P1.6 AEROSOL INTERCATIONS ON CLOUDS WITH EMPHASIS ON THE ARABIAN PENINSULA

R. T Bruintjes[1], V. Salazar[1], D. Breed[1], Jia Li[2], Peter Buseck[2], T Jensen[1], K. Ross[3], S. Piketh[3] and J. Reid[4]

[1]National Center for Atmospheric Research, P.O. Box 3000, Boulder, CO 80307, USA.
[2]Arizona State University, Tempe, Arizona, 85287, USA
[3]University of Witwatersrand, Johannesburg, South Africa)
[4]Naval Research Laboratories, Monterrey, California

1. INTRODUCTION

Biomass burning and human activities are releasing large amounts of aerosols into the atmosphere. In addition, desert dust particles have increased substantially in the atmosphere over the past 30 years due to larger areas of soil exposed to the atmosphere and the drying of lake beds. These particles enhance scattering and absorption of solar radiation. They also produce brighter clouds that are less efficient at releasing precipitation. These in turn lead to large reductions in the amount of solar irradiance reaching Earth's surface, a corresponding increase in solar heating of the atmosphere, changes in atmospheric temperature structure, suppression of rainfall, and less efficient removal of pollutants. These aerosol effects can lead to a weaker hydrological cycle, which connects directly to the availability of fresh water that could result in a major environmental issue (Twomey, 1977; Albrecht, 1989, Ramanathan et al. 2001).

Aerosol effects on clouds and precipitation is obviously complex - a multi-order problem. Evidence of cloud and precipitation changes due to aerosols (changes in "natural" Cloud Condensation Nuclei (CCN) and Ice Nuclei (IN)) is becoming widespread. There is ample evidence that biomass burning, desert dust and other anthropogenic sources of aerosols affect the radiative properties of clouds and precipitation processes in clouds leading also to changes in the dynamical processes in clouds (i.e. effects on cloud structures and lifetimes). Increased CCN lead to higher droplet concentrations and narrower droplet spectra (which manifests itself as a higher cloud albedo) leading to suppressed drizzle formation and longer lasting stratiform clouds (ship-track studies, Albrecht, 1989). Desert dust on the other hand affects ice processes by providing efficient IN. However, when and if desert dust gets coated with sulfates it might act as a giant CCN further complicating aerosol-cloud effects. Recent satellite studies of cloud microstructure downwind of biomass burning in Indonesia, Australia, and other areas have suggested similar effects (suppressed precipitation formation in the affected clouds, Rosenfeld, 2000). The intriguing evidence of increased positive lightning flashes in storms affected by the Mexican fires of 1998 is yet another example of the multi-order effects aerosols have on clouds, precipitation and the microphysics relevant to cloud electrification (Lyons, 1998).

The effects of aerosols on cloud microphysical and dynamical processes and precipitation were also highlighted by the results from experiments to enhance rainfall by hygroscopic seeding. Evidence exists that the particles produced by hygroscopic flares: 1) broaden the droplet spectrum and 2) seem to increase the lifetime of precipitating convective clouds possibly by changing the downdraft/updraft structures (WMO, 2000).

Biomass burning particles, desert dust, sulfates from anthropgenic sources and hygroscopic seeding are entrained into clouds and participate in the microphysical processes in clouds. The combined affect of these particles determine cloud microphysical and dynamical processes.

2. AFFECTS OF DIFFERENT PARTICLES

The initial droplet spectra in clouds depend on the chemistry, size and concentration of CCN. Recent results from biomass burning experiments indicate that the chemistry of particles initially emitted from fires undergo chemical transformations in the atmosphere that affect both the chemical composition and the size of the particles (Li et al, 2003, Posfai et al., 2003). More KCl particles occur in young smoke (Fig. 1), whereas more K_2SO_4 and KNO_3 particles are present in aged smoke.

