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Workshop on Aerosol Impact in the Environment: from Air Pollution to Climate Change

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Introduction to Aerosol/radiation/climate interactions - pt.2

M. Mallet Lab. d'Aerologie, Toulouse, France





Introduction to aerosol radiations & climate interactions

Marc Mallet

(CNRS - Laboratoire of Aerologie)



Outlines

1) Definition & physical-chemical-optical properties

1.1 Aerosol Physical & Chemical properties 1.2 Aerosol Optical properties

1.3 The aerosol mixing state

2) Aerosols & impact on climate

2.1 Aerosol radiative properties 2.2 The aerosol « direct » & « semi-direct » effects 2.3 Aerosol & the regional climate

Definition

« Aerosol is defined as a suspension of fine solid or liquid particles in a gas »

Aerosols could be emitted directly as particles (primary aerosol) or formed in the atmopshere by gas-to-particle conversion processes (secondary aerosol)

Primary aerosols :

The main aerosol types are :

- mineral dust particles,
- Primary black & organic carbon,
- sea salt particles,
- volcanoes,

- Secondary aerosols :
- The main species are :
- sulfates aerosols
- nitrates particles,
- « Secondary » organics,...

> Atmospheric aerosols are generally considered to be the particles that range from a few nanometers (nm) to tens of micrometers (μ m) in diameter

1.1 Aerosol physical and chemical properties



The parametrization generally used : the log-normal size distribution

Equation:

$$\frac{dN}{d\log r} = \sum_{i=1}^{n} \frac{N_i}{\sqrt{2\pi}\log\sigma_i} \exp\left[-\frac{1}{2}\left(\frac{\log r - \log r_{g,n(i)}}{\log\sigma_i}\right)^2\right]$$



With: $r_{g,n}$: Number Median Radius (NMR), maximum of the aerosol concentration

 σ_g : geometric standard deviation, quantifies the width of the distribution

 $\ensuremath{\mathsf{N}}$: total aerosol number concentration in the considered mode

These functions have been chosen as they match well with observed shapes of ambient aerosol distribution

Generally used to treat aerosol size in models and to calculate optical properties and associated climate impact

Aerosol Chemical Composition

Example of the contribution of different aerosol species to the total particle mass during summer 1999-2000 in Paris



- large contribution of sulfates & organics

- none negligible <u>contribution</u> of BC, dust & nitrates

Among the different species, carbonaceous particles are very important \rightarrow interactions with radiations

Cachier et al. , 2005

Carbonaceous aerosols : BC, OC and Cbrown

The carbonaceous fraction of ambient particulate matter consists of **black carbon** and a variety of **organic compounds** (organic carbon)



Black carbon aerosol is emmited directly into the atmosphere during combustion \rightarrow main absorbing particle



Organic carbon is either emitted directly by sources (primary OC) or can be formed in situ by condensation of low-volatility products (secondary OC) \rightarrow mainly scattering

Emerging research suggests that a variety of particulate OM can absorb radiation, particularly at the **shorter visible and UV wavelengths** (Adler et al., 2009; Barnard et al., 2008; Dinar et al., 2008; Hoffer et al., 2006; Kirchstetter et al., 2004; Rincon et al., 2009,...)

The large variability in the absorption capacity of OM is **related to the composition of the OM fraction**, <u>which can include HULIS</u>, lignin and polycyclic aromatic compounds,

OC is not necessary a purely scattering aerosol !

Brown Carbon Spheres in East Asian Outflow and Their Optical Properties

Duncan T. L. Alexander,¹ Peter A. Crozier,²* James R. Anderson³

Atmospheric aerosols play a substantial role in climate change through radiative forcing. Combustion-produced carbonaceous particles are the main light-absorbing aerosols; thus, quantifying their optical properties is essential for determining the magnitude of direct forcing. By using the electron energy-loss spectrum in the transmission electron microscope, we quantified the optical properties of individual, submicrometer amorphous carbon spheres that are ubiquitous in East Asian–Pacific outflow. The data indicate that these common spheres are brown, not black, with a mean refractive index of 1.67 - 0.27 (where $i = \sqrt{1}$) at a wavelength of 550 nanometers. The results suggest that brown carbon aerosols should be explicitly included in radiative forcing models.



