Aerosol indirect effects and the importance of the CCN budget for marine low clouds

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MBL clouds likely responsible for much of the global aerosol indirect forcing

LEFT: Annual mean aerosol first indirect forcing; RIGHT: uncertainty in simulated first indirect forcing due to uncertainty in model (GLOMAP) parameters. Note the large aerosol indirect forcing and uncertainty in the ENA. Adapted from Carslaw et al. (2013)
Aerosol indirect effects

- **Twomey** ($N_a - N_d - \text{albedo}$) [aerosol concentration – cloud droplet concentration – cloud optical thickness]
  - Five decades of evidence linking cloud droplet concentration with aerosol concentration.
  - Twomey effect: strong sensitivity of albedo to unperturbed aerosol state.

- **Light precipitation sensitivity to $N_d$**
  - Drizzle common in the cloud-capped PBL, esp. over ocean
  - Consistent indication that Sc clouds with same amount of condensate precipitate less readily when they have larger $N_d$
  - Unclear from field data about susceptibility of precip. in Cu

- **Importance of cloud dynamical responses to aerosol**
  - Are polluted clouds more vigorous?
  - Cloud top entrainment
FIRST INDIRECT EFFECT

lower albedo
macrophysically identical clouds
fewer larger drops

higher albedo
more smaller drops

SECOND INDIRECT EFFECT

lower albedo
macrophysically different clouds
more efficient precipitation
⇒ more LWC depletion
⇒ less cloud cover/longevity

higher albedo
less efficient precipitation
⇒ less LWC depletion
⇒ more cloud cover/longevity
Key physical processes: Stratocumulus

- Large scale subsidence
- Evaporative cooling
- Latent heating
- Turbulent mixing
- Solar heating
- Longwave cooling
- Drizzle
- Surface fluxes energy & moisture
- Entrainment

Sea surface

FREE TROPOSPHERE

BOUNDARY LAYER
\( N_a - N_d \) relationship

- Formalized as the ACI metric (e.g. Feingold et al.):
  \[
  ACI = \frac{\partial \ln N_d}{\partial \ln N_a}
  \]
  - where \( N_a \) usually refers to as the concentration of accumulation mode aerosol particles.
- More complex relationship known as *cloud droplet concentration closure* (whereby entire CCN spectrum or size distribution/composition is specified, along with updraft speed statistics)
- Consequences for cloud albedo first proposed by Twomey (1974, 1977)
• First $N_d$ closure experiments by Twomey and Warner (right)
• Revisited in the 1990s (Martin et al. 1994, below)
  – Linear relationships for low $N_a$
  – "saturation" effect at larger $N_a$

Fig. 1. Comparison of mean droplet concentration observed in cloud with the concentration computed from the observed spectra of cloud nuclei for an updraft of 3 m sec$^{-1}$. The dashed line represents exact agreement between observed and computed values.


Cloud droplet concentration closure

- Excellent closure in small Cu and Sc
- Activation largely a solved problem (although not necessarily in application to climate models)


Figure 2. Aerosol-CDNC closure: predicted versus observed droplet concentration. Observed values use method 1 screening (see text) for adiabaticity. The short-dashed line represents an unweighted least squares linear fit to the data in log-log units. The long-dashed line represents a fit to the data when method 2 screening is used. The solid line represents perfect model-observation agreement. The term “cloud base” reiterates that observations used in this plot were generally taken in adiabatic regions within 100 m of cloud base.
ACI \tau

- More recently (2000s) attention has turned to evaluation of Twomey effect on cloud radiative properties, e.g. using ACI metrics:

\[ ACI \tau = \left( \frac{\partial \ln \tau}{\partial \ln N_a} \right)_{LWP} \]

where \( N_a \) usually refers to as the concentration of accumulation mode aerosol particles.

- Discrepancies between aircraft (e.g. Painemal and Zuidema values close to Twomey expectations) and remote sensing estimates that are often lower (see McComiskey et al. 2012) – scale issues?

