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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

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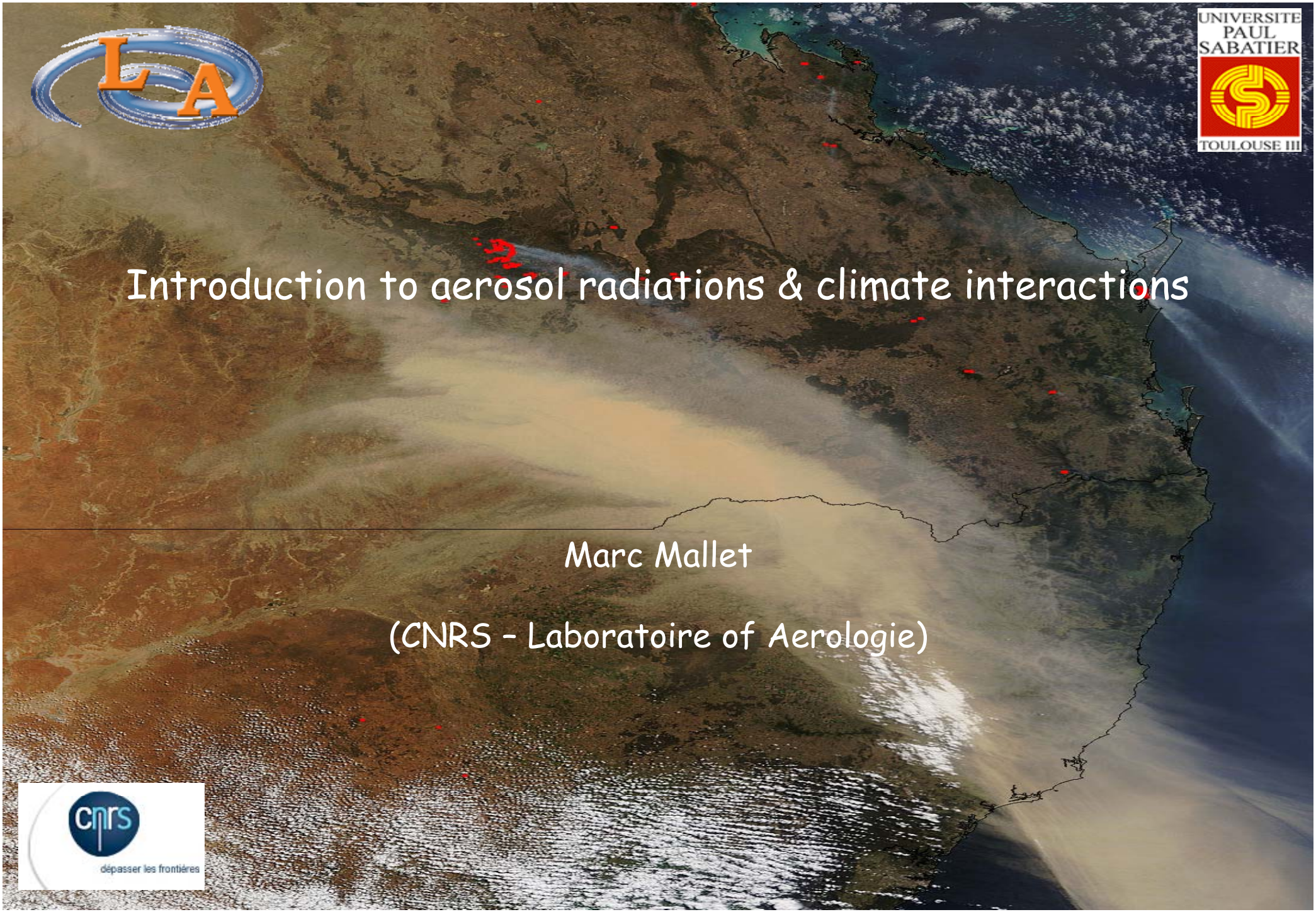
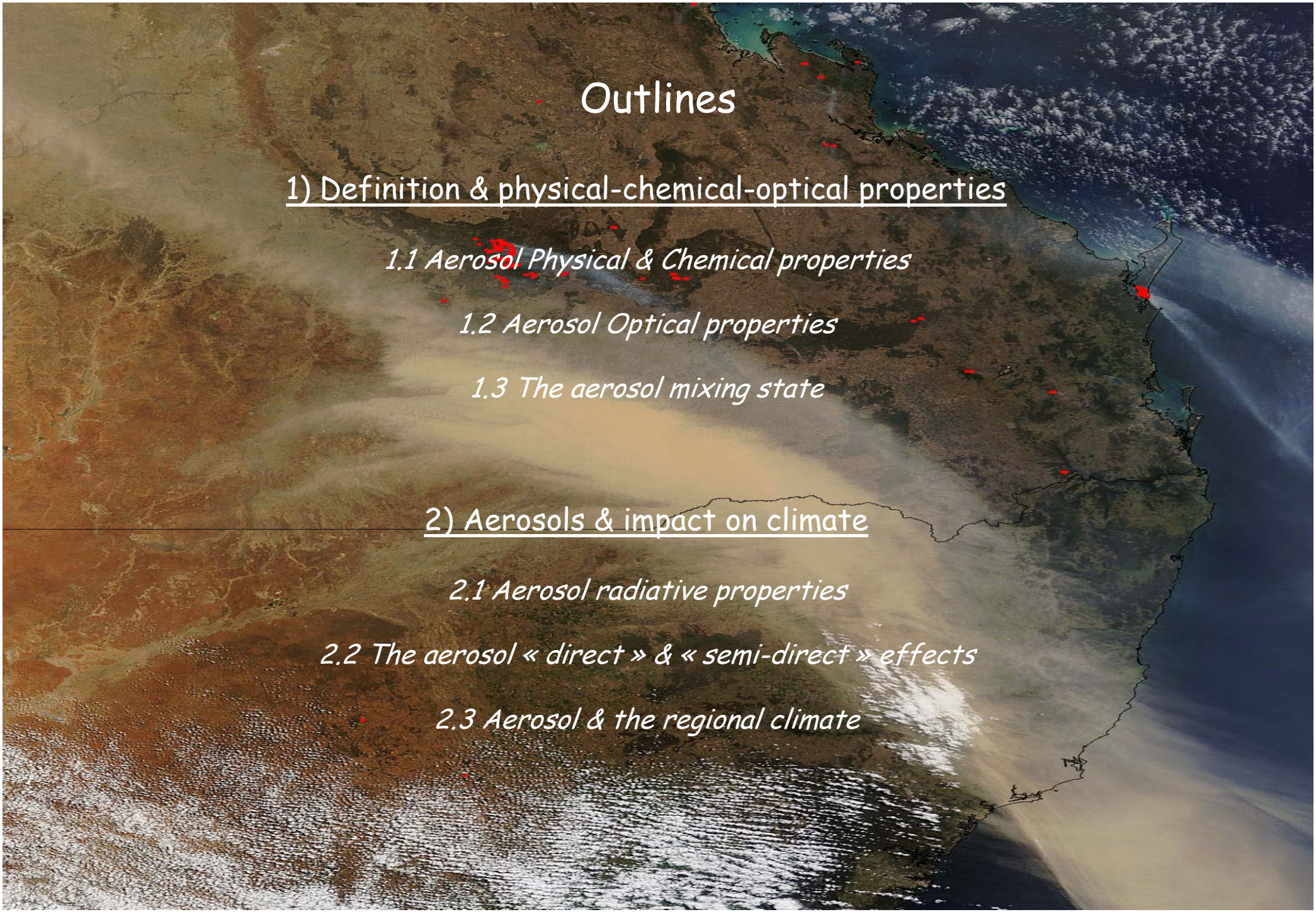