Fig. 2. Wavelength dependent mass absorption cross-section (MAC) of C_{Brown} with a form as given by Sun et al. (2007) and where the absolute magnitude of the k_{Brown} (solid black line) has been deduced from Barnard et al. (2008). Dashed lines indicate k_{Brown} upper and lower bounds for our modeling.

Organic aerosols are generally considered as purely « scattering » in modelling simulations...

RegCM regional climate model

Table 1. Number distribution characteristics and optical properties (at 380 and 550 nm) for the four carboneaceous types considered in the study. $r_0 = 'dry'$ modal radius, $\sigma =$ standard deviation, $\rho_p =$ particle density, m = refractive index, $\sigma_{\text{ext}} = dry$ extinction cross section, α hygroscopic growth parameter, $f_{\text{rh}=0.8} =$ hygroscopic growth factor at 80% relative humidity

$\sigma_{\rm ext}$				$\sigma_{\rm ext}$ (n	$n^2 g^{-1}$)	g ⁻¹)		
Species	(µm)	σ	$(g \text{ cm}^{-3})$	380–550 nm	380 nm	550 nm	α	$f_{\rm rh} = 0.8$
BC _{hb}	0.0118	1.7	1.5	1.87-0.569i	14.6	9.6	0	0
BC_{hl}	0.03	1.9	1.5	1.87-0.569i	20.2	12.1	0.2	1.37
OC _{hb}	0.06	2.	1.7	1.55-0.005i	6.1	2.7	0	0
$\mathrm{OC}_{\mathrm{hl}}$	0.1	2.	1.7	1.55–0.005i	9.8	4.9.	0.25	1.49
					So	lmon et	al. Tell	lus, 2006

The regional atmosphere chemistry model REMOTE (Regional Model with Tracer Extension)

				Density,	
Author	Aerosol	r _{gN} , μm	σ_g	$kg(m^{-3})$	Refractive Indices at 0.55 μ n
Langmann et al. [1998]	sulfate	0.05	1.8	1600	1.43 -i 2.0*10 ⁻⁸
Hess et al. [1998]	sulfate	0.1	2.0	1760	1.53 -i 6.0*10 ⁻³
Koepke et al. [1994]	sulfate	0.07	2.03	1700	1.43 -i 1.0*10 ⁻⁸
Penner et al. [1998]	sulfate	0.05	2.0	1200	$1.53 - i \ 1.0^{*}10^{-7}$
Penner et al. [1998]	BC	0.0118	2.0	1800	$1.75 - i 4.4 * 10^{-1}$
Hess et al. [1998]	BC	0.01	2.0	1000	$1.75 - i \ 4.4 * 10^{-1}$
Cooke et al. [1999]	OC	0.02	2.0	1800	
This study	sulfate	0.05	1.8	1600	1.53 -i 1.0*10 ⁻⁷
This study	BC	0.0118	2.0	1800	$1.75 - i 4.4*10^{-1}$
This study	OC	0.05	2.0	1200	► 1.53 -i 1.0*10 ⁻⁷

Marmer et al., JGR, 2007

Canadian Centre for Climate Modelling and Analysis (CCCma) Atmospheric General Circulation Model.

Table 4. Hydrophobic OC Accumulation Mode ($r = 0.2 \ \mu m$, $\sigma = 2.0$) Optical Properties for the GCM Shortwave Four-Band Radiation Scheme						
Band	$\lambda, \ \mu { m m}$	β , m ² g ⁻¹	ω	g		
1	0.25-0.69	4.6971	0.9236	0.7097		
2	0.69 - 1.19	2.4246	0.9756	0.6650		
3	1.19 - 2.38	0.8997	0.9897	0.5976		
4	2.38 - 4.00	0.1338	0.4227	0.4399		
			D. Baümer et al.,	JGR, 2007		

\rightarrow need to be updated & improved in futur versions for specific plumes (BB ?)



