Observational constraints on aerosol indirect effects and controlling processes

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Motivating questions

- What are spaceborne aerosol measurements telling us about CCN?
- What are CCN over the remote oceans telling us about (anthropogenic aerosol sources)?



Rosenfeld et al., Science, 2008

Wood et al J. Geophys. Res., 2012

Regional gradients: Strong aerosol indirect effects in an extremely clean background





Does satellite AOD inform about CCN?

VOCALS as a testbed for understanding aerosol variability



• Break down aerosol optical depth τ into constituent parts

$$\frac{d\tau}{\tau} = \frac{d\sigma}{\sigma} + \frac{dG_{\sigma}}{G_{\sigma}} + \frac{dh}{h} = \frac{dN_{a}}{N_{a}} + 3\frac{dD_{3}}{D_{3}} + \frac{dG_{\sigma}}{G_{\sigma}} + \frac{dh}{h}$$

dry extinction

aerosol layer depth

$$\frac{d\tau}{\tau} = \frac{d\sigma}{\sigma} + \frac{dG_{\sigma}}{G_{\sigma}} + \frac{dh}{h} = \frac{dN_a}{N_a} + 3\frac{dD_3}{D_3} + \frac{dG_{\sigma}}{G_{\sigma}} + \frac{dh}{h}$$

Three longitude bins: $80-85^{\circ}W$, $75-80^{\circ}W$, $70-75^{\circ}W$ dX is the increase from offshore to coastal bin; X is the mean value





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Three longitude bins: 80-85°W, 75-80°W, 70-75°W *dX* is the increase from offshore to coastal bin; *X* is the mean value

dD3 -0.38 $[D_3 decreases from 0.28 to 0.25 \mu m]$ dh -0.25 [MBL depth decreases from h 1.5 to 1.2 km] (a) Cloud boundaries and LCL 1.8 Cloud top 1.6 Cloud top (C-130, REx) (RHB) 1.4 1.2 Cloud altitude (km) base (RHB) 0.8 Bretherton et al. (2010); Surface Near surface Cloud base (C-130, REx) LCL (RHB) LCL (C-130, REx) 0.6 de Szoeke et al. (2012) 0.4 -85 -82.5 -77.5 -75 -72.5 -80



Closure achieved for dry aerosol scattering

$$\frac{d\tau}{\tau} = \frac{dN_a}{N_a} + 3\frac{dD_3}{D_3} + \frac{dG_\sigma}{G_\sigma} + \frac{dh}{h}$$

$$\downarrow \qquad \qquad \downarrow \qquad \qquad \downarrow$$

$$\frac{d\tau}{\tau} = 0.09 \qquad \qquad \frac{dN_a}{N_a} = 0.72$$

$$3\frac{dD_3}{D_3} = -0.38$$

$$\frac{dh}{h} = -0.25$$

$$\frac{dG_\sigma}{G_\sigma} = 0.0-0.24^*$$

$$= 0.09-0.17 \qquad \qquad 0.09-0.33 \qquad ^{\text{highly uncertain}}$$

 \Rightarrow Large increase in conc. offset by reduction in size, and MBL depth

Changes in size are small, but important 0.25 μm



"The droplet mode size is nearly invariant" Kleinman et al. (ACP, 2011)

What controls CCN and cloud microphysical variability in the marine boundary layer? A simple CCN budget for the PBL

ENTRAINMENT NUCLEATION/SECONDARY DRY DEP. $\dot{N} = \dot{N}_{FT} + \dot{N}_S + \dot{N}_{PROD} + \dot{N}_P + \dot{N}_{DRY} + \dot{N}_{ADV}$ SURF. SOURCE PRECIP. SINK ADVECTION

- Assume nucleation/secondary processes unimportant
- Dry deposition is negligible (Georgi 1990)
- Sea-spray formulation (e.g. Clarke et al. 2006)
- Ignore advection
- Precipitation sink primarily from accretion process
- Equivalency of CCN and cloud drop conc. N_d

What controls CCN and cloud microphysical variability in the marine boundary layer? A simple CCN budget for the PBL

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Steady-state CCN budget



- Concentration relaxes to FT concentration N_{FT} + wind speed dependent surface contribution dependent upon subsidence rate (Dz_i)
- Precipitation sink controlled by precipitation rate at cloud base P_{CB} . Use expression from Wood (2006).

Precipitation important in controlling gradient in cloud droplet concentration

- Assume constant FT aerosol concentration
- Precipitation from CloudSat estimates from Lebsock and L'Ecuyer (2011)
- Observed surface winds
- Model N_d gradients mostly driven by precipitation sinks



Wood et al. (J. Geophys. Res. 2012)

Precipitation is primary control of N_d away from coastal zones

Model reproduces significant amount of variance in N_d over oceans \Rightarrow implications for interpretation of AOD vs r_e relationships



Thoughts

- Much more work is required to interpret remotelysensed AOD measurements as providing useful information about CCN
 - VOCALS region shows a doubling of Nd from the remote ocean to the coast but only a 10% increase in AOD
 - Differences explained by decreasing MBL depth and aerosol size
 - Need assimilation approaches, e.g. Saide et al. (2012)
- A large fraction of the variability in cloud droplet concentration over the remote oceans is driven by precipitation sinks as opposed to aerosol sources
 - Confounds interpretation of cloud vs aerosol relationships as indicative of aerosol indirect effects caused by anthropogenic pollution sources

Separating aerosol impacts from meteorological impacts on clouds

- Aerosol impacts on clouds are not simply explained by Twomey's arguments
- Changes in macrophysical cloud properties produce radiative impacts of same order as those from Twomey (e.g. Lohmann and Feichter 2005, Isaksen et al. 2009)

Aerosol impacts on cloud

• An observed change in cloud property *C* is caused by changes due to meteorology *M* and aerosols *A*:

$$\delta C = \left(\frac{\partial C}{\partial M}\right)_A \delta M + \left(\frac{\partial C}{\partial A}\right)_M \delta A$$

meteorology-driven

aerosol-driven

- To determine aerosol-driven changes on *C*, one needs to measure meteorology-driven changes
- This is a particularly arduous task, as the following examples demonstrate

Stevens and Brenguier (2009)

Shiptracks



$$\delta C = \left(\frac{\partial C}{\partial M}\right)_A \delta M + \left(\frac{\partial C}{\partial A}\right)_M \delta A$$
$$= 0$$

Shipping lanes

- Shipping emissions increase along preferred lanes
- **Control** clouds upstream; perturbed clouds downstream

$$\delta f = \left(\frac{\partial f}{\partial LTS}\right)_A \delta LTS + \left(\frac{\partial f}{\partial A}\right)_M \delta A$$



20S

40S

<->

120W

10^{1.3}

 $10^{0.7}$

10^{1.9}

clean A cloud cover increase of 0.02 represents a radiative forcing of 2 W m⁻²

Peters et al. (ACP, 2011)

90W

 10^{3}

 $10^{2.4}$

[metric tons yr⁻¹, (1°x1°)⁻¹]

60W

10^{3.6}

(Mostly) regulating feedbacks in stratocumulus