Introduction to aerosol radiations & climate interactions

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 :

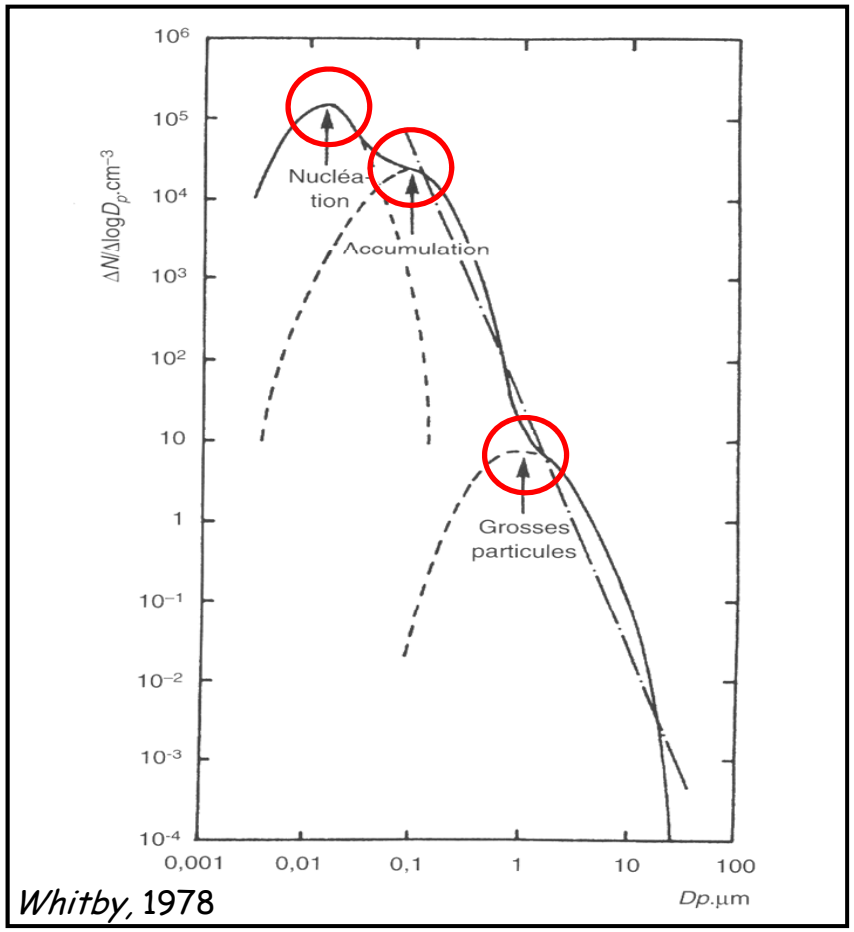
- sulfates aerosols
- nitrates particles,
- « Secondary » organics,...