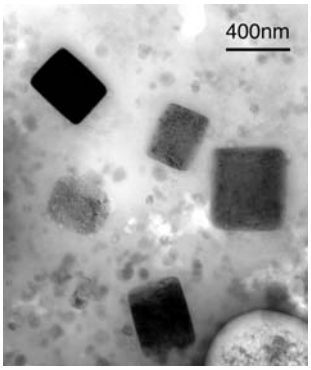

Figure 1: TEM images of the initial KCl particles emitted from savannah fires in Southern Africa.

Corresponding author's address: Roelof T. Bruintjes, National Center for Atmospheric Research, P.O. Box 3000, Boulder, CO 80307, USA; Email: roelof@ucar.edu

This change indicates that KCl particles from the fires were converted to K_2SO_4 and KNO_3 through reactions with S- and N-bearing species from biomass burning as well as other sources (Fig. 2). These chemical reactions have been previously observed (Posfai et al., 1995) and seem to be similar to those observed in polluted marine environments as reported by McInnes et al. (1994). These reactions were also found in recent studies in the United Arab Emirates where large amounts of SO_2 are emitted into the atmosphere from the oil-related industries (Fig. 3). Within a half hour of emission the KCl particles were converted to K_2SO_4 and KNO_3.

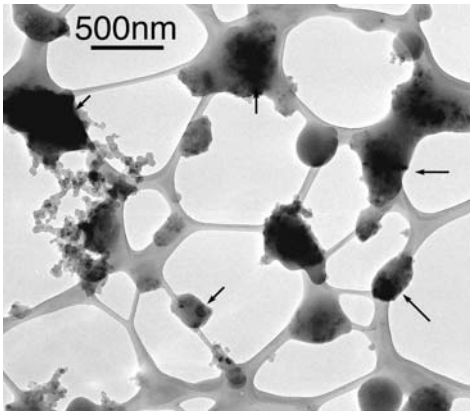

Figure 2: TEM images from reacted particles in smoke plumes that aged for more than 20 minutes from two different fires in southern Africa. Particles are soot, tar balls and KSO_4 and KNO_4 (arrowed) with possible organic coatings.

Figure 3: TEM image of sea-salt and partially reacted sea-salt particles over the UAE.

These data suggest that anthropogenic pollution and biomass-burning aerosol particles experience chemical transformations as they age and interact with other atmospheric gases and particles that, in turn, modify their efficiency as CCN particles and hence cloud characteristics (Bruintjes et al., 2004).

Less known is how the aerosol particle size distributions change during the chemical transformation processes. If changes do occur during these transformations, they may have important implications for the CCN activity and the characteristics of the initial cloud droplet size distribution and precipitation development in clouds growing in these environments.

The concentrations of the initial KCl particles produced by the fires seem to be a function of the flaming temperature of the wildland fires (Ward and Hardy 1991). It is assumed that the potassium and chlorine salts are released from the biomass material and volatilized by the extreme temperatures in the flames before re-condensing into particles during the cooling stage. Ward and Hardy (1991) found that fires with higher intensity (longer flame lengths) produce proportionately larger particles than in low-intensity and smoldering fires. However, they did not investigate the differences in chemistry between the particles. Larger and smaller particles, especially if they are inorganic salts or inorganic salts mixed with other substances will behave differently as CCN. In addition, chemical transformation processes may also depend on the sizes of the initial particles produced. These effects may have important effects on cloud processes.

During the Feasibility study for rainfall enhancement in the United Arab Emirates the effects of desert dust on cloud were also investigated. An important aspect that was studied during the 2002 field efforts was the effect of sulfate coated desert dust on the precipitation process in clouds. To assess the impact of the desert dust, particles were collected on filters that were analyzed using transmission electron microscopy (TEM). TEM and SEM images of the particles indicated that all three major aerosol types occur over the UAE (mineral dust, sea salt and anthropogenic (sulfates)) and are abundant over most regions of the UAE. While many of the desert dust particles contained sulfate incursions on them (Fig. 4), there was no clear indication of dust particles completely coated with sulfate. Instead, aggregates of sea salt (Fig. 3), sulfates, and mineral dust were found to be most common during all periods of observations.

