Mixed dust-black carbon forcing: How large is it?

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The global mean radiative forcing of the climate system for the year 2000, relative to 1750

Radiative Forcing (Watts per square metre)

- Halocarbons
- N₂O
- CH₄
- CO₂
- Tropospheric ozone
- Stratospheric ozone
- Sulphate
- Organic carbon from fossil fuel burning
- Biomass burning
- Mineral Dust
- Black carbon from fossil fuel burning
- Aviation-induced contrails
- Cirrus
- Aerosol indirect effect
- Land-use (albedo) only

Level of Scientific Understanding

- High
- Med.
- Med.
- Low
- Very Low
- Very Low
- Very Low
- Very Low
- Very Low
- Very Low
- Very Low
- Very Low
Climate applications

Hemispherical flux

\[ F_{\text{up}}(\lambda, xy, z, t) \] and
\[ F_{\text{down}}(\lambda, xy, z, t) \]

Remote sensing applications

Radiance

\[ I(\lambda, xy, z, \theta, t) \]

Radiation transfer codes

- optical depth \( \tau(\lambda, xy, z, t) \)
- single scattering albedo \( \omega_o(\lambda, xy, z, t) \)
  \( \omega_o = \frac{K_{sc}}{K_{sc} + K_{ab}} \)
- asymmetry parameter \( g(\lambda, xy, z, t) \)

Optical Model

Mie theory (or others)

- particle number (or mass) size distribution
  \[ N(r) = f(xy, z, t) \]
- refractive index (or composition)
  \[ m = n - ik; \quad m = f(\lambda, r, xy, z, t) \]
- particle shape

Sokolik et al. (2001)
Complex particle morphology

flyash

NaCl

CaCO₃

Ca silicate

soot
Particle morphology and composition

Wind-blown mineral dust:
- nonspherical particle
- mixtures of minerals controlled by the dust source and evolution during transport in the atmosphere

Fly ash (heavy industries/coal combustion):
- spheres
- metal oxides (SiO2; Al2O3)
- CaO (cement production)

Black carbon:
- Emitted total carbon is partitioned between OC and BC depending on material and combustion conditions
- From oil combustion: porous spheres
- From biomass burning and diesel combustion: fractal-like aggregated spheres
Black carbon

- All assume spherical shape

No comprehensive studies addressed fractal-like structure of BC in radiative forcing assessments

- Limited data on spectral refractive indices
Why we need to account for dust mineralogical composition (Sokolik et al., JGR, 1998; Sokolik and Toon, JGR, 1999)

- Dust is a collective term referring to widely varying mixtures of minerals.
- Minerals each have different physical and chemical properties (e.g., density, hygroscopicity, chemical reactivity, etc.)
- Spectral refractive indices vary widely from mineral to mineral.
- Optical properties of dust are determined by the relative abundance of each mineral and the details of how the minerals are mixed together.
- The abundance of various constituents depends on the dust source region, how it was mobilized, and chemical and physical transformation processes during dust transport in the atmosphere.

Selected minerals:
- Clays (illite, kaolinite, montmorillonite)
- Quartz
- Hematite (or other iron oxides)
- Calcite
- Gypsum

Claquin et al. (1998):
First global map of soil mineralogical composition

Miller et al. (2004):
GISS AGCM treats minerals as individual tracers
Asian dust: Extinction coefficient (a) and single scattering albedo (b) calculated for six models of dust size distributions: (1) Clarke et al. (2004), (2) D’Almeida (1987), (3) D’Almeida (1991), (4) OPAC (1998), (5) D’Almeida (long-range), (6) Bahrain-Aeronet.

Lafon et al. (2004): new technique for measurements of iron oxides
What is new in non-spherical dust modeling

Micshchenko et al. => mixtures of oblate/prolate spheroids

T-matrix


DDA (Discrete Dipole Approximation)

Kalashnikova et al. (2004, in press): incorporated Asian dust and Saharan dust analogs into MISR retrievals

Advanced approach: analysis of a statistically large number of dust samples => supervised classification of particle shapes and aggregation => probability distribution of shape/aggregation important for optics at the solar wavelength
Time series of aerosol single scattering albedo (Nephelometer/PSAP) and chemical composition of particles with radius less than 1 µm measured at Amami-Oshima site in April 2001 (Nakajima et al., 2003)
Dust in the marine boundary layer
ATOFMS data (Guazzotti et al.)
Processes resulting in the formation of multicomponent aerosols containing dust

- Adsorption of water vapor on dust particle surfaces
- Cloud processing
- Coagulation of dust with other aerosol or cloud particles
- Heterogeneous chemistry: uptake of $\text{SO}_2$, $\text{NO}_2$, $(\text{H}_2\text{SO}_4$ or $\text{HNO}_3)$ gases on dust particle surfaces followed by heterogeneous chemical reactions resulting in the formation of sulfates or nitrates.
Negligible effect of RH on coated Asian dust: implications to dust CCN

Humidograms for (a) $D_p < 10 \mu m$ and (b) $D_p < 1 \mu m$ on approximately DOY 101.9 (11 April) during the peak of the dust event sampled on the R/V Ronald Brown during ACE-Asia (Carrico et al., 2003)
Aggregation of dust with black carbon

Dust in marine BL: ATOFMS data, ACE-Asia, YD 101 (Prather, Guazzotti et al)

![Bar chart showing the percentage of dust particles aggregated with black carbon across different size classes.](chart.png)
Aggregation of dust with black carbon

ACE-Asia, C-130 data (Anderson)

\[ n_{bc} = (1.74, 0.44) \]

Negligible differences in optics!
Radiative forcing efficiency

Diurnal Forcing Efficiency at Kosan: April 12–19, 2001

\[ F_{TSBR} = 2.7 - 86.3 \tau_{440} \text{ [W m}^{-2}] \]

Bush, Leitner and Valero
Scripps Institution of Oceanography
University of California, San Diego

440 nm AERONET AOD (Courtesy Brent Holben: brent@aeronet.gsfc.nasa.gov)
April 13

SeaWiFS

TOMS

Kosan, Korea:
\( \tau_{sp(0.5\mu m)} = 0.6 - 1 \)

GOCART (Chin et al.)

