8.1 THE ROLE OF SEA-SPRAY IN CLEANSING AIR POLLUTION OVER OCEAN VIA CLOUD PROCESSES

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ABSTRACT

Tropical Rainfall Measuring Mission (TRMM) satellite observations have shown that anthropogenic aerosols suppress precipitation in convective clouds as deep as 6 km over land, and one would expect that the same would occur when these aerosols affect deep clouds over ocean. However, using similar methods, we show here that convective clouds developing in polluted air over ocean do precipitate and wash down the pollution aerosols. The probable cause for that is the sea spray, providing giant CCN that initiate the precipitation. Model simulations show that initiation of precipitation in deep clouds is much more sensitive to the sea spray than the initiation of drizzle in shallow clouds. Once precipitation have initiated, the air gets cleansed and eventually becomes "truly" maritime, i.e., containing small number concentrations of CCN, and shallow clouds that form in this air drizzle readily and possess low albedo. At the final account, it is the sea salt that helps cleansing the atmosphere from the anthropogenic air pollution, via cloud processes.

1. OBSERVED POLLUTED AND CLEAN CLOUDS OVER OCEAN

The INDOEX experiment showed how huge amounts of air pollution spill off the South Asia continent to the Indian Ocean and spread all the way to the ITCZ. These aerosols are composed of a mixture of smoke from biomass burning, urban air pollution and desert dust, all of which act to suppress precipitation by providing large concentrations of small CCN (Rosenfeld, 1999, 2000; Rosenfeld et al., 2001). This was confirmed in INDOEX by aircraft measurements in the trade-winds cumulus clouds over sea. The average droplet concentration in clouds feeding on the polluted air was 315 cm^{-3}, whereas in the pristine air it was only 89 cm-3. Respectively, drizzle concentration in polluted air was suppressed to 1/4 of the value in pristine air (McFarquhar and Heymsfield, 2001).

We wanted to see how these observations in the shallow clouds might translates to suppression of precipitation in the deeper convective clouds, using similar methods as previously applied to clouds forming in polluted air over land (Rosenfeld, 1999, 2000; Rosenfeld et al., 2001), and what happens when the clouds travel over ocean with the trade winds and eventually reach the ITCZ. According to these methods, using the TRMM measurements, clouds in the polluted air over land had r_{eff} smaller than the precipitation threshold of 14-μm (Rosenfeld and Gutman, 1994) and did not develop radar precipitation echoes when their tops were below the –10°C isotherm level (black curve in figure 1), which was at a height of about 6 km. Effect of precipitation suppression over land was recently simulated by Khain et al, (2001) using spectral-microphysics cloud model.

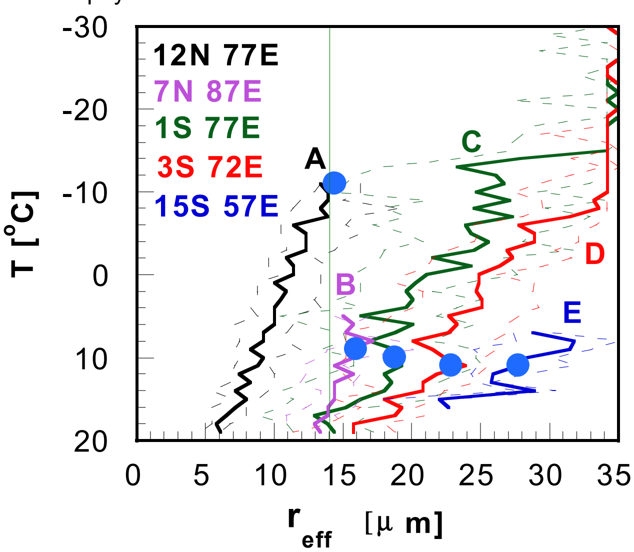

Figure 1: The temperature – effective radius (T-r_{eff}) relations as observed by TRMM satellite in deep convective clouds occurring in: (**A**) heavily polluted air over south India on 24 March 1999; (**B**) polluted air in the northeasterly trade winds on 11 February 1999 (**C**) reaching the northern rain band of the ITCZ, (**D**) and to the south of the northern cloud band of the ITCZ, still in northern hemisphere air mass; (**E**) and lastly in the southern hemisphere trade winds. The vertical green line marks the precipitation threshold of 14 μm. The blue dots denote the warmest cloud top temperature that still contained detectable TRMM radar precipitation echoes. Note that the r_{eff}<14 μm over land, so that precipitation is completely suppressed there below the -10°C isotherm, whereas deep clouds in polluted air over sea (curve B) precipitate readily, with some further decreasing threshold depth further to the south (C to E) in the cleaner clouds.