Twomey albedo susceptibility

Combine Twomey’s expression for cloud optical thickness:

\[ \tau = kN^{1/3}L^{2/3}h \]

with simple expression for albedo, \( \alpha \approx \tau/(	au + 7) \), to estimate the rate of change of albedo with \( N \) (termed the albedo susceptibility, \( \text{Platnick and Twomey 1994} \)), to obtain:

\[ \frac{d\alpha}{dN} = \frac{\alpha(1 - \alpha)}{3N} \]

This form, although not in the original Twomey (1974,77) papers, is the most instructive way to visualize the key results:

- **Clouds with low \( N \) are most susceptible to an increase in \( N \)**
- Clouds with albedos \( \sim 0.5 \) are more susceptible to increases in \( N \) than clouds with either lower or higher albedo.

Minimum allowed cloud droplet concentration influences indirect forcing in models

\[ \Delta A \propto \ln\left( \frac{N_{\text{perturbed}}}{N_{\text{unperturbed}}} \right) \]

Low \( N_d \) background \( \Rightarrow \) strong Twomey effect
High \( N_d \) background \( \Rightarrow \) weaker Twomey effect

Quaas et al., AEROCOM (Atmos. Chem. Phys., 2009)  
Hoose et al. (GRL, 2009)
Precipitation “susceptibility” metric

Analogous to albedo susceptibility, precipitation susceptibility expresses sensitivity of precipitation rate to cloud droplet concentration:

A Proposed New Construct for Warm Clouds

Current constructs of aerosol effects on clouds are useful provided that they can be related to observations that prove causality. We propose here a simple construct that may assist in addressing aerosol effects on precipitation. Consider an analog to the albedo susceptibility $S'_0$, which we will term “precipitation susceptibility,” $R'_0$ defined as:

$$R'_0 = - \frac{d \ln R}{d \ln N_d}. \quad (14.4)$$

$R'_0$ expresses the relative change in precipitation $R$ for a relative increase in $N_d$ in warm clouds. The minus sign is applied so that a positive $R'_0$ will reflect the conventional wisdom that for warm rain, $R$ and $N_d$ are negatively correlated.

Precipitation susceptibility in marine low clouds from ground based remote sensing, aircraft and LES


**Figure 4.** Precipitation susceptibility with respect to CCN number concentration as a function of liquid water path in AMF observations from this study and to cloud droplet number concentration in VOCALS observations averaged over a 5 km length scale (squares) [Terai et al., 2012, Figure 7] and large-eddy simulations (LES, triangles; adapted from Sorooshian et al. [2009]) of precipitating cumulus initialized using soundings from the Rain in Cumulus over the Ocean (RICO) field campaign [Rauber et al., 2007]. Error bars in AMF data represent 95% confidence intervals, which take into account the errors associated with $R_{cb}$ in Figure 2.
Cloud macrophysical responses: ship tracks are useful control experiments for aerosol influences on clouds

- LWC reductions in some ship tracks (see also Coakley and Walsh 2002)

- Effect likely related to increased cloud top entrainment caused by (a) TKE invigoration due to drizzle suppression; (b) reduced cloud drop sedimentation (Ackerman et al. 2004, Bretherton et al. 2007)

Chen et al. (Atmos. Chem. Phys., 2012)
The need to quantify sources and sinks in the MBL CCN budget

- Aerosol indirect effect magnitude sensitive to unperturbed (preindustrial) aerosol loading
  - Can be understood theoretically from combination of Twomey effect and sublinear $N_d-N_a$ relationship
- Unperturbed aerosol loading dependent upon balance of natural sources and sinks of CCN
- Perturbed aerosol loading dependent upon flux (transport) of pollution aerosol and aerosol sinks
- New natural surface aerosol sources have been identified
- Processes controlling CCN budget (e.g. precipitation, entrainment) are also critical mediators of aerosol indirect effects
What factors control the magnitude and uncertainty of the global AIE?