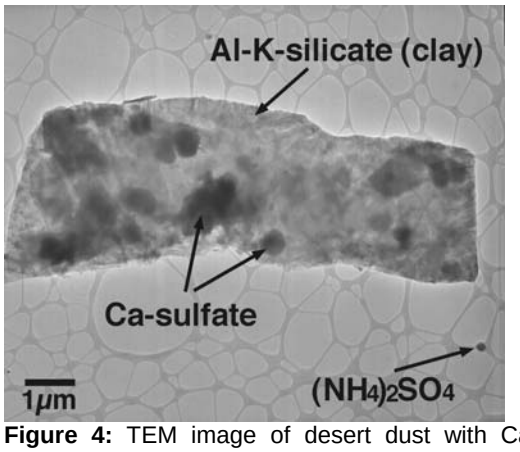

Figure 4: TEM image of desert dust with Ca-sulfate incursions.

The mineral dust particles comprised most of the particles larger than 1 μm in diameter and all the particles larger than 10 μm diameter. The sea salt particles comprised most of the particles in the size range 0.5 to 5 μm diameter but it was evident that many of them had already reacted or partially reacted with sulfate-bearing species.

The above-mentioned studies indicate that in order to increase our understanding of aerosol-cloud interactions and precipitation development in clouds in natural and polluted environments, we need to integrate chemistry and cloud microphysical measurements. Very few studies in the past have integrated these measurements.

3. CASE STUDY (SAUDI ARABIA)

During a 35-day field effort in July and August 2004 as part of a feasibility study for rainfall enhancement in the Abha region of Saudi Arabia, aerosol and cloud measurements indicated complex interactions and strong temporal and spatial variations in these effects. The vertical profile of aerosol measurements on two days (16 July and 28 July 2004, Figs. 5) over the Red Sea just of the southern coast of Saudi Arabia indicated widely different sources and types of aerosols entering the region and interacting with the clouds.

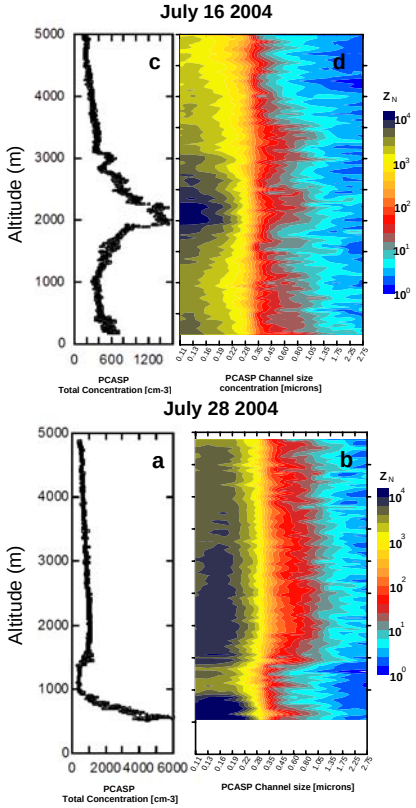

Figure 5: PCASP vertical profile from 300m over sea level up to 5 kilometers over the Red Sea during July 16 and 28 2004. $Z_N=Log_{10}(dN/dr)$

While the 16 July measurements were conducted during the early morning before the onset of the sea-breeze the measurements on 28 July were conducted during mid-afternoon after the onset of the see-breeze. The aerosol concentrations on 28 July were substantially larger than on 16 July with a peak concentration near the surface and fairly "clean" layer between 1000 and 3000m MSL with higher concentrations aloft. The concentrations of particles on 16 July 2004 peaked at around 2000m MSL and according to satellite and model simulations were most probably biomass burning particles advected from Africa (Fig 6).