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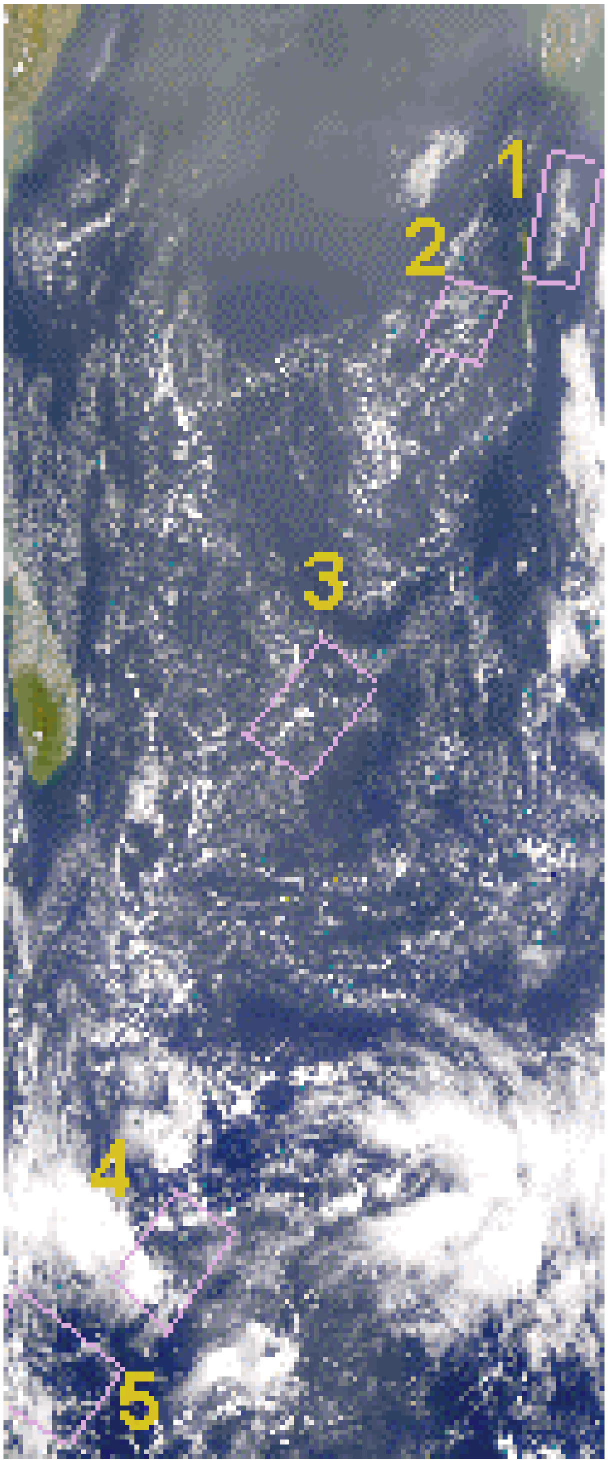

Figure 2: SEAWIFS image of clouds forming in polluted air flowing towards the ITCZ, over the Bay of Bengal and the Indian Ocean to its south, on 11 February 1999, 07:39 GMT, about two hours after the TRMM overpass. Frame 1 is between Myanmar and Andaman Islands. Frames 1, 2 and 3 correspond to curves B, C and D of Figure 3, respectively. Frames 3, 4 and 5 correspond to curves B, C and D of Figure 1.

The amount of pollution in the air did not diminish much when it moved over sea for many hundreds of km, as indicated by the consistent observed turbidity of the air (see Figure 2). This was quantified by modal aerosol optical depth (AOD) of about 0.35 for areas 2 and 3 of Figure 2, reducing to about 0.27 in area 4 upon reaching the ITCZ. However, clouds developing in the polluted air over sea had larger r_{eff} than clouds of the same depth over land, and increasingly so further away from land (figure 1). The TRMM measurements show that unlike polluted clouds over land, deep clouds developing in polluted air over ocean behaved like maritime clouds, with rainout signature (i.e., the air goes up, but the rain forms quickly and falls through the updraft) as described by Rosenfeld and Lensky (1998), and warm glaciation temperature as inferred from the indicated r_{eff} reaching its saturation value near -10^oC. The TRMM precipitation radar showed that clouds over sea precipitated at intensities > ~1 mm hr^{-1} almost always when their top exceeded the height of 3 km (about 10^oC isotherm), with no obvious differences in the precipitation echoes from clouds forming in the polluted and pristine air. The observation that the r_{eff} at a given cloud depth was greater in clouds at longer distance downwind away from the pollution source over land suggest that the pollution particles affected the cloud microstructure to produce smaller droplets, but unlike over land, this did not seem to suppress the precipitation in the clouds over ocean.