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

1.1 Aerosol physical and chemical properties

-> large range of atmospheric concentration !

*With **a log scale for the diameter**, both small and large particles size ranges are depicted*



Whitby, 1978

Nucleation mode (10nm < D_p < 100 nm) : formed in the atmosphere by gas-to-particle conversion processes,

Optically (SW & LW) active

Accumulation mode (0.1 < D_p < 2.5 µm) : formed both by ***coagulating of particles*** in the nuclei mode and from condensation of vapors onto existing particles -> SW radiations

Coarse mode (D_p > 2.5 µm) : primary aerosols directly introduced in the atmosphere
-> LW radiations,

The parametrization generally used : the log-normal size distribution

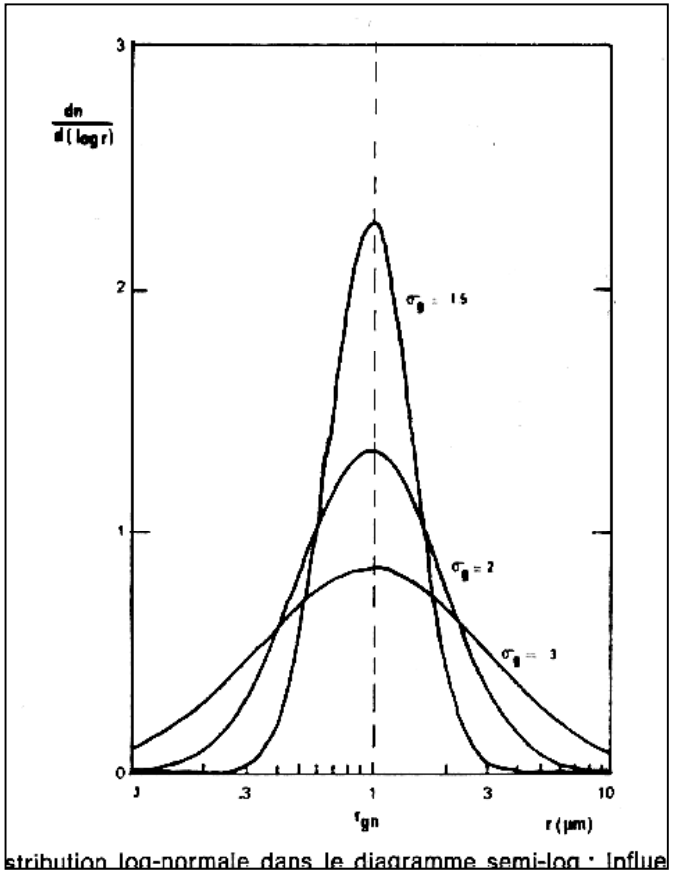
Equation:

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

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

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

N : total aerosol number concentration in the considered mode



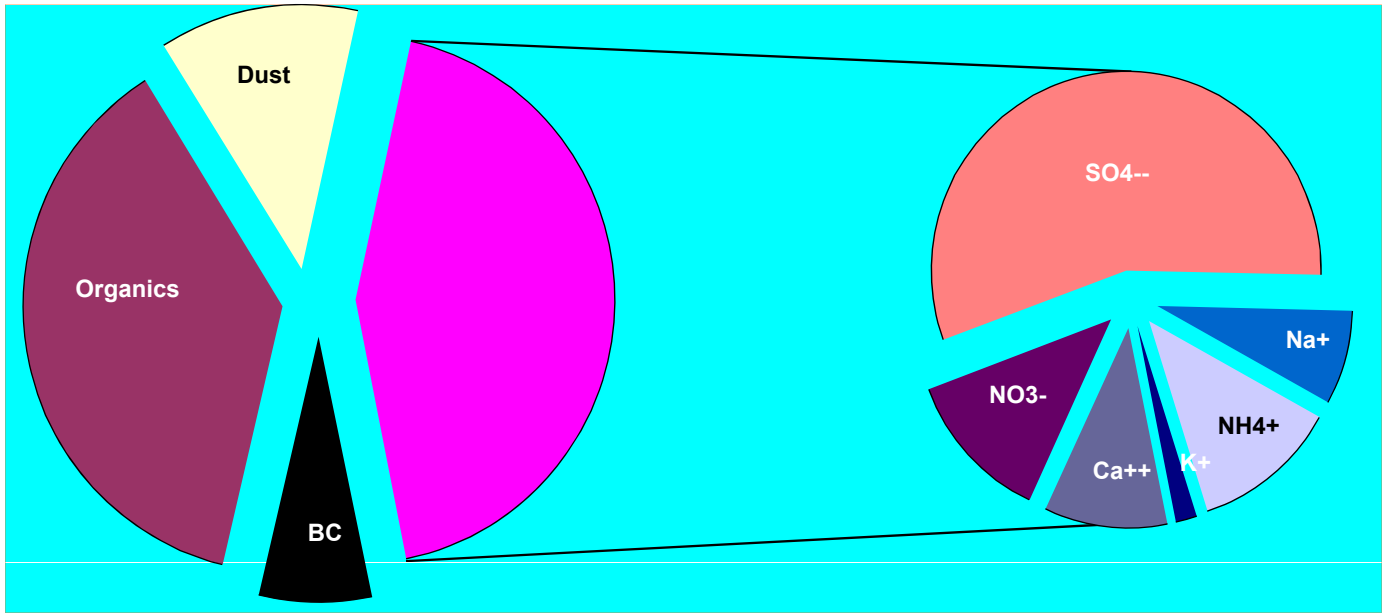
stribution log-normale dans le diagramme semi-log · Influe