1.2 Aerosol optical properties



-> the main optical aerosol properties : extinction, scattering & absorption

Aerosol optical properties could be estimated in-situ -> example from the AMMA-SOPO experiment

Aerosol optical properties

• light absorption coefficient (σ_{abs}) from MAGEE aethalometer (at 7 λ)



- aerosol optical depth (440, 670, 870 and 1020nm)
- aerosol volume size distribution $(0.05 < r < 10 \ \mu m)$ for the total atmospheric column (Dubovik et al., 2002)
- aerosol single scattering albedo and asymmetry parameter (at 4 λ) for the total atmospheric column (Dubovik et al., 2002)

• vertical profiles of aerosols

E = P

PHOTON (LOA)

Aerosol optical properties on Djougou during the dry season



The quantitative measurement of aerosol light absorption is still a challenge !

Instrument name	Company	Principle	Nominal wavelength(s)	Artifacts due to .
Aethalometer (AE22 & AE31 & AE42 & AE45)	Magee Scientific Company, 2020 Stuart Street, Berkeley, CA 94703, USA. Tel.: +1510-845-2801. http:// www.mageesci.com		880 nm (AE45) 370 & 880 nm (AE22) 370 & 880 nm (AE42) 370 & 470 & 520 & 590 & 660	- modification of particle and filter morphology upon particle deposition
Particle Soot Absorption Photometer (PSAP)	Radiance Research Inc., 535 N.W. 163rd Street, Seattle, WA 98177, USA. Tel.: +1206-366-7981	Aerosol absorption coefficient from real time filter transmission	565 nm	- optical interaction of deposited particles and filter medium
Micro Soot Sensor (AVL 483)	AVL LIST GMBH, Hans-List-Platz 1, A 8020 Graz, Austria, Tel.: +6143 316 787x0. info@avl.com. http:// www.avl.com	BC mass density (for source characterization) from real time, in-situ photoacoustic signal	808 nm	- poor angular integration of light scattered by deposited particles
Multi-Angle Absorption Photometer (MAAP model 5012)	Thermo Fisher Scientific Inc., 81 Wyman Street, Waltham, MA 02454, USA. Tel.: +18662820430. http:// www.thermo.com	Aerosol absorption coefficient, BC mass density from real time filter transmission with scattering correction, automatic filter change	670 nm	(Moonsmuller, 2009)
Multi-Filter Rotating Shadowband Radiometer (MFR-7)	Yankee Environmental Systems, Inc., 101 Industrial Blvd., Turner Falls, MA, USA. Tel.: +1 413 863 0200. info@yesinc.com. http:// www.yesinc.com	Aerosol absorption optical depth and SSA from sun and sky radiance	415 & 500 & 615 & 673 & 870 & 940 nm	
Photo-Acoustic Soot Spectrometer (PASS-1 & PASS- 3)	Droplet Measurement Technologies, 5710 Flatiron Parkway Suite B, Boulder, CO 80301, USA. Tel.: +1 303 440 5576. info@dropletmeasurement.com. http:// www.dropletmeasurement.com	Aerosol absorption coefficient from real time, in-situ photoacoustic signal	781 nm & custom (PASS-1) Three custom wavelengths (PASS-3)	Pasadena, CA 250 -
Single Particle Soot Photometer (SP2)	Droplet Measurement Technologies, 5710 Flatiron Parkway Suite B, Boulder, CO 80301, USA. Tel.: +1 303 440 5576. info@dropletmeasurement.com. http:// www.dropletmeasurement.com	BC mass of individual particles from laser-induced incandescence	1064 nm	v 200 ν 200 ν 50 ν 100 ν 100
Sun Tracking Photometer (CE 318-1 & CE 318-2)	CIMEL Electronique, 172 rue de Charonne, 75011 Paris, France. Tel.: +33143487933. cimel@cimel.fr. http://www.cimel.fr	Aerosol absorption optical depth and SSA from sun and sky radiance	440 & 670 & 870 & 936 &1020 nm	50 WWW www. WWWW www. WWW walke waa wa waa waa waa waa waa waa waa waa
		•	Extinction-mi	nus-scattering technique is a
			promising di	rect technique for the
Anoce	ible new way		t of light absorption using :	
r poss	ible new way		cavity ring-do	wn (CRD) techniques
		-	Cavity Attenu technology (ated Phase Shift (CAPS) (-> see Moonsmuller et al., 2009)

In parallel, column-averaged optical properties could be estimated from remote sensing surface techniques... (see O. Dubovik presentation)



Calculations of aerosol optical properties -> the Mie theory

The absorption and "elastic" scattering of light by a **spherical** particle is a classical problem in physics and the mathematical formalism is -> Mie theory.