Why do the deep clouds react so differently to the pollution over land and over ocean? Potentially relevant differences between land and ocean are:

- Sea spray forms large (diameter>1-μm) sea salt aerosols, that are the first to form droplets at cloud base, therefore reducing the super saturation there and preventing the activation of smaller pollution aerosols into cloud droplets (Cooper et al., 1997; O'Dowd et al., 1999; Yin et al., 2000). This reduces the droplet number concentrations and leads to enhanced coalescence, which can progress sufficiently to form rain in convective clouds deeper than about 3 km.

- Updraft velocities over ocean are weaker than over land, therefore a smaller fraction of the aerosols is nucleated into cloud droplets (Zipser and LeMone, 1980; Lucas et al., 1994). In addition, more time is available for the calescence to progress and form warm precipitation.

2. SATELLITE INFERENCES FOR SEA SPRAY INITIATING PRECIPITATION

Additional satellite observations of clouds occurring in polluted air mass moving from land to sea (figure 3) provided insight to the relative importance of these two potential explanations. An area of clouds with elevated basis (curve B) had the same small r_{eff} as the clouds upwind over land (curve A). When low-level clouds formed in addition to the high base clouds the r_{eff}

increased, especially near the base (curve C). Further downwind, when elevate base clouds disappeared and all the clouds were fed from the low level base, the r_{eff} increased further with decreasing T or increasing height (curve D). The r_{eff} further increased downwind towards the ITCZ and beyond, as shown in figure 1.

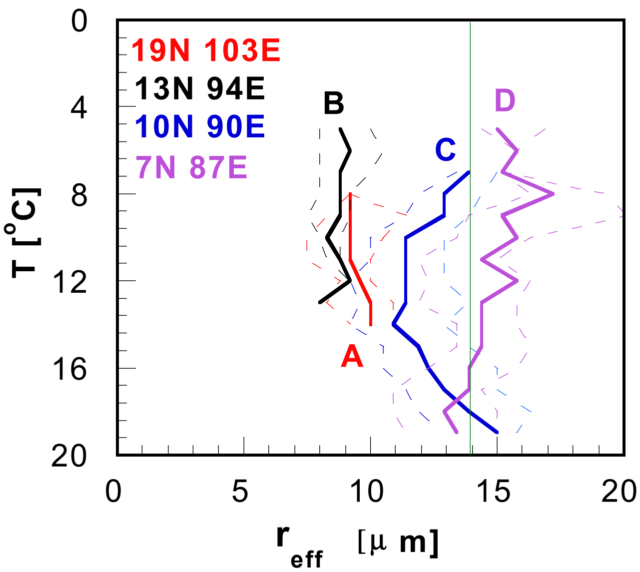

Figure 3: T-r_{eff} relations as in figure 1, for clouds developing in (**A**) heavily polluted air over north Thailand on 11 February 1999, moving to the (**B**) north Bay of Bengal and forming elevated base clouds with small r_{eff}, but when clouds with low bases are added (**C**) the lower (warmer) clouds have much larger r_{eff}, that (**D**) expand to the whole cloud depth when only the low base clouds remain. The r_{eff} continues growing further downwind, as shown in Figure 1.

A possible explanation of these observations would be that the air pollution from the land overrides a cleaner marine air near the surface. However, according to back-trajectory analysis using the NOAA-HYSPLIT model (1997), the wind direction was from land at the surface level. The AOD was about 0.35, and ship aerosol and LiDAR measurements showed that the pollution was generally confined to the lowest three km and reached the surface (Lelieveld et al., 2001). Another possible explanation is the updraft velocity at cloud base, forming smaller droplets for greater upward velocities. However, the elevated clouds seem at the satellite imagery less convective than the clouds with the lower bases, therefore highly unlikely having greater updrafts at cloud base or higher. The remaining possible explanation is the presence of large sea salt CCN at the marine boundary layer of the atmosphere, but not at the higher levels or over land.

