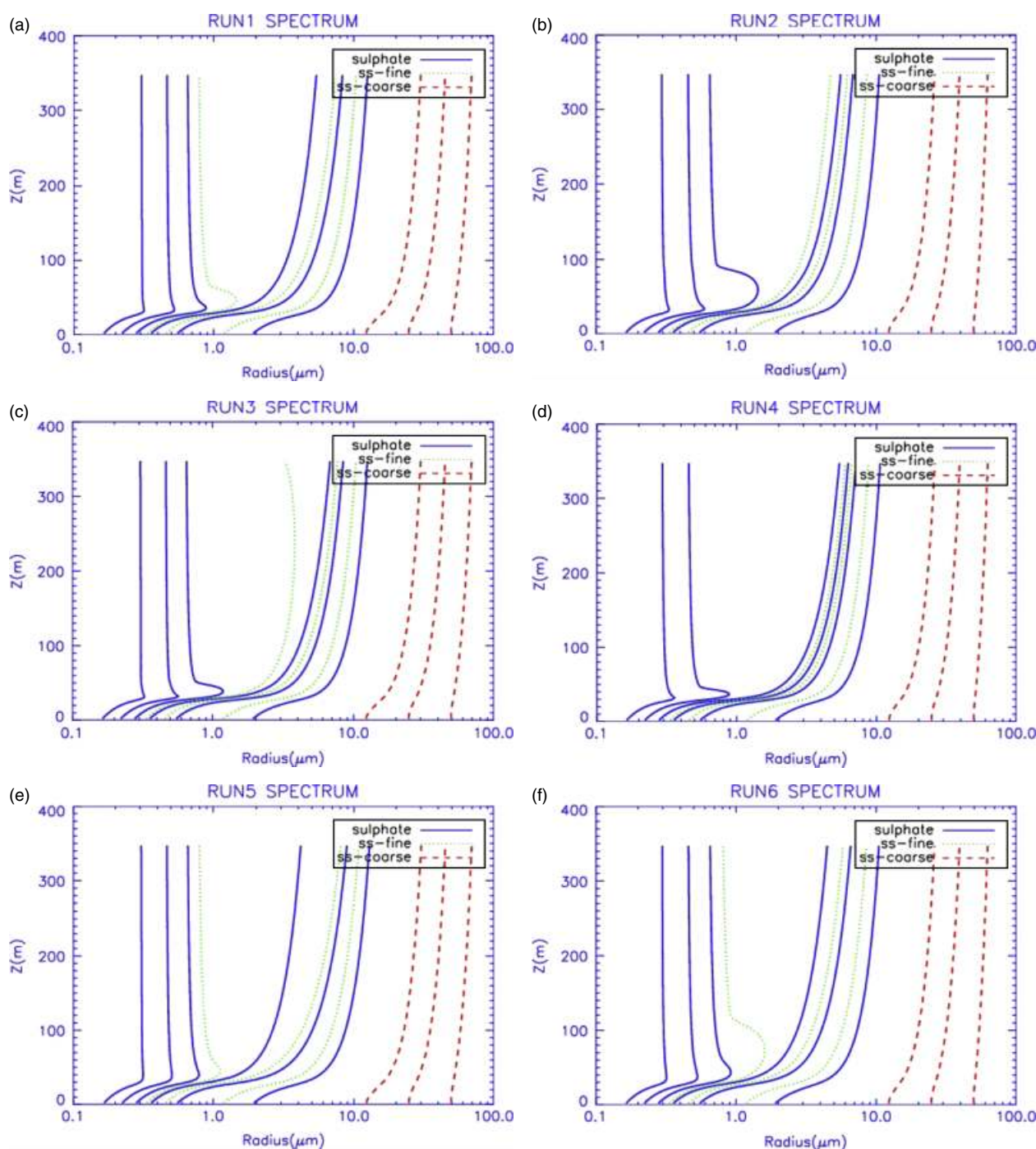
**Figure 4.** Run 1 Droplet spectra (sulphate bin) showing insignificant growth of the smaller sulphate bins 1, 2 and 3. In contrast, the other three bins show substantial growth.