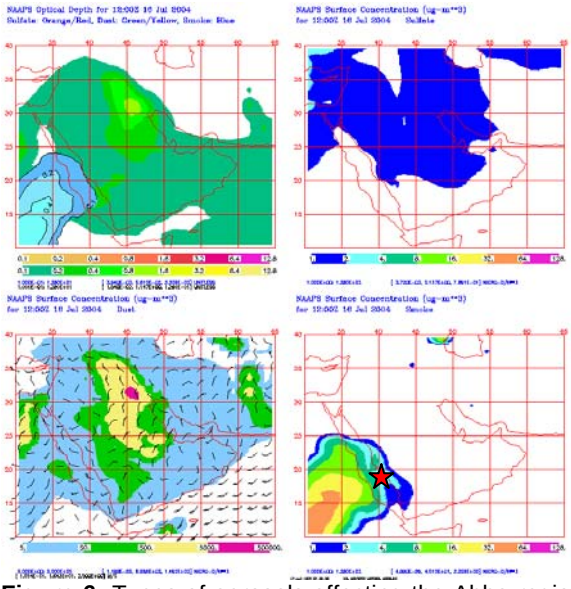

Figure 6: Types of aerosols affecting the Abha region on 16 July 2004.

While the particles near the surface on 28 July were generally small (~0. to 0.2μm diameter) the particles in the upper layer were somewhat larger with higher concentrations in sizes larger than 1.6μm diameter (Fig. 5). The low-level particles could have been due to ships in the area or DMS production by the ocean while the higher level particles were most probably dust (Fig. 7).

On both days these particles were advected inland with airflow and affected cloud microphysical properties in the region. The concentration of cloud droplets around 16 July was the highest concentrations measured during the field program Fig. 8). This was also the period with the strongest influx of biomass burning particles during the field campaign. In addition, the mean diameter, mean volume diameter and effective radius of the cloud droplets showed a minimum during this period. The 28 July period generally showed lower concentrations of cloud droplets but the other parameters were fairly similar indicating that the CCN activity of these particles was most probably less effecient.

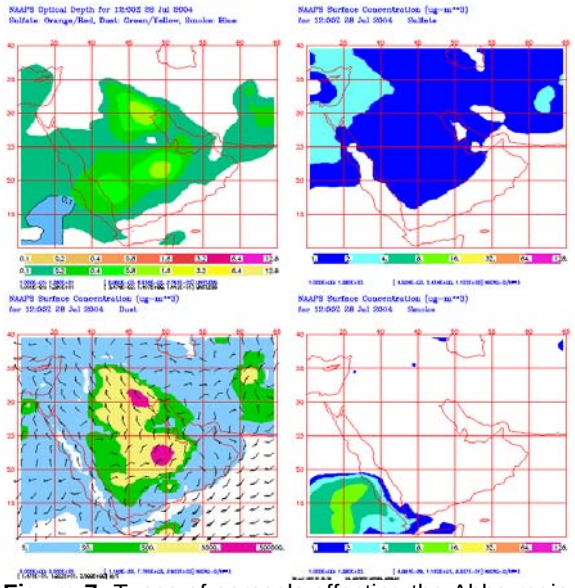

Figure 7: Types of aerosols affecting the Abha region on 28 July 2004.