- Accumulation mode width
- Cloud thickness
- Low cloud fraction
- Updraft velocity
- Cloud water replenishment time
- Threshold radius for autoconversion
- Anthro BC+POM emissions
- Anthro SO2 emissions
- Anthro SOA emissions
- Primary mode radius
- Mass fraction new particles
- Mass fraction of secondary aerosol mass on accumulation mode

…..also Carslaw et al. (Nature, 2013)  

Ghan et al. (J.Geophys. Res., 2013)
Simple CCN budget in the MBL

\[ \dot{N} = [\dot{N}]_{\text{ent}} + [\dot{N}]_{\text{sfc}} + [\dot{N}]_{\text{coal}} + [\dot{N}]_{\text{dry dep}} \]

Model accounts for:

- Entrainment
- Surface production (sea-spray only)
- Coalescence scavenging
- Dry deposition

Model does not account for:

- New particle formation – significance still too uncertain to include
- Advection
Conceptual model of background FT aerosol

Clarke et al. (J. Geophys. Res. 1998)
Production terms in CCN budget

\[
\left[ \dot{N} \right]_{ent} = \frac{w_e (N_{FT} - N)}{z_i}
\]

\[
\left[ \dot{N} \right]_{sfc} = \frac{\beta U_{10}^{3.41}}{z_i}
\]

We use Clarke et al. (J. Geophys. Res., 2007) at 0.4% supersaturation to represent an upper limit.
Loss terms in CCN budget: (1) Coalescence scavenging

\[
\dot{N}_{\text{coal}} = -KNP_{CB} \frac{h}{z_i}
\]

Comparison against results from stochastic collection equation (SCE) applied to observed size distribution

\[
\dot{N} \approx -\pi E_0 N \int_0^\infty r^3 w_T n(r) dr = -\frac{3}{4\rho_w} E_0 NP
\]

Loss terms in CCN budget: (2) Dry deposition

\[
\left[ \frac{\dot{N}}{N} \right]_{\text{dry dep.}} = -N \frac{W_{\text{dep}}}{Z_i}
\]

\[
\frac{\left[ \frac{\dot{N}}{N} \right]_{\text{coal}}}{\left[ \frac{\dot{N}}{N} \right]_{\text{dry dep.}}} = \frac{KP_{CB}h}{w_{\text{dep}}}
\]

\[w_{\text{dep}} = 0.002 \text{ to } 0.03 \text{ cm s}^{-1} \text{ (Georgi 1988)}\]

\[K = 2.25 \text{ m}^2 \text{ kg}^{-1} \text{ (Wood 2006)}\]

For \( P_{CB} = > 0.1 \text{ mm day}^{-1} \) and \( h = 300 \text{ m} \)

\[
\frac{\left[ \frac{\dot{N}}{N} \right]_{\text{coal}}}{\left[ \frac{\dot{N}}{N} \right]_{\text{dry dep.}}} = 3 \text{ to } 30
\]

For precip rates \( > 0.1 \text{ mm day}^{-1} \), coalescence scavenging dominates
Steady state (equilibrium) CCN concentration

\[ N_{eq} = \left( \frac{N_{FT} + \frac{\beta U_{10}^{3.41}}{Dz_i}}{1 + \frac{hkP_{CB}}{Dz_i}} \right) \]

\[ \frac{w_e}{z_i} = D = \text{surface divergence} \]
Drizzle helps control CCN and cloud microphysical variability in the MBL.