Sea salt aerosols that originate from sea spray are known to be added to the air mass during the travel over ocean, at a size distribution that is closely related to the surface wind velocity (Woodcock, 1953). LiDAR measurements (Sugimoto et al., 2000) show that the sea salt particles are nearly evenly distributed within the mixed layer up to the cloud base, where they are ingested with the updrafts into the clouds. Apparently the impact of the sea spray was able to compete with the pollution particles and create clouds with moderate average concentrations of droplets (250 to 400 cm^{-3}), as measured by McFarquhar and Heymsfield (2001). This droplet concentration was sufficient at most cases to prevent precipitation from stratocumulus clouds, which would be in the form of drizzle for these shallow clouds (Albrecht, 1989). Albrecht (1989) noted that the drizzle develops in stratocumulus containing droplet concentration less than about 150 drops cm^{-3}, and droplet radii > ~15 μm. He also recognized that the drizzle washes down the CCN and so causes the subsequent clouds to contain smaller concentrations of larger drops, thereby creating more favorable conditions for drizzle, establishing a positive feedback loop, which in the extreme leads to elimination of all the CCN and collapse of the marine boundary layer and dissipation of the clouds (Ackerman et al., 1993). Conversely, stratocumulus clouds in polluted air develop very little drizzle, so that the pollution is not washed down by cloud processes.

3. SIMULATED PRECIPITATION FROM POLLUTED CONVECTIVE CLOUDS OVER LAND AND OCEAN

According to the TRMM Precipitation Radar (PR), the polluted clouds start precipitating when their tops exceed about 3 km above sea level (see Figure 1). The PR cannot detect drizzle in the shallow pristine maritime clouds, because of the strong dependency of the echo intensity on the drop size.

Apparently, the height that polluted cloud tops have to exceed for start precipitating is much smaller over ocean (~3 km) than over land (~6 km). The satellite analysis presented in Figure 2 suggests that the giant CCN from the sea spray plays a major role in this difference. This qualitative assessment was further investigated using a one-dimensional warm rain cloud parcel model (Khain et al, 2000) with 2000 size bins for CCN and drops, for obtaining high accuracy of the diffusional growth of CCN and cloud droplets and coalescence processes.

The aerosols were taken from the measurements of NOAA's research vessel Ron Brown in the Indian Ocean during February and March 1999. Four spectra were extracted:

- Pristine maritime, at Julian day 79.271 near the southern edge of the cruise, in little-polluted maritime air to the south of the ITCZ, denoted as "clean" in Figure 4.
- Polluted air over sea near the northern edge of the cruise, at Julian day 69.25, denoted as "Polluted" in Figure 4.
- Polluted air of the second spectrum, but with sea spray truncated to Yunge distribution at radii>0.6 μm, denoted as "Pol, R<0.6" in Figure 4.
- Polluted air of the second spectrum, but with sea spray truncated to Yunge distribution at radii>0.3 μm, denoted as "Pol, R<0.3" in Figure 4.

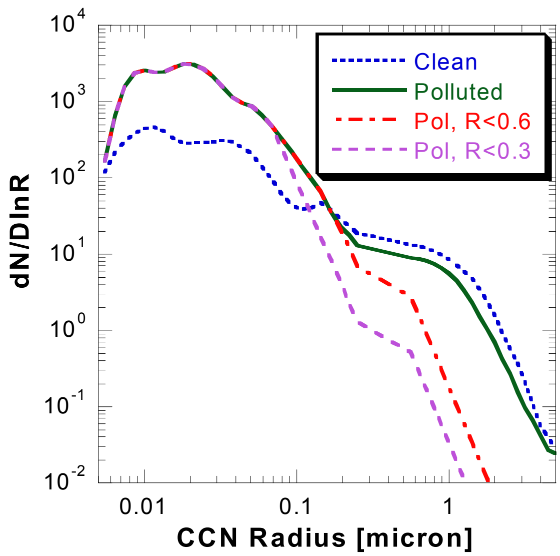

Figure 4: CCN aerosol size distribution in equivalent NaCl particles, used for the cloud simulation. The distributions are for clean air with sea spray (SS), polluted air with SS, and polluted air with SS eliminated below radius of 0.6 and 0.3 micron.