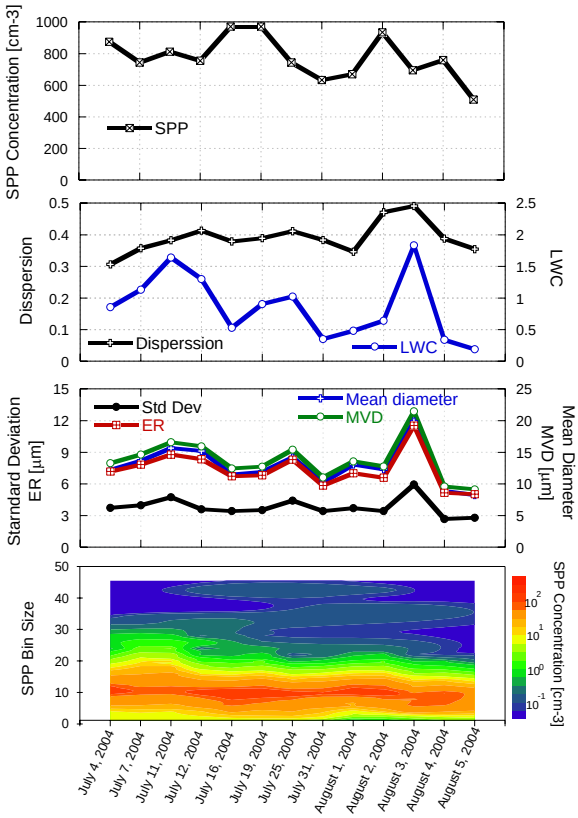

Figure 8: Daily peak cloud droplet concentrations, liquid water content, standard deviation, dispersion, mean diameter, mean volume diameter, and effective radius during the field program period.

These are only preliminary analyses at this stage and many affects of the different particle types on cloud microphysical processes needs to further explored. However, the data indicate that cloud microphysical processes in one area may be highly dependent on the influx of type and concentration of particles and variations in types and concentrations could affect cloud microphysical processes significantly. However, new satellite and model data for the first time now provide the opportunity to further explore these differences and their affects on cloud microphysical and precipitation processes. These differences will also have important implications for cloud seeding operations to enhance rainfall or suppress hail.

In addition to the role of biomass burning particles as natural CCN from marine and biomass burning sources, KCl and NaCl are also used in hygroscopic flares for cloud seeding to enhance rainfall (WMO 2000). Modeling studies have indicated that a particle diameter of ~ 1 μm should be optimal for cloud seeding to enhance rainfall (Cooper et al, 1997). Recent studies at NCAR indicate that the hotter the flare burns, the larger the salt particles produced by the flares. While it was initially thought that cooler burning flares would produce larger particles it should be noted that the flare burning temperature is comparable to the temperatures observed in wildland fires. There is also circumstantial evidence that hotter wildland fires produce larger particles (Ward and Hardy, 1991). The size of the re-condensed particles is possibly related to cooling rate. The hotter the flare burns the larger the rate of cooling. Because the hygroscopic particles are released at cloud base, they would be directly incorporated in cloud droplets before chemical reactions could take place as found in the biomass burning and polluted marine experiments.

4. CONCLUSIONS

Cloud condensation nuclei (CCN) and Ice nuclei (IN) are the particles that form cloud droplets and ice crystals, a small fraction of which may eventually form raindrops. The objective of hygroscopic seeding is to alter the natural CCN to enhance the formation of the select few cloud droplets that become raindrops while glaciogenic seeding attempts to enhance precipitation formation by increasing ice crystal concentrations and buoyancy effects through the freezing of supercooled water. Thus, actual measurements of CCN and the natural droplet spectra, and ice nuclei and ice formation should be an important objective of the field program. In addition, ice processes also depend on the characteristics of the cloud droplet spectra. The background aerosol characteristics and spatial and temporal variations therefore play an important role in determining the cloud microphysical characteristics and associated precipitation development. This again will have a direct affect of the effectiveness of cloud seeding operations.

Some general conclusions are summarized as follows:

- Biomass burning and industrial particles are mostly sub-micron in diameter with KCl dominant initially in flaming fires.