Several interesting features are revealed from Figure 5 where we explicitly show the growth profiles for all bins. First, we note that the cloud base forms at  $\sim 26$  m for all cases. The activated bins are those where the profiles slope to the right, i.e. the radius increases with height. The non-activated bins are represented by the parallel vertical lines. For the polluted case all the coarse-mode salt bins get activated but one of the fine-mode salt bins is left un-activated. For Run 1, the higher pollution eases the activation so that the larger sulphate bins grow more efficiently depleting moisture. This suppresses the growth of the smallest fine-mode salt bin. This effect is suppressed in Run 2 – we can see that the smallest salt bin has indeed activated as some of the available moisture is now freed enabling the activation of the smallest salt bin.

In Runs 3 and 4 (Figure 5(c) and (d)) the dry sulphate aerosol number concentration is halved and it can be seen that all the salt bins now grow. Sulphate bin 3 activates in Run 4 as enough moisture is now available. In contrast, when we examine Runs 5 and 6 (Figure 5(e) and (f)) we find that in these two runs one obtains the maximum number of un-activated bins owing to the maximum aerosol number concentration densities. It is worth noting that the number of un-activated bins also depends on the overall sub-micron aerosol number concentrations. Our smallest fine-mode salt particles are of the order of  $0.5 \mu\text{m}$  with a number density never exceeding a few  $100 \text{ cm}^{-3}$ . It is also possible that situations can arise where this number concentration can easily be exceeded – for cyclonic depressions one encounters very high wind speeds injecting much larger amounts of fine-mode sea salt particles over Chennai. In such situations there would be a severe competition for the available moisture and only the largest of the aerosol particles could form cloud droplets. However, for the runs that we have explored, we get a deeper insight into the activation process, when we examine the super-saturation profiles shown below in Figure 6.

We note that the maximum super saturation is the highest for Run 4 and lowest for Run 5. This is to be expected since Run 4(5) is characterized by the lowest (highest) dry sulphate aerosol number concentration. The pollution signature is clearly evident from the comparison of Runs 3 & 4 – for Run 3 corresponding to the polluted run we find that the maximum super-saturation has significantly lowered suggesting that the presence of a condensable vapour eases the process of activation. This trend is generally true for all the model runs.

When we examine the growth profiles of individual bins we obtain a still deeper insight into the activation process. We have shown this explicitly for Runs 1 and 2 (Figure 7) for the sulphate bin 5. We find that the bin in the polluted case grows more than for the clean case (this trend is preserved for Bin 6 also) and the reason is immediately apparent when we examine Figure 7. The higher  $\text{SO}_2$  level in Run 1 clearly lowered the level of maximum super-saturation, so that the aerosol particle readily activated, despite the fact that the ascent rate of the parcel in Run 1 was half that of run 2. This is true even when the aerosol number concentrations are halved (Runs 3 and 4) and doubled (Runs 5 and 6). This is a significant result in the sense that, we are seeing that higher pollution can also cause sulphate aerosol drops to grow in polluted environments. In repeated cloud processing cycles these may also grow to sizes large enough to induce collision/coalescence, because of their significant number concentrations. So far, we have discussed how the large sulphate bins activate in a polluted air mass. Let us now concentrate on the salt bins. When we examine a fine-mode salt particle we find that the fine-mode salt particle does not grow in the polluted run but it grows in the clean run. In Figure 8 we have shown the growth curves for bin 7, i.e. a fine-mode salt bin. This happens because the enhanced growth of Bins 5 and 6 for the polluted run depletes the parcel of moisture to such an extent that a fine-mode salt particle is not able to grow as the updraught speed is half that of a clean run. Coarse-mode salt particles are naturally large to easily overcome the Kohler barrier and grow to droplets with radii in excess of  $20 \mu\text{m}$  (the minimum size requirement for the initiation of collision/coalescence) (Ghosh *et al.*, 2005b); however, they are far fewer in number as compared to the sulphate aerosol particles. This is evident from Figure 3 where we observe a coarse mode at approximately  $35 \mu\text{m}$  (with a number concentration of  $0.12 \text{ N/cm}^3$  or  $123\,000 \text{ N/m}^3$ ) exceeding the critical size of  $20 \mu\text{m}$ . It is must also be borne in mind that our model results correspond to one cloud cycle spanning a vertical distance of approximately  $350 \text{ m}$ . Since the model results correspond to a non-entraining cloud parcel, the cloud liquid water increases adiabatically with height. For example, the liquid water content (lwc) increases from  $0.33 \text{ g kg}^{-1}$  at  $150 \text{ m}$  to  $0.84 \text{ g kg}^{-1}$  at  $350 \text{ m}$  above the cloud base. In reality, it is quite possible that aerosol spectra produced from one cloud cycle can remain aloft to be subjected to a



