Aerosol Size Distributions & Optical Properties Found in the Marine Boundary Layer Over the Atlantic Ocean


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Investigation Overview

Purpose

- investigate marine aerosols
- emphasizing submicron particles (r < 0.05 µm)

Part 1

- Size distribution data & growth scheme in remote tropics

Part 2

- Analysis of measured and calculated properties
1983 Lynch Cruise

✈ Sailed from Charleston, SC to the Canary Islands, and onto Scotland

✈ 3/11 – 4/17, 1983

Fig. 1. Map showing the 1983 Lynch cruise track.
Instrumentation

- Aerosol Size Distributions
  - *Differential Mobility Analyzer (DMA)* (0.006-0.5 µm radius)
  - *Calspan Impactor* (0.5-50 µm particle radius)
- Total Aerosol Concentrations
  - *Gardner Counter*
- Continental Air Mass Detection
  - *Radon Monitor* (Radon 222)
- Optical Depth
  - *Sun Photometer*
- Aerosol Light Scattering
  - *Integrating Nephelometer & Visiometer*
1. Apply **electric charge** to particles
2. Particles **migrate** (mobility = size & charge) into clean sheath air flow under electric field
3. A narrow range of particle mobilities are **separated** from the aerosol by withdrawing a portion of the sheath air flow
4. Varying the electric field provides a range of particle mobilities, and the concentration in each range can be measured
5. Convert to a size distribution by using the distribution of charges produced by the charger and the known relation between mobility and size.

http://www.cac.yorku.ca/people/mozurke/Analyzer.htm (accessed 1/22/04)
Sun Photometer

1. Sunlight causes the LED detector to generate an **electrical current**
2. LED current **transformed** to a voltage and boosted by the **amplifier**
3. The voltage is measured with a digital voltmeter

http://www.concord.org/haze/spworks.html (accessed 1/22/04)
Nephelometer

- Detects scattering properties by measuring light scattered by the aerosol and subtracting light scattered by the gas, the walls of the instrument and the background noise in the detector
- Light that passes through the lens is separated by dichroic filters into wavelengths
- The reflected light passes through a filter into a photomultiplier tube
- Nephelometer measures extinction, caused by scattering and absorption of light, over a distance

\[ b_{\text{ext}} = b_{\text{scat}} + b_{\text{abs}} = \ln(I/I_0)/x \]
\[ x = \text{length of light path} \]
\[ I = \text{intensity of light after distance} \]
\[ I_0 = \text{initial intensity of light} \]
\[ b_{\text{scat}} \text{ and } b_{\text{abs}} \text{ are the scattering and absorption coefficients} \]

4 Case Studies

- **T1 and T3**
  - Meteorological disturbance brought air of African origin

- **T2 and T4**
  - Precipitation scavenging removed particles
Transition in Particle Sizes

Curve 1 (3/12/83):
- Total aerosol 5460 cm⁻³
- Radon 25pCi m⁻³
- Overcast sky w/stratocumulus

Curve 2:
- Total aerosol 1920 cm⁻³
- Radon 7pCi m⁻³
- Clear sky w/10% cumulus

Curve 3:
- Total aerosol 1325 cm⁻³
- Radon 7-8pCi m⁻³
- Overcast 80-100% w/80-100% altocumulus & stratocumulus
- Hump associated w/nonpft cloud effect on particle size distribution (cause for double-peaked distributions?)

Curve 4 (3/18/83):
- Total aerosol 243 cm⁻³
- Radon 2pCi m⁻³
- Overcast 85%
Double-Peaked Characteristic

- 3/18 and 3/19 size distributions exhibited double-peak char.
- Distributions did change but the double-peaked characteristic always recovered
- In the remote (tropical) marine Atlantic the size distribution is sustained by an unknown mechanism
- **Idea was**: Separation of interstitial and CCN from aerosol cycling thru nonppt clouds

Fig. 10. Composite of 49 size distributions taken during a 35-hour period on March 18 and 19, typical of conditions when there was no continental influence.
Case T1
PPT Changes Size Distribution

- Supports idea that aerosol cycling thru nonppt clouds causes the double-peak characteristic.
- Air mass was of Saharan origin, which resulted in a transition in particle size distribution.
- Even though extensive cloud cover, this was an exception to the double peak observation assoc w/nonppt cloud presence.