- Initial KCl particles react to form potassium sulfate and nitrate with the latter usually smaller.
- Flare and biomass burning temperatures both range between 1000 and 2000^{o}C.
- Hot flaming fires and flares volatilize potassium and chloride and nucleate as KCL particles.
- The size of re-nucleated particles possibly depends on cooling rate. Experiments are ongoing to quantify these effects.
- Measurements in both forest fires and flare seeding indicate that several complex processes act simultaneously to produce and modify the CCN characteristics.
- Hotter burning flares seem to produce larger particles. This is different than originally anticipated.
- CCN activity is continuously changed in the atmosphere through chemical gas-to-particle and particle-to-gas processes.
- Spatial and temporal variations of aerosol concentrations, composition and sizes are important for cloud microphysical processes in clouds
- New information from satellites and models should be used in real-time to determine and evaluate seeding operations.

These results have direct implications for the dispersing and targeting of hygroscopic seeding material, especially in polluted environments. Based on these experiments it may be possible to tailor the particle size distribution from the flares to specific situations and environments. The flare development technology also provides an excellent opportunity to include other materials such as desert dust and organic materials in the flares in the future to study the specific effects of these substances on water and ice nucleation and other microphysical processes in clouds.

5. ACKNOWLEDGEMENTS

This study was supported by the Southern African Regional Science Initiative - SAFARI 2000. This research was sponsored by a grant (NAG5-7939) from NASA through the University of Virginia to NCAR, Ice Crystal Engineering, The Presidency of Meteorlogy and Environment in Saudi Arabia and the Department of Water Resources Studies (DWRS) in the United Arab Emirates. The ASU research was funded by a grant from the NASA Radiation Science Program. Samples were collected on board the UW-CV-580 under the direction of Peter Hobbs. The National Center for Atmospheric Research is sponsored by the National Science Foundation.

6 REFERENCES

Albrecht, B.A., 1989: Aerosols, cloud microphysics, and fractional cloudiness. Science, **245**, 1227-1230.

Cooper, W.A., R.T. Bruintjes, and G.K. Mather, Calculations pertaining to hygroscopic seeding with flares. *J. Appl. Meteor.*, **36**, 1449-1469, 1997.

Li, J., M. Posfay, P.V. Hobbs, and P.R. Buseck, 2003: Individual aerosol particles from biomass burning in southern Africa: 2. Compositions and aging of inorganic particles. J. of Geophys. Res., **108**, D13, 8484, SAF20.

Lyons, W. A., T. E. Nelson, E. R. Williams, J. A. Cramer, and T. R. Turner, Enhanced positive cloud-to-ground lightning in thunderstorms ingesting smoke from fires. *Science. **282***:77-80, 1998.

McInnes, L.M., D.S. Covert, P.K. Quinn, and M.S. Germani, Measurements of chloride depletion and sulfur enrichment in individual seasalt particles collected from the marine boundary layer, *J. Geophys. Res*., **99**, 8257-8268, 1994.

Posfai, M., J.R. Anderson, P.R. Buseck, and H. Sievering, Compositional variations of sea-salt-mode aerosol particles from the North Atlantic, *J.Geophys. Res*., **100**,23063-23074, 1995.

Posfai, M., R. Simonics, J. Li, P.V. Hobbs, and P. R. Buseck, Individual aerosol particles from biomass burning in southern Africa: 1. Compositions and size distributions of carbonaceous particles. J. of Geophys. Res., **108**, 8483, SAF20, 2003.

Ramanathan, V, P.J. Crutzen, J.T. Kiehl, and D. Rosenfeld, 2001. Aerosols, Climate, and the hydrological cycle. *Science* **294**, 2119-2124.

Rosenfeld, D. 2000: Suppression of rain and snow by urban and industrial pollution. *Science*, **287**, 1793-1796.

Twomey, S., 1977: The influence of pollution on the short-wave albedo of clouds. *J. Atmos. Sci*., **34**, 1149-1152.

Ward, D.E., and C.C. Hardy, 1991: Smoke emissions from wildland fires. Environmental International, **17**, 117-134.

World Meteorological Organization (WMO), 2000: Report of the WMO workshop on hygroscopic seeding, *WMP Report No. 35, World Meteorological Organization*, WMO/TD No. 1006, Geneva, Switzerland, 68pp.