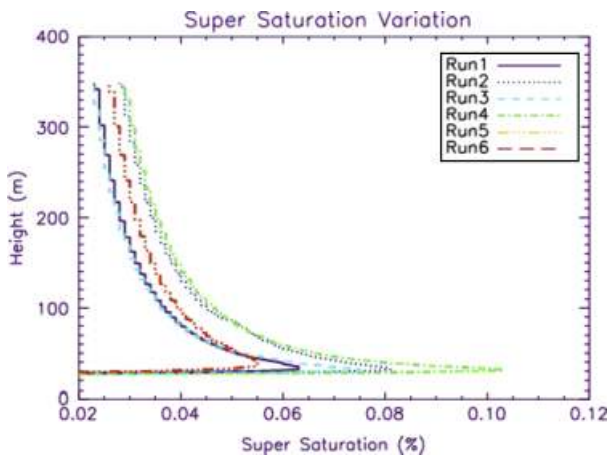
**Figure 5.** Droplet growth profiles for various runs. Note that the cloud base forms approximately at 26 m. The aerosol bins grow into activated cloud droplets only when the curves slope to the right. Note that the coarse-mode salt bins are always activated. Note also that not all of the fine-mode salt bins and the sulphate bins activate into cloud droplets. (a) Run 1 (normal aerosol loading and polluted run); (b) Run 2 (normal aerosol loading and clean run); (c) Run 3 (half aerosol loading and polluted run); (d) Run 4 (half aerosol loading and clean run); (e) Run 5 (double aerosol loading and polluted run); (f) Run 6 (double aerosol loading and clean run).

further cycle of cloud processing. This will result in a shift of the droplet spectra to larger sizes, thereby triggering collision/coalescence which will have to be treated as a time-dependent process.

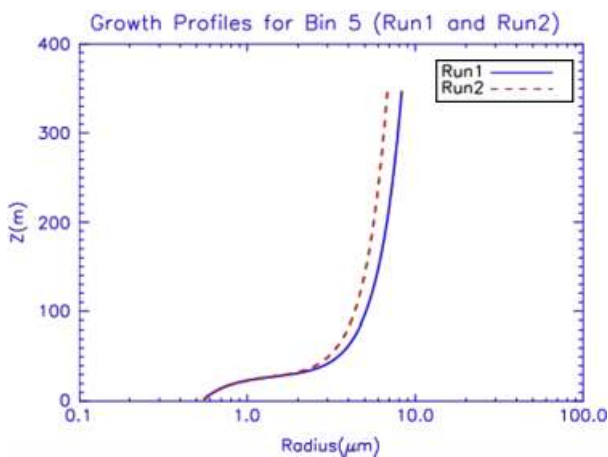
## 5. Concluding remarks

Chennai on India's southern tip receives air masses from mainland India as well as from over the oceans.