Fig. 11. Change in the size distribution during transit through weak front on March 16. Curves 1 and 2 are the average size distributions taken before and after passage of the front.
Case T2

- Ship passed thru high thunderstorm activity
- Heavy ppt around and north of the ship
- After some rain the size distribution of curve 2 was observed
- Interpret low concentration in 2nd size distribution to be result of ppt scavenging in local t-storms
Case T3

- Air trajectory indicated the air had been over Africa 7 days earlier.
- Cu & Cb w/showers in vicinity of ship and change in size distribution of submicron particles.
- Inc in wind speed, no sea spray and continentally derived elements in lg particles indicated increase in [lg particles] due to air mass change and not locally generated sea-salt particles.
- 2nd exception to double peak observation in presence of nonppt clouds.
Case T4

- Curve 1 & 2: air came from N. Atlantic passing near Iberian Peninsula 4 days prior to arriving at ship.
- 2 days after curve 1 and 2, significant change in submicron size distribution (Curve 3).
- Local conditions were similar but back trajectory of the air for the 3 periods was very different.

Back trajectory was behind cold front in region of widespread ppt.

Trajectories were always in front of the cold front and air didn’t encounter a lot of ppt.

Fig. 15. Curves 1 and 2 are size distributions taken in air which had passed in front of a cold front and had encountered no (recent) precipitation, whereas the size distribution represented by curve 3 was taken in air which had passed through the cold front and encountered heavy precipitation 3 days prior to the measurement.
Approach to Canary Islands

- Downwind of Grand Canary, the double peaked feature disappeared.
- Downwind of Tenerife air was more polluted than in remote tropical Atlantic and peak occurred at larger radius.

Fig. 19. Curve 1 is the size distribution before the ship was downwind of the islands. Curve 2 was taken downwind of Tenerife during heavy overcast conditions. Curve 3 was taken downwind of Grand Canary Island under clear conditions. The locations at which the three size distributions were taken are shown by the numbers along the ship’s track in Figure 17.
Air Adveecting off Iberian Peninsula

- Midday at 1 and 2, increase in small particles
- Small particle increase associated with homogeneous nucleation and growth of new particles from gas phase reaction products
- Photochemically induced since occur in air which spent the morning daylight hours over water prior to arriving at the ship

Fig. 20. Back trajectories for air encountered off the Iberian Peninsula where the tie marks indicate 6-hour intervals. Radon concentration (squares), CN concentrations (circles), and integrated aerosol concentrations (solid curve) measured along the track of the ship are plotted. (Scales are linear.) Time periods indicated by 1 and 2 refer to periods when the size distributions in Figures 22 and 23 were taken.
Gas-phase rxn products of marine origin provide source for small particles in MBL

DMS Rxn w/OH:
\[
\text{OH} + \text{CH}_3\text{SCH}_3 \rightarrow \text{CH}_3\text{SCH}_2 + \text{H}_2\text{O}
\]
Proposed life cycle of small marine particles in remote tropics

10-20 nonppt cloud cycles over 3-10 day period

Rate of 10-100 cm$^{-3}$ d$^{-1}$,
Growing to ~0.04 µm
Nonppt Cloud Hypothesis

- **Supporting evidence:**
  - Particles under both peaks are too volatile to be sea-salt
  - Double-peaks associated with air passing thru regions of boundary layer clouds
  - Minimum occurs at 0.04-0.07 $\mu$m radius range, agrees w/cloud super-saturation
  - Particle number under 0.09 $\mu$m peak is in 50-130 cm$^{-3}$ range, correlates with [cloud droplet] in clouds
  - Convertible gas source from sea surface able to sustain size distribution
Proposed Mechanism Time Scale

✓ Assumes time for dry aerosol to increase radius by 50% is constant

✓ Assumes cloud droplets have 7 µm & [200 cm⁻³]

✓ Assumes aerosol spends 5% time in cloud

✓ Assumes the size distribution observed in remote tropical Atlantic
Conclusions

A. [Small particle] decays as air masses move off continents

B. Separation of interstitial and CCN from aerosol cycling thru nonppt clouds

C. “Background aerosol” in remote tropical marine boundary layer (MBL) maintained by in-situ formation of new particulate matter from gas phase

D. Gas-phase rxn products of marine origin provide source for small particles in MBL