The key parameters that govern the scattering and absorption of light by a particle are (1) the wavelength λ of the incident radiation; (2) the size of the particle, usually expressed as a dimensionless size parameter α and (3) the refractive index m



Population n(Dp) of different-sized particles with identical ref. ind. (m)

$$b_{\text{ext}} = \int_{0}^{D_{\text{wet}}^{\text{max}}} \frac{\pi D_{\text{wet}}^2}{4} Q_{\text{ext}}(m, \alpha_{\text{wet}}) n(D_{\text{wet}}) dD_{\text{wet}}$$

*b*_{ext} : Extinction coefficient

Scattering coefficient is computed with the same formula from Q_{scat}

Calculations of aerosol optical properties - the aerosol Refractive Index (RI)

m (λ) = n (λ) - ik (λ)



Limitations of the Mie Theory : Spherical or non-spherical?





Fig. 1. Opper panel: Satellite image of smoke extending from northern California to Reno, Nevada on 10 July 2008. The smoke sources and wind trajectory were similar for much of July. Beneath panel: Conceptual model of emission and aging of urban and biomass burning aerosol.

- Assumption realistic for BB

...the problem is more complex for mineral dust aerosols...



Spherical and non-spherical dust aerosols exhibit dissimilar phase functions and upscatter fractions,

- Not exact for fresh polluted urban aerosols, such as soot !

These difference lead to significant changes in the DRF,

The impact of dust non-sphericity should be considered in satellite and model estimates of aerosol DRF (problem due to the numerical coast),

Optical properties & hygroscopic aerosol properties





Biomass burning

Mineral dust

Mixing !

The aerosol mixing state

<u>Context</u>: coatings of Non-Absorbing (NA) components on strongly Absorbing (A) core can increase the absorption of the composite aerosol (e.g., Fuller et al., 1999; Jacobson, 2001; Stier et al., 2006).



The different kind of aerosol mixing states



Aerosols can appear as :

- externally mixed (1 & 2)
- heterogeneously internally mixed (3,4 & 5)
- homogeneously internally mixed (6 & 7)

Homogeneous internally mixed form by evaporation of droplets containing several species with similar solubility and by simultaneous condensation of semi-volatile species.

They can be found in a variety of combinaisons :

(1) Mixtures of several NA components (NaCl & NA organics) \rightarrow 7

(2) Mixtures of NA comp. (Amm. Sulf.) with weakly or strongly absorbing (Cbrown, BC) $\rightarrow 6$

Coated particles can be form by processes such as **condensation of semi-volatile species on pre-existing particles**, evaporation of droplets containing **two species with different solubilities**.

They can be found :

(1) NA core (NaCl) coated with another NA species $\rightarrow 4$

(2) NA core coated with a weakly absorbing species (Cbrown) $\rightarrow 5$

(3) absorbing core (soot or dust) coated with NA or weakly species \rightarrow 3

Importance of the aerosol mixing on optical properties

The shell acts as a lens and *focuses more photons onto the core*

This lensing effect <u>has been shown theoretically</u> to increase the absorption by an individual *BC* particle by 50-100 % (Bond et al., 2006)



Absorbing enhancement due to lensing <u>has been observed</u> for :
BC particles coated with SOA (Schnaiter et al., 2005),
absorbing spheres coated with organic material (Lack et al., 2009),
absorbing mineral dust coated in aqueous inorganic material,

Importance of the aerosol mixing on optical properties



Based on calculations using core/shell Mie Theory, Lack and Cappa (2010) have shown that the enhancement of light absorption by BC when it is coated by moderate absorbing material (Cbrown) **is reduced** to the one induced by NA coatings

This reduction is sensitive to both the coating thickness and imaginary refractive index, and can be up to 50 % across the visible radiation for reasonable core/shell diameters,

Treatment of the aerosol mixing in climate models

There are **number of approaches** employed to calculate the equivalent RI for optical calculations of internally mixed particles into climate models