- **Simple** budget model for CCN/$N_d$ in the MBL:

\[
\dot{N} = [\dot{N}]_{ent} + [\dot{N}]_{sfc} + [\dot{N}]_{coal}
\]

\[
N_{eq} = \left(\frac{N_{FT} + \frac{\beta U^{3.41}}{DZ_i}}{1 + \frac{hkP_{CB}}{DZ_i}}\right)
\]

- Assume constant FT aerosol conc.
- Model $N_d$ gradients mostly driven by precipitation sinks

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Cloud effects on aerosols

Observations of the Geographical Variation of Cloud Nuclei

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(Manuscript received 16 December 1968)

ABSTRACT

Airborne measurements at flight level of cloud nuclei have been carried out during approximately 100,000 mi of flight over wide areas of the world. A thermal diffusion chamber was used to obtain cloud-nuclei spectra which present the concentration of active nuclei as a function of supersaturation over the range of supersaturation of interest in cloud formation. The results confirm previous conclusions that continental air masses are systematically richer in cloud nuclei and indicate that the median of cloud nuclei over the open ocean is reasonably predictable and varied little from region to region. Over continents the median spectrum does not seem to vary greatly even when North America was compared to Africa and Australia; however, the variability about the median is much greater over land when compared to ocean areas. The results indicate that the lifetime of cloud nuclei, at least over the oceans, is about 3 days. The results require a widespread and relatively uniform source of cloud nuclei both over the oceans and over the land, and it is indicated that the sources of nuclei are not largely dependent on industrial or other man-made pollution.

These semi-quantitative arguments suggest that washout is sufficient to account for the observed depletion rate in the case of the nuclei which grow into cloud droplets and become involved in coalescence and rain formation. The removal of the inactivated nuclei, which

Fig. 4. The decay of nucleus number (for 0.75%, supersaturation) in continental air moving out over the ocean; mid-latitude observations (a); tropical or subtropical observations (b).
Ultra-Clean Layers – a common occurrence?

- Aircraft observations from VOCALS identified Ultra-Clean Layers (UCLs) at the level of the detraining cloud layer in ALL open cell cases (POCs) sampled; common during recent CSET campaign over NE Pacific.
- In UCLs, CCN concs. are two orders of magnitude lower than those near the surface.
- Impacts of such low CCN on clouds and susceptibility to aerosol perturbations unclear.
Sources of remote MBL CCN

Free troposphere:
- Long range transport of continental emissions
- Homogeneous nucleation of DMS oxidation products


Entrainment rate

Sea salt

Organics

Cloud Condensation Nuclei

Marine boundary layer:
- Particle growth – condensation, coagulation, cloud processing
- Secondary organic aerosol formation

Biological activity
Wind speed
DMS
Phytoplankton
DOM
Sea salt
Marine aggregates

H$_2$SO$_4$ → $\text{SO}_4^{2-}$

$\text{SO}_2$ → DMS

Gas phase organics

Wind speed
Bubble bursting

Biological activity
Surfactants
Wind speed
Bubble bursting
Summary

• The magnitude of the global aerosol indirect forcing is critically dependent upon:
  – Marine boundary layer cloud macrophysical responses (turbulence, PBL structure, LWP, entrainment, cloud cover) to aerosol perturbations
  – Unperturbed (preindustrial) CCN concentration and particle size

• Processes controlling MBL CCN budget are also critical for determining magnitude of aerosol indirect forcing
Liquid clouds immediately above and below a biomass burning aerosol layer over the SE Atlantic, W. of Angola [MODIS Terra, RGB]
Light precipitation drives cloud transitions

DYCOMS-2 Field Campaign
Stevens et al. (2005, BAMS)
Polluted shallow cumulus may have shorter lifetimes

Lessons

• $N_a$-$N_d$ relationships mostly an engineering rather than a science challenge
  – Scale effects important; updrafts important
• Observations show good evidence for aerosol-driven precipitation suppression, but also for precipitation-driven aerosol suppression
  – Together with “contamination” of the CCN signal by near-cloud radiative/swelling effects, results in major challenges in interpreting satellite aerosol-cloud relationships
• Cloud macrophysical responses to changing CCN can be observed but obtaining adequate controls is challenging
  – Ship track clouds can increase or decrease condensate in response to aerosol increases.
High resolution measurements of the EIL atop stratocumulus