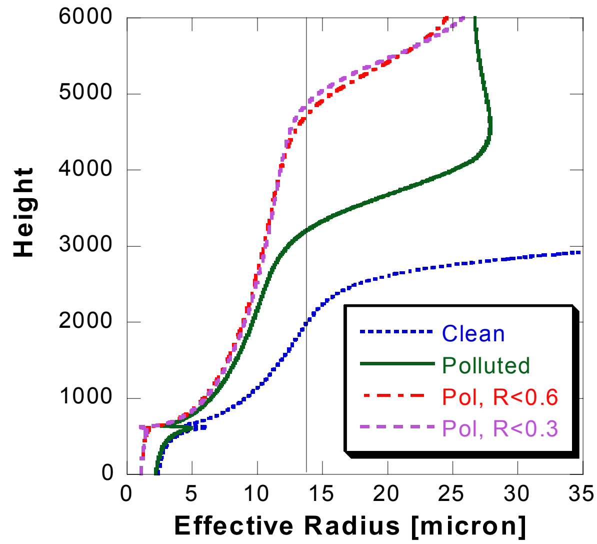

Figure 6: Simulated effective radius of cloud drops as a function of height for the four aerosol spectra shown in Figure 4. The vertical line at 14 μm is the precipitation threshold (Rosenfeld and Gutman, 1994).

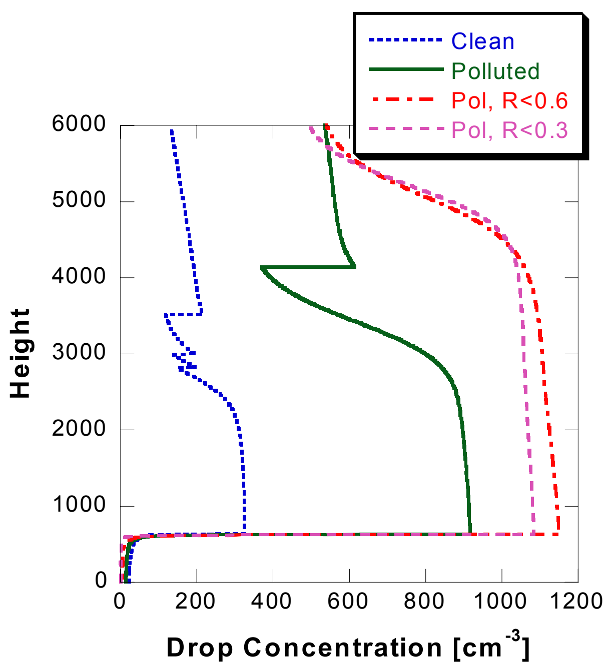

Figure 5: Simulated drop concentration as a function of height for the four aerosol spectra shown in Figure 4.

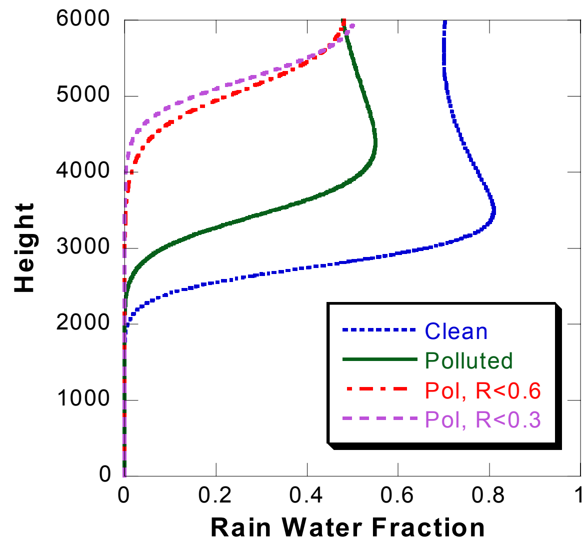

Figure 7: Simulated drop concentration as a function of height for the four aerosol spectra shown in Figure 4.

The measured aerosol spectra were converted to NaCl equivalent particles by to the following procedure:
Aerosols smaller than diameter of 1 μm were assumed to be mostly pollution particles, and were assumed to contain 15% equivalent of NaCl.
Aerosols greater than 1 μm were assumed to be exclusively composed of sea spray, and left as is. This is justified by selecting the polluted case with large aerosols in concentrations that do not exceed those of the maritime case, where the aerosols > 1 μm can be assumed to be exclusively sea salt.
Polluted aerosols over land, without sea spray, were simulated by replacing the large aerosols greater than diameter of 0.6 or 1.2 μm by Yunge distribution.

The simulated cloud base updraft was 1 ms^{-1}, and the vertical velocity increased with height, reaching a maximum of 9 ms^{-1} at height of 5 km. These are typical vertical velocities for maritime environment, but smaller than typical for continental convective clouds.