1) <u>The « linear » mixing rule</u>, in which the effective RI of the aerosol is calculated by linearly averaging the real and imaginary parts, respectively, weighted by their volume fractions,

2) The « Maxwell-Garnett » approximation, which is one of the most widely used methods for calculating the bulk dielectric properties of inhomogeneous materials (some limitations : size of inclusions $<< \lambda$, volume fraction of the inclusion should be small),

3) Mie scattering code for coated aerosols provide a more explicit calculations. As such codes have high « numerical coast », they are generally applied off-line in climate models,

How can we measure/observe the aerosol mixing state?

Example of <u>nitrate-coated mineral particles</u> collected in regional polluted haze episodes over northern China from <u>transmission electron microscopy</u> (TEM)





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2) Aerosols & impact on climate

2.1 Aerosol radiative properties

2.2 The aerosol « direct » & « semi-direct » effects

2.3 Aerosol & the regional climate -> some examples of the West-African region (AMMA experiment)

2.1) Aerosols radiative properties

Key radiative parameters of the RTE



«A radiative forcing is a change imposed on the Earth's radiation balance »

D	daylight fraction
S ₀	solar constant
T _{at}	atmospheric transmission
A _c	cloud fraction
R _s	surface albedo



aerosol optical depth



δ

aerosol singlescattering albedo



average aerosol up-scatter fraction

Aerosol Optical Depth



RONET Project,

Aerosol Single Scattering Albedo (SSA)

$$\succ \omega_{o}(\lambda) : \text{ratio of scattering to extinction}$$
$$\omega_{o}(\lambda) = K_{\text{scat.}}(\lambda) / (K_{\text{scat.}}(\lambda) + K_{\text{abs}}(\lambda))$$
$$(0 < \omega_{o}(\lambda) < 1)$$
$$\Rightarrow \text{Black carbon}: \omega_{o,550} = 0.26 - 0.40$$
$$\Rightarrow \text{Sulfate \& sea salt}: \omega_{o,550} = 0.99$$





SSA for BB and mineral dust from recent in-situ observations -> the AMMA-SOPO experiment

Mean BB SSA of **0.81** (visible range) lower than the one (~0.85) obtained during SAFARI (South-Africa)

SSA close to 1.0 (visible range) for submicronic dust and 0.90 for the total (fine & coarse) aerosol size distribution



Figure 12. Schematic zonal cross section showing the northward transport of biomass burning aerosol in warm, ascending air (red arrows) and the westward/southward transport of mineral dust in a cooler airflow channeled by the mountainous regions to the north (blue arrows). The "Harmattan front" is shown by the solid line which marks the boundary between the two air masses with arrows representing mixing of the dust with the biomass burning smoke. The intertropical front marks the boundary between the moist monsoon air and the smoke-laden air. A deep convective cloud associated with the ITCZ is represented to the south.



Osborne et al., 2008 Johnson et al., 2008; Haywood et al., 2008 McConnell et al., 2008

Recent SSA estimations over « urban atmosphere » show very low values (visible λ)...



Large SW radiative heating due to highly absorbing aerosols :

-> impact on the « urban » meteorology : urban / peri-urban breeze, Urban Bound. Layer, Air Quality ??

AERONET observations offer a general view of SSA for different aerosol species



Large variability of urban/Ind and Biom. Burn. SSA at 440 nm,

Desert dust SSA ~ 0.90 at 440 nm, <u>SSA increases with λ </u> contrary to BB and UI aerosols

Oceanic aerosols SSA ~ 1.00 for the whole λ : purely scattering species

Some examples of recent SSA aerosol properties retrieved from space...



Cooling versus Heating of an aerosol layer

-> the importance of SSA on the aerosol direct forcing !



Above a critical surface albedo ($R_s \sim 0.2-0.3$) absorbing aerosols could change the sign of TOA forcing !