MATCH (Rasch & Collins)
Measured solar radiative forcing

Diurnal Forcing Efficiency at Kosan: April 12–19, 2001

\[ F_{\text{TSR}} = 2.7 - 0.63 \tau_{440} \quad [\text{W m}^{-2}] \]

Bush, Leitner and Valero
Scripps Institution of Oceanography
University of California, San Diego

440 nm AERONET AOD (Courtesy Brent Holben: brent@aeronet.gsfc.nasa.gov)
Problems with the concept of radiative forcing efficiency

Large differences in AOT depending on a measurement/modeling approach

Time series of AOT as a function of day in April 2001 at Gosan and Amami-Oshima sites. Results from CFORS (solid line), SPRINTARS (broken line), satellite (circles), and surface method (triangles) (Nakajima et al. 2003)
Problems with the concept of radiative forcing efficiency

The 24 hour mean clear sky net shortwave ARF as a function of AOT at 500 nm at Gosan and Amami-Oshima. Theoretical values with Junge aerosol size distribution and US standard atmosphere are also presented as a reference. The imaginary part of the aerosol refractive index is changed from 0 to -0.05 as labeled. (Nakajima et al., 2003)
Radiative fluxes are controlled by effective optical properties

Effective optical properties = aerosol + gases + clouds

\[ \tau_{\text{eff}} = \sum \tau_i \]
\[ \omega_{\text{eff}} = \frac{\sum \omega_i \tau_i}{\sum \tau_i} \]
\[ g_{\text{eff}} = \frac{\sum g_i \tau_{i,s}}{\sum \tau_{i,s}} \]

In the presence of both absorbing and non-absorbing aerosols:

\[ F(\tau_{\text{eff}}, \omega_{\text{eff}}, g_{\text{eff}}) \neq \sum F_i \]
Aerosol radiative impacts in the presence of clouds

TOA radiative forcing from remote sensing:

\[ \Delta F_{\text{TOA}} = -F_{\text{TOA}}^{\uparrow}(\text{aerosol + cloud}) - F_{\text{TOA}}^{\uparrow}(\text{clear}) \]

Radiative forcing from climate/radiation models:

\[ \Delta F_{\text{TOA}} = -F_{\text{TOA}}^{\uparrow}(\text{aerosol + cloud}) - F_{\text{TOA}}^{\uparrow}(\text{cloud}) \]
TOA radiative forcing of dust/pollution in clear and cloudy conditions

over the ocean
(low sun)

over the land
(high sun)

\[ \text{[aer+cloud]} - \text{[clear]} \quad \text{[aer+cloud]} - \text{[cloud]} \]

\[ \text{[aer]} - \text{[clear]} \]
ACE-Asia radiative forcing assessments

Nakajima et al. (2003)
ACE-Asia TOA radiative forcing assessments

Conant et al. (2003)

Large differences in spatial distribution of TOA forcing!!!

Takemura et al. (2003)
ACE-Asia radiative forcing assessments

April, 2001

Conant et al. (2003): TOA forcing = - 3 W/m²
Surface forcing = - 17 W/m²

Nakajima et al. (2003): Aerosol indirect = 1-3 W/m²
Takemura et al. (2003): Aerosol indirect = - 1.8 W/m²
(SPRINTARS)
Processes resulting in the formation of multicomponent aerosols containing dust

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The percentage of sampling days when dust dominated aerosol light extinction is as high as ~ 50 – 70% in some of the sites. “Dominated” means that it caused more light extinction than any of the other major categories -- SO4, NOx, EC, and OC.
Dust/BC mixtures

Time series of aerosol single scattering albedo (Nephelometer/PSAP) (squares) and absorption coefficients (bars) measured at Yasaka, Japan, between 20 March and 5 April 2001 (Holler et al., 2003)
Vertical profiles of regional optical properties (no STP correction) measured on the DC-8 and P3-B for the intensive portion of the TRACE-P experiment near the Asian continent, the case of urban pollution

(a) Vertical profile of altitude-averaged (0.25 km altitude bins) total scattering coefficients for DC-8/P3-B flights near the Asian continent below 6 km. (b) Same as Figure a, but for the altitude-averaged total absorption coefficients. (c) Same as Figure a, but for the altitude-averaged total single scatter albedo (Moore et al. 2003)
April 17

TOMS

MATCH (Rasch & Collins)

$\tau_{\text{sunphot}} = 0.5 - 0.7$

GOCART (Chin et al.)
Radiative forcing efficiency

(a) The solar aerosol forcing at the Earth's surface (broadband and diurnal averaged) as a function of aerosol optical thickness (AOT); (b) the solar aerosol forcing at the TOA (broadband and diurnal averaged). The open circles in both cases represent the aerosol forcing obtained from observations, squares represent the aerosol forcing obtained from radiative transfer model, and the solid line is a linear fit to the observations. (Markowicz et al. 2003)
SEM/TEM show very complex morphology

...but resolution scale is 10-20 nm