The main results of the simulations are:

- The effective radius of 14 μm as a precipitation threshold (Rosenfeld and Gutman, 1994) is confirmed.
- The tops of polluted clouds without sea spray have to exceed the height of 5 km for start precipitating.
- The tops of polluted clouds with sea spray has to exceed the height of 3 km for start precipitating, 2 km less than without sea spray.
- The unpolluted clouds start developing significant precipitation at 2.3 km, 700 m lower than the polluted clouds with sea spray.
- There is little sensitivity to the exact size of the truncation of the sea spray. This means that the larger sea spray particles are dominating the restoration of the precipitation in the polluted clouds.
- The sea spray in the polluted air decreased the maximum cloud base super saturation to the extent that greater super saturation formed higher in the cloud, where additional CCN were activated into cloud droplets. The increase in super saturation is observed in the area of active coalescence that leads to a decrease in drop concentration within cloud updraft. This explains the sudden increase in the drop concentration with height shown in Figure 5.
- The renewed nucleation of small droplets aloft in the polluted air with sea spray explains the observed smaller r_{eff} aloft in the precipitating clouds that form closer to the pollution source (see the r_{eff} above the blue dots in Figure 1).

Additional simulations (not shown) tested the sensitivity to the cloud base updraft. As expected, all heights increased with greater updraft velocities, but the relative changes remained similar. The higher base of clouds over land and the greater cloud base updrafts that typically occur there can explain the observation of clouds exceeding 6.5 km before start precipitating (Curve A in Figure 1). Similar observations were obtained by satellite and aircraft for clouds in polluted air over Texas (Rosenfeld and Woodley, 2000), Thailand (Rosenfeld and Woodley, 1999), Indonesia (Rosenfeld and Lensky, 1998; Rosenfeld, 1999) and the Amazon (Rosenfeld and Woodley, 2001).

The simulated drop concentrations should be compared to the peak concentrations in the aircraft measurements, which represent the undiluted parcel at the cores of the updrafts. The peak concentrations are roughly the double of the average concentrations. Given that, there is a reasonable agreement between the model results and the aircraft measured average drop concentrations.

4. SUMMARY AND CONCLUSIONS

The winter monsoon is a flow of cool and highly polluted air mass off the Southeast Asia continent to the relatively warm waters of the ocean. The warming and moistening of the air at the sea surface as it flows towards the ITCZ causes cumulus convection with tops often reaching 3 km, becoming gradually deeper with the approach to the ITCZ.

The particulate air pollution would have caused these convective clouds to become microphysically “continental”, and avoid precipitating, as observed for their counterparts that form over land in the afternoon (Curve A in Figure 1). However, the sea spray “seeds” the clouds over sea and affects them by two ways:

- Provide giant CCN that create initial larger cloud droplets that start the coalescence processes.
- Decrease the maximum super saturation at cloud base, and so prevent the nucleation of the smaller pollution particles into cloud drops. This reduces the cloud number concentrations, so that the condensed vapor must be divided between smaller number of drops, which then become larger and coalesce faster into raindrops.

The bulk of the cloud droplets is nucleated on the larger pollution particles. The sea spray adds only a small number of large droplets that coalesce with the other drops and eventually precipitate them as raindrops. At the final account, the sea spray cleanses the atmosphere from the pollution particles that nucleated cloud droplets. The second generation of clouds that forms in the air is already cleaner, with smaller drop number concentration thus more efficient coalesce and further cleansing by rainfall. This process eventually converts the clouds to become microphysically “maritime”.

This cleansing process would advance slower in shallower clouds, because a minimum depth is required to form precipitation, which according to the cloud simulation here is about 3 km for the marine polluted clouds (see Fig. 3). The best conditions for cleansing air pollution over sea would be deep clouds and high surface winds, that raise much sea spray. If the oceans were no salty, air pollution would remain much longer in the lower troposphere and spread to much greater areas of the oceans. The clouds would have to grow vertically to about 5 km for start precipitating in polluted air over a hypothetical salt-free ocean. This height is well above the tops of common convective clouds under high subtropical inversion, and therefore such deeper clouds cover much smaller areas of the oceans, leaving a major role for the sea spray in making the difference between non-precipitating to precipitating clouds.

At the bottom line, it is the sea salt that serves as catalyst to cleanse by cloud processes the atmosphere from the anthropogenic air pollution, and plays a major role in making the oceanic air mass so pristine.

5. ACKNOWLEDGEMENTS

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