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

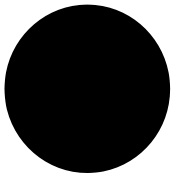
- none negligible contribution of BC, dust & nitrates

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

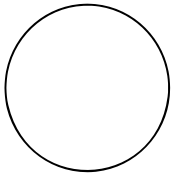
Cachier et al. , 2005

Carbonaceous aerosols : BC, OC and Cbrown

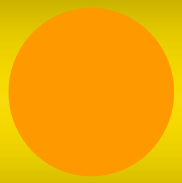
The carbonaceous fraction of ambient particulate matter consists of ***black carbon*** *and a variety of* ***organic compounds*** *(organic carbon)*



Black carbon aerosol is ***emmited directly into the atmosphere during combustion***
→ 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)*
→ 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****,* ***which can include HULIS,*** *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. Anderson[3]

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 $1.67 - 0.27i$ (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.

Science, 2008

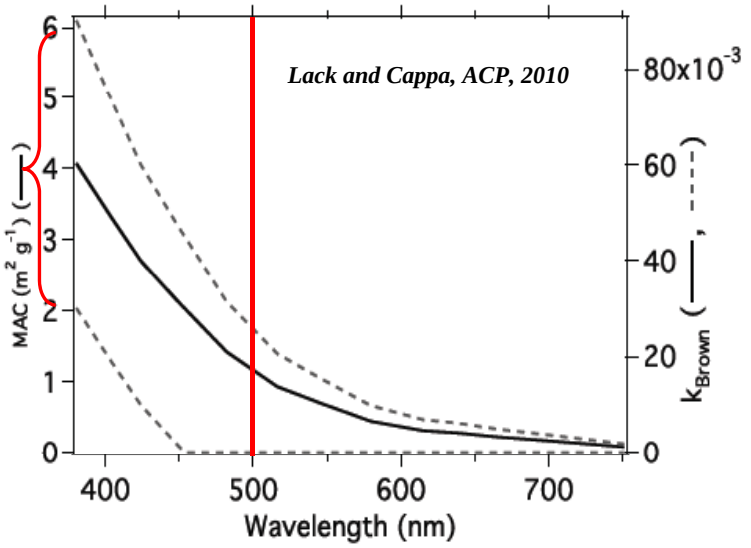


Fig. 2. Wavelength dependent mass absorption cross-section (MAC) of C_{Brown} with a form as given by Sun et al. (2007) and where the absolute magnitude of the k_{Brown} (solid black line) has been deduced from Barnard et al. (2008). Dashed lines indicate k_{Brown} upper and lower bounds for our modeling.

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, σ = standard deviation, ρ_p = particle density, m = refractive index, σ_{ext} = dry extinction cross section, α hygroscopic growth parameter, $f_{rh=0.8}$ = hygroscopic growth factor at 80% relative humidity

Species	r_0 (μm)	σ	ρ_p (g cm^{-3})	$m = n\text{-}ik$ 380–550 nm	σ_{ext} (m^2 g^{-1}) 380 nm	550 nm	α	$f_{rh=0.8}$
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
OC_{hl}	0.1	2.	1.7	1.55–0.005i	9.8	4.9.	0.25	1.49

Solmon et al. Tellus, 2006

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

Table 1. Aerosol Physical and Optical Properties as Found in Literature

Author	Aerosol	r_{gN}, μm	σ_g	Density, kg(m^{-3})	Refractive Indices at 0.55 μm
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

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

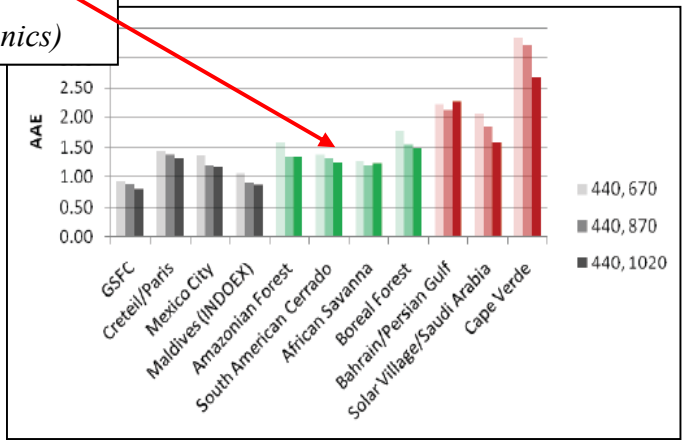
Canadian Centre for Climate Modelling and Analysis (CCCma) Atmospheric General Circulation Model.