For the critical R_s:

-> SSA ~ 0.95 induces « cooling » effect at TOA

-> SSA ~ 0.80 induces « heating » effect at TOA

Aerosol asymmetry parameter (g)





The importance of the vertical profiles of anthropogenic vs natural particles on the direct radiative forcing



Figure 10. Aerosol backscatter from the CALIPSO lidar (1032 nm) during transects across N Africa: (a, b) 7 January 2007 and (c, d) 18 January 2007. The highlighted boxes in Figures 10b and 10d show close-ups of the area from approximately 22°N to 5°N. The dust layers are shown by red and grey (backscatter > $4-10 \times 10^{-2}$ km⁻¹sr⁻¹), while yellow represents biomass burning influenced aerosol $(backscatter > 1-2 \times 10^{-2} km^{-1} sr^{-1}).$



SW RT calculations show that the radiative effect of biomass burning aerosol was sensitive to the vertical distribution of aerosol.

(a)

dust

1.000

When the observed low-level dust layer was included in the model, the absorption of solar radiation by BB increased by 10%.

This absorption enhancement was caused by the dust reflecting solar radiation up into the biomass burning aerosol layer.

 \rightarrow the radiative forcing of anthropogenic absorbing aerosol can be sensitive to the presence of natural aerosol species. (Jonhson et al., JGR, 2008)

2.2) Aerosol direct & semi-direct effect

Aerosol direct effect

represents the capacity of particles to scatter/absorb solar and/or IR (dust) radiations

\rightarrow depends on the physical, chemical, optical and mixing state of aerosols



^{IPCC, 2007} Scattering Aerosol ("<u>whitehouse"</u> aerosol) Sulfates ~ -0.4 W m ⁻² Organic Carbon ~ -0.05 W m ⁻²



Absorbing particle ("<u>greenhouse"</u> aerosol) IPCC, 2007 BC ~ <u>+ 0.20 W m ⁻²</u>

Satellite retrievals of direct radiative effect at the global scale

Table 2.3. The direct aerosol radiative effect (DRE) estimated from satellite remote sensing studies (adapted and updated from Yu et al., 2006).

Reference	Instrument ^a	Data Analysed	Brief Description	Clear Sky DRE (W m ^{_2}) ocean
Bellouin et al. (2005)	MODIS; TOMS; SSM/I	2002	MODIS fine and total τ_{aer} with TOMS Aerosol Index and SSM/I to discriminate dust from sea salt.	-6.8
Loeb and Manalo-Smith (2005)	CERES; MODIS	Mar 2000 to Dec 2003	CERES radiances/irradiances and angular distribution models and aerosol properties from either MODIS or from NOAA-NESDIS ^b algorithm used to estimate the direct radiative effect.	–3.8 (NESDIS) to –5.5 (MODIS)
Remer and Kaufman (2006)	MODIS	Aug 2001 to Dec 2003	Best-prescribed aerosol model fitted to MODIS data. τ_{aer} from fine-mode fraction.	-5.7 ± 0.4
Zhang et al. (2005); Christopher and Zhang (2004)	CERES; MODIS	Nov 2000 to Aug 2001	MODIS aerosol properties, CERES radiances/irradiances and angular distribution models used to estimate the direct radiative effect.	-5.3 ± 1.7
Bellouin et al. (2003)	POLDER	Nov 1996 to Jun 1997	Best-prescribed aerosol model fitted to POLDER data	-5.2
Loeb and Kato (2002)	CERES; VIRS	Jan 1998 to Aug 1998; Mar 2000.	τ_{aer} from VIRS regressed against the TOA CERES irradiance (35°N to 35°S)	-4.6 ± 1.0
Chou et al. (2002)	SeaWiFs	1998	Radiative transfer calculations with SeaWiFS τ_{aer} and prescribed optical properties	-5.4
Boucher and Tanré (2000)	POLDER	Nov 1996 to Jun 1997	Best-prescribed aerosol model fitted to POLDER data	–5 to –6
Haywood et al. (1999)	ERBE	Jul 1987 to Dec 1988	DRE diagnosed from GCM-ERBE TOA irradiances	-6.7
Mean (standard deviation)				-5.4 (0.9)

Reasonable agreement of the global mean, diurnally averaged clear-sky DRE from various studies : **mean DRE of - 5.4 W m⁻²** standard deviation of 0.9 W m⁻²

Notes:

^a SSM/I: Special Sensor Microwave/Imager; VIRS: Visible Infrared Scanner; ERBE: Earth Radiation Budget Experiment.