Large eddy scale $<<$ EIL thickness

"nibbling rather than engulfment"

Katzwinkel et al. (BLM, 2011)
Light precipitation sensitivity to $N_d$

First quantitative measurements in Sc. clouds (Brost et al. 1982)

Precipitation increases strongly with cloud top height in warm precipitating trade cumulus (Byers and Hall 1955)
Entrainment in trade cumulus: new considerations

- Upward mass transport in cumulus clouds primarily balanced by near-cloud downdrafts
- Implications for species longevity in the trade Cu MBL, and for lateral entrainment

Jonker et al. (2008)

Heus et al. (2009)
New capabilities open new frontiers

• ARM Climate Research Facilities, especially new remote sensing and aerosol sampling suites, offer long-duration sampling of cloud physical and dynamical processes. New statistical approaches to cloud sampling. New permanent ARM site in the Azores.

• Need for more routine measurements over the remote ocean (e.g. NSF Ocean Observatories Initiative “super buoys”)

• Aircraft facilities now offer longer range (e.g. Global Hawk, HIAPER), unprecedented remote sensing

• **Big capabilities ⇒ need big thinking**
Where new field observations can help

• Factors controlling cloud top entrainment in stratocumulus and cumulus.
  – Role of shear and evaporative cooling uncertain.
  – Need to exploit new measurements approaches, both in-situ (very high resolution measurements) and remote sensing (new Doppler radar and lidar approaches)

• Microphysical impacts (both CCN and ice formation) on marine low clouds over the extratropical oceans
  – IN remain poorly understood and measured
  – Role of aerosol in influencing extratropical cloud regimes (e.g. open/closed cells, collapsed boundary layers)
  – Aerosol transport mechanisms to the remote ocean
  – What controls cloud droplet concentration over the extratropical oceans?
Stratocumulus cloud thickness

Data are from the published literature and available datasets from recent stratocumulus field campaigns (Nicholls and Leighton 1986; Boers and Krummel 1998; Miles et al. 2000; Pawlowska and Brenguier 2003; Comstock et al. 2004; vanZanten et al. 2005; Wood 2005a; Lu et al. 2007; McFarquhar et al. 2007; Lu et al. 2009; Wood and coauthors 2011)
What remains to be done?

• Understanding of entrainment and key processes affecting it remains poor
  – New measurement techniques required
  – Large eddy models do not yet explicitly model entrainment

• Large scale model treatments of low clouds are improving but low cloud feedback not narrowing significantly. Why?

• Aerosol impacts on Sc and (perhaps as importantly) Sc impacts on aerosols are not well known
Shallow, well-mixed STBL

Deep, cumulus-coupled STBL
Differences in precipitation "efficiency" between continental and marine clouds

A STUDY OF CONVECTIVE PRECIPITATION BASED ON CLOUD AND RADAR OBSERVATIONS

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(Manuscript received 6 March 1956)

ABSTRACT

Observations of precipitation and cloud-top height in the central United States and in the Caribbean area, obtained from radar-equipped airplanes, have been analyzed in terms of the fraction of clouds of a given height which contain precipitation. These data are compared with observations of a similar type taken by Braham, Reynolds and Harrell in New Mexico. It is concluded that the condensation-coalescence process can account for the formation of precipitation in convective clouds in all three regions but in varying proportions of clouds.

Fig. 6. Percentage of cumuli with precipitation as function of cloud-top height and temperature for three different geographical regions (lower part of diagram). Schematic clouds in upper part of diagram fall in height categories where 20 per cent were observed to contain precipitation echoes.
Canonical profiles

Albrecht et al. (J. Geophys. Res., 1995)
Entrainment interfacial layer
Recent MBL Cloud focused field programs