Table 4. Hydrophobic OC Accumulation Mode (r = 0.2 μm, σ = 2.0) Optical Properties for the GCM Shortwave Four-Band Radiation Scheme

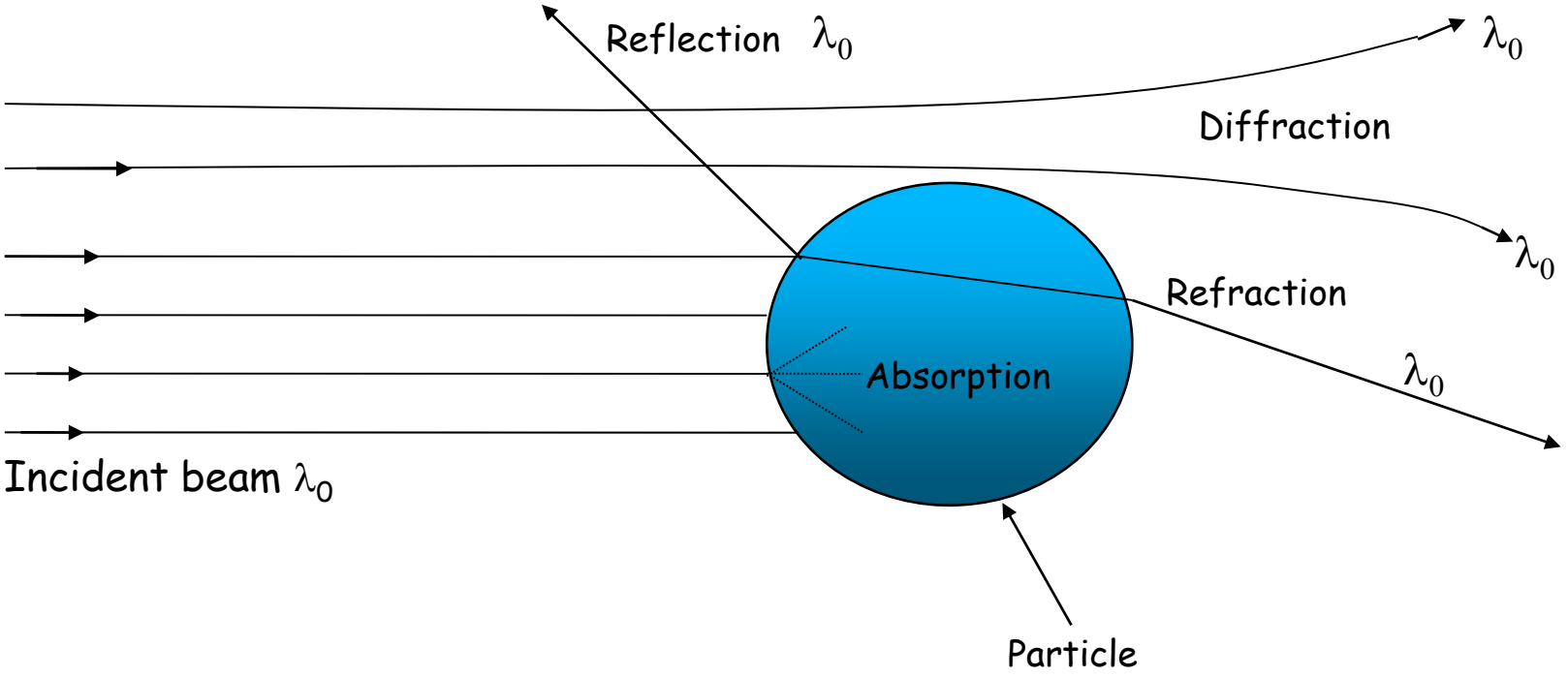
Band	λ, μm	β, m^2 g^{-1}	ω	g
1	0.25–0.69	4.6971	0.9236	0.7097
2	0.69–1.19	2.4246	0.9756	0.6650
3	1.19–2.38	0.8997	0.9897	0.5976
4	2.38–4.00	0.1338	0.4227	0.4399

D. Baümer et al., JGR, 2007

AERONET datas could be used to contrain modelisation outputs -> Abs. Angs. Exp. (AAE > 1 for abs. organics)