^b NESDIS: National Environmental Satellite, Data and Information Service

Kaufman et al. (2005) estimated the anthropogenic-only component of the aerosol <u>fine-mode fraction</u> from the MODIS product to deduce a clear sky RF over ocean of -1.4 W m^{-2}

Aerosol direct effect at global scale from modelling studies

Since the IPCC 2001, more complete aerosol modules in a larger number of global atmospheric models now provide estimates of the direct RF,

→ considerable enhancement over the models → now include the most important anthropogenic and natural species,

Some of the more complex models now account : - explicitly for the dynamics of the aerosol size distribution, - parametrize the internal/external mixing of the various aerosol components,

« The mean and median of the sulphate direct RF from grouping all these studies together are identical at -0.41 W m⁻² »

« The mean and median for the direct RF of fossil fuel organic carbon from grouping all these studies together are identical at -0.05 W m^{-2} with a standard deviation of 0.03 W m⁻² »

« The mean and median of the direct RF for fossil fuel BC from grouping all these studies together are +0.19 and +0.16 $W m^{-2}$, respectively, with a standard deviation of nearly 0.10 $W m^{-2}$ »

Aerosol semi-direct effect

An additional, hypothesized effect of absorbing tropospheric aerosols is <u>the semi-direct effect (SDE)</u> [Hansen et al., 1997; Ackerman et al., 2000],

Absorbing aerosols are thought to warm the layer in which they are located \rightarrow decreased relative humidity \rightarrow less cloud cover \neq indirect effect,



Aerosol semi-direct effect from satellite observations

Smoke Invigoration Versus Inhibition of Clouds over the Amazon

llan Koren,¹ J. Vanderlei Martins,^{2,3} Lorraine A. Remer,³ Hila Afargan¹

The effect of anthropogenic aerosols on clouds is one of the most important and least understood aspects of human-induced climate change. Small changes in the amount of cloud coverage can produce a climate forcing equivalent in magnitude and opposite in sign to that caused by anthropogenic greenhouse gases, and changes in cloud height can shift the effect of clouds from cooling to warming. Focusing on the Amazon, we show a smooth transition between two opposing effects of aerosols on clouds: the microphysical and the radiative. We show how a feedback between the optical properties of aerosols and the cloud fraction can modify the aerosol forcing, changing the total radiative energy and redistributing it over the atmospheric column.

The semi-direct effect ...from the Large Eddy Simulations (LES) point of view...

LES was used to investigate the SDE for marine stratocumulus & examine the dependency on the vertical distribution (*indirect effect is excluded*)



Recent work using GCM odels indicates a more complex interaction

Cloud cover increase with increasing aerosol absorptivity: A counterexample to the conventional semidirect aerosol effect

Jan Perlwitz1,2 and Ron L. Miller1,2

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Contrary to the expected decrease in low cloud cover due to heating by tropospheric aerosols, Perlwitz and Miller, 2010 find a significant increase with increasing absorptivity of dust in regions with high dust load

The cloud cover change is directly linked to the change in relative humidity (RH) as a result of changes in specific humidity (SH) and temperature (T),

(1) more absorption by aerosols \rightarrow larger diabatic heating and increased warming \rightarrow decreasing RH,

(2) however, a corresponding increase in SH exceeds the T effect on RH.



2.3) Aerosol & the regional scale

-> Example over the WA region (AMMA experiment)



BOA direct forcing efficiency (W.m²:E)

·150 ^{__}

Due to the direct radiative forcing at the regional scale, more and more studies are now focused on the effect of particles on the regional climate

In most of cases, the *regional DRF* due to aerosols is clearly larger than GHG !

A large difference between BOA and TOA DRF *is observed* due to absorbing aerosols !

What is the impact of DRF on the atmospheric dynamic, heating rate profiles, hydrological cycle, cloud cover,??

Impact of mineral dust on the West. Africa regional climate case of March 2006 - Meso-scale simulations



-750-850-550-450-350-250-150-50 0 50 150 250 350 450 550 See F. Solmon presentation for long term climatic simulations... Tulet, Mallet et al., JGR, 2008

Thank you for your attention!