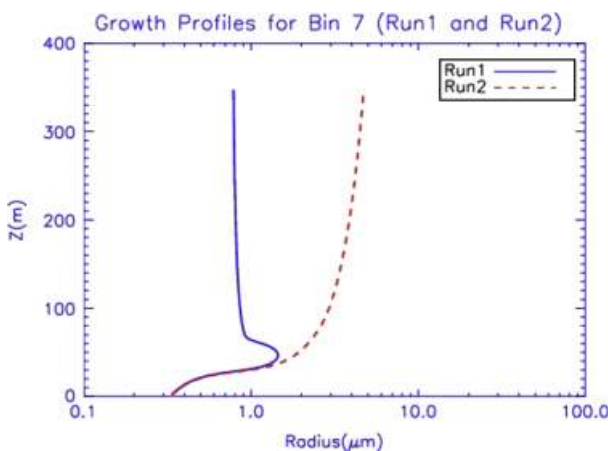
As a result, dry aerosol spectral measurements show multiple modes comprising of sea salt as well as ammonium sulphate particles. The city receives much of its rainfall during the NE monsoon rains (unlike the rest of the sub-continent where the SW monsoon activity is far stronger) and washout effects cleanse the city's air. Chennai is a coastal city with a hot tropical climate which is the city's saving grace – air pollution effects are ameliorated during part of the year because



**Figure 6.** Super-saturation profiles for the various model runs. Note that for the pairs of clean and polluted runs (i.e. Runs 1 and 2, Runs 3 and 4, Runs 5 and 6, the maximum super saturation attained is lower for the polluted run).



**Figure 7.** Growth profiles for a coarse-mode sulphate bin for clean and polluted cases. Note that the polluted run is marked by a higher growth rate than a clean run.



**Figure 8.** Growth profiles for a fine-mode salt bin for clean and polluted cases. Note that the clean run is marked by a higher growth rate than a polluted run.

of mixing and dilution effects. The Indian economy is developing at a rapid pace and we anticipate that

in the coming decade, both the aerosol loading levels as well as air pollution levels will increase which will profoundly affect the cloud microphysical development. Whilst observational data on the aerosol distribution are available from large-scale field campaigns such as the Indian Ocean Experiment (INDOEX) and ACE – Asia, there are no modelling studies to elucidate the mechanistic details of the aerosol activation process for the sub-continent. This is in contrast to the situation in Europe and the United States where both observational analysis as well as comparable modelling studies are available (IPCC, 2007). This is the first microphysical modelling study for the Indian sub continent where NEERI observations have been applied to cloud microphysical applications.

In this study we have shown simulations with varying levels of ammonium sulphate loading and for current levels of  $\text{SO}_2$  pollution over Chennai. Our main aim was to find out to what extent is the maximum super saturation lowered when a mixed population of aerosol particles grow in a humid atmosphere containing a condensable pollutant gas such as  $\text{SO}_2$ . We find that this effect is strong in comparison to the effects induced by changes in updraught speeds – we find that for Chennai the former effect can supersede the latter effect when the updraught speeds are less than  $0.5 \text{ ms}^{-1}$  (as is often the case in Chennai (NEERI, 2007)). We also find that in polluted air masses, some of the particles are more privileged than others since they grow more efficiently whilst suppressing the growth of some other aerosol particles. This suggests that in polluted air some of the aerosol particles, particularly the larger ones, grow more in polluted air masses than in clean air. This effect can be exacerbated for multiple cloud cycles that are possible over deep convective clouds- some may even grow to more than  $20 \mu\text{m}$  thereby triggering the onset of collision and coalescence. Only for the smallest salt bins, the reverse is true-the smallest particles activate for the clean cases more efficiently as the updraught speeds are double. These sensitivity studies have revealed that the process of aerosol activation comprising of a mixture of aerosol particles and grown in polluted air masses is complex and often non-intuitive. In a later study we intend to include biomass aerosol particles along with sea-salt and ammonium sulphate particles in the microphysical parcel model and study the spectral development of the cloud droplets. We anticipate significant amounts of biomass over Chennai as the city has many slums where fire wood and cow dung cakes are still used by millions of people for household cooking.

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