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 7λ)*

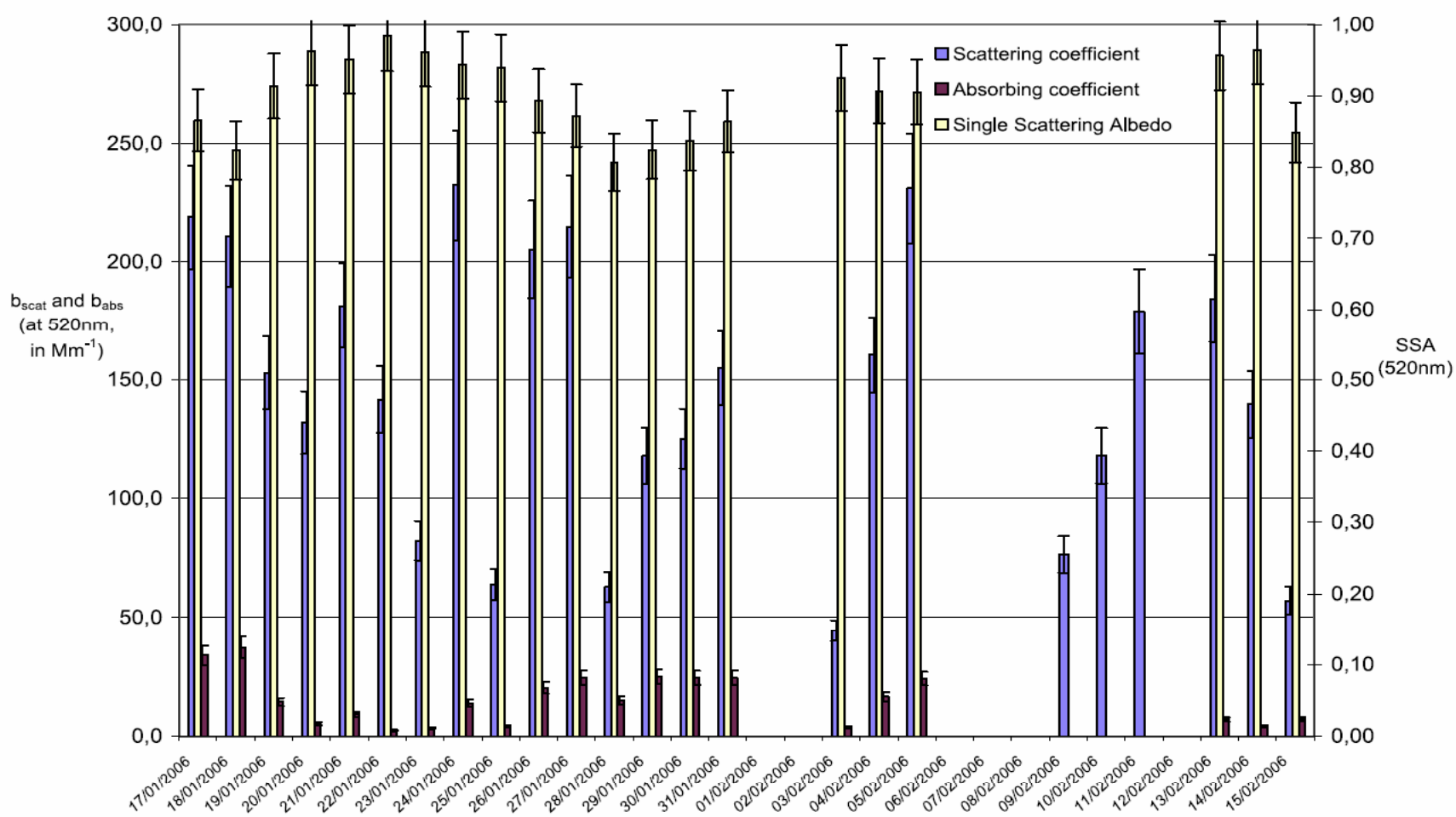
• *light scattering coefficient (σ_{scat}) from ECOTECH nephelometer (at 520nm)*

PHOTON (LOA)

• *aerosol optical depth (440, 670, 870 and 1020nm)*
• *aerosol volume size distribution (0.05 < r < 10 µm) 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*

Aerosol optical properties on Djougou during the dry season



Mallet et al., JGR, 2008
Pelon, Mallet et al., JGR, 2008

Absorbing coefficient
(corrected from Weingartner et al., 2003)
$2 \leq \sigma_{abs} \leq 37\ Mm^{-1}$ *(520nm)*
mean $\sigma_{abs} = 15.2\ Mm^{-1}$

Scattering coefficient
(corrected from Heintzenberg et al., 2006)
$44 \leq \sigma_{scat} \leq 230\ Mm^{-1}$ *(520 nm)*
mean $\sigma_{scat} = 145\ Mm^{-1}$

mean SSA ~ 0.91 ± 0.05

0.81 < SSA < 0.98 (520 nm)

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

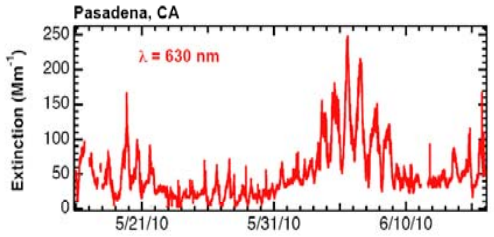
Table 3
Some commercially available instruments related to the measurement of aerosol light absorption.

Instrument name	Company	Principle	Nominal wavelength(s)
Aethalometer (AE22 & AE31 & AE42 & AE45)	Magee Scientific Company, 2020 Stuart Street, Berkeley, CA 94703, USA. Tel.: +1 510-845-2801. http://www.mageesci.com	BC mass density from real time filter transmission, automatic filter change	880 nm (AE45) 370 & 880 nm (AE22) 370 & 880 nm (AE42) 370 & 470 & 520 & 590 & 660 & 880 & 950 nm (AE31)
Particle Soot Absorption Photometer (PSAP)	Radiance Research Inc., 535 N.W. 163rd Street, Seattle, WA 98177, USA. Tel.: +1 206-366-7981	Aerosol absorption coefficient from real time filter transmission	565 nm
Micro Soot Sensor (AVL 483)	AVL LIST GMBH, Hans-List-Platz 1, A 8020 Graz, Austria, Tel.: +61 43 316 787x0. info@avl.com. http://www.avl.com	BC mass density (for source characterization) from real time, in-situ photoacoustic signal	808 nm
Multi-Angle Absorption Photometer (MAAP model 5012)	Thermo Fisher Scientific Inc., 81 Wyman Street, Waltham, MA 02454, USA. Tel.: +1 866 282 0430. http://www.thermo.com	Aerosol absorption coefficient, BC mass density from real time filter transmission with scattering correction, automatic filter change	670 nm
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)
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
Sun Tracking Photometer (CE 318-1 & CE 318-2)	CIMEL Electronique, 172 rue de Charonne, 75011 Paris, France. Tel.: +33 1 43 48 79 33. cimel@cimel.fr. http://www.cimel.fr	Aerosol absorption optical depth and SSA from sun and sky radiance	440 & 670 & 870 & 936 & 1020 nm

Artifacts due to :

- *modification of particle and filter morphology upon particle deposition*
- *optical interaction of deposited particles and filter medium*
- *poor angular integration of light scattered by deposited particles.*

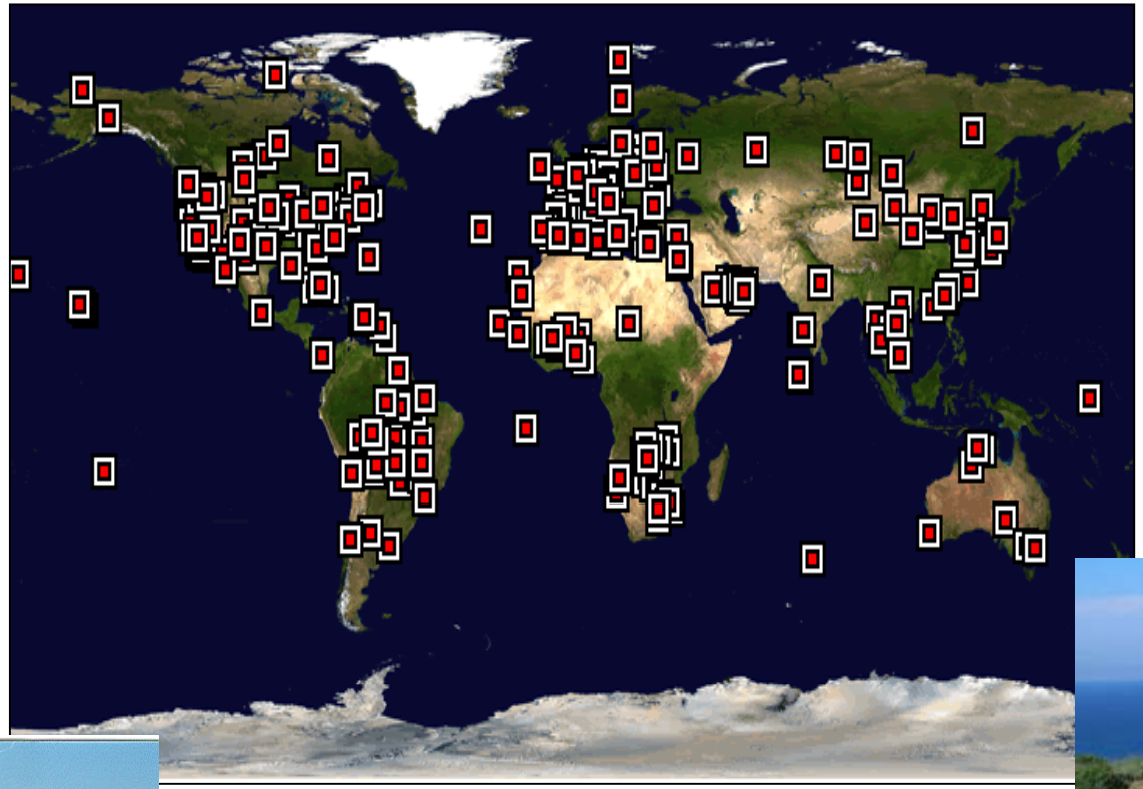
(Moonsmuller, 2009)



A possible new way…

- Extinction-minus-scattering technique is a **promising direct technique for the measurement of light absorption** using :
  - cavity ring-down (CRD) techniques
  - Cavity Attenuated Phase Shift (CAPS) technology (-> see Moonsmuller et al., 2009)

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



Aerosol remote sensing from the surface
- the AERONET network -

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**

Aerosol size parameter
$\alpha = \pi D_p/\lambda$

Mie calculations

Aerosol refractive index
$m(\lambda) = n(\lambda) - i(\lambda)$

Scattering and extiction efficiencies of a spherical particle :
$Q_{scat}(m,\alpha)$
$Q_{ext}(m,\alpha)$

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

$$b_{\text{ext}} = \int_0^{D_{\text{wet}}^{\max}} \frac{\pi D_{\text{wet}}^2}{4} Q_{\text{ext}}(m, \alpha_{\text{wet}}) n(D_{\text{wet}}) \mathrm{d}D_{\text{wet}}$$

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)

$$m(\lambda) = n(\lambda) - ik(\lambda)$$

Absorbing aerosols ($k \neq 0$) :

Black carbon : large range of values !
1.80 – 0.74i to 2.00 - 1.00i

Bond et al. (2006) : 1.95 - 0.79i

Moderate absorbing aerosols ($k \neq 0$) :

Some mineral dust aerosols (McConnell et al., 2010)

Cbrown : 1.67 – 0.27i (0.55 µm) (Alexander et al., 2008)

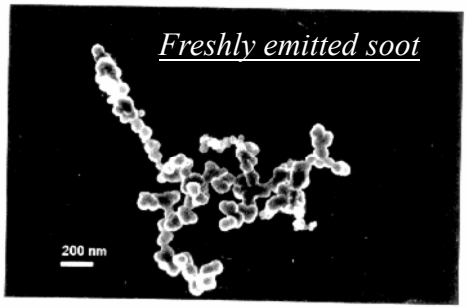
Scattering aerosols ($k \sim 0$) :

Sulfate : $m = 1.53 - 6.10^{-3}i$

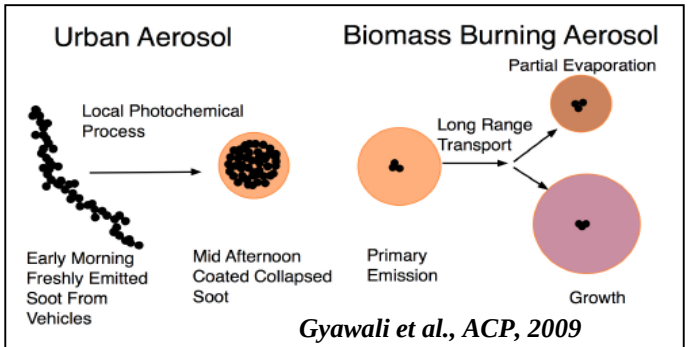
Sea salt : $m = 1.55 - 210^{-8}i$

Organics : m = 1.55 - 0.005i

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



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

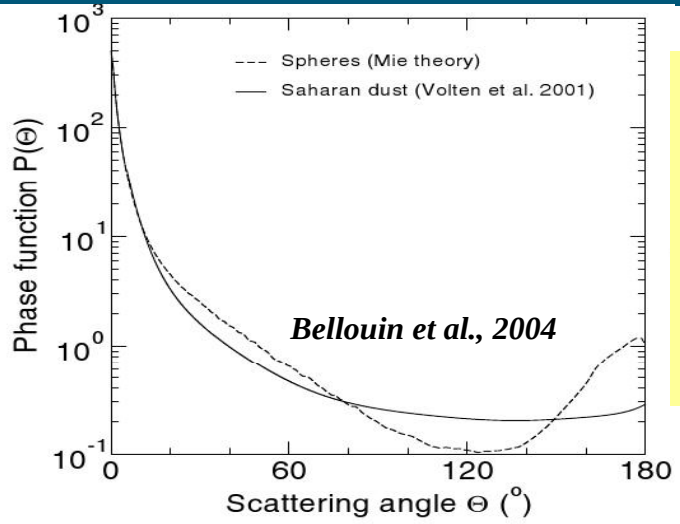


Gyawali et al., ACP, 2009

Fig. 1. Upper 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...



Bellouin et al., 2004

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

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

➢ represents the « affinity » of a particle with H_2O,

➢ critical property for indirect (cloud modification) and direct (change in optical properties -> RI) effects

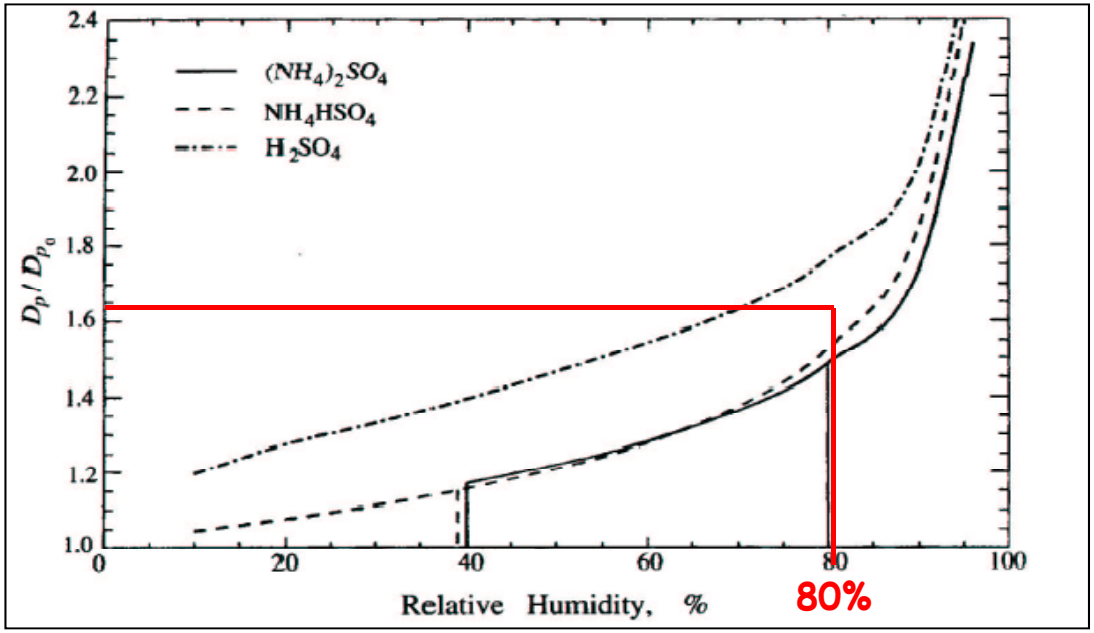
Hydrophilic particles

→ sulfate, nitrate, sea salt, ammonium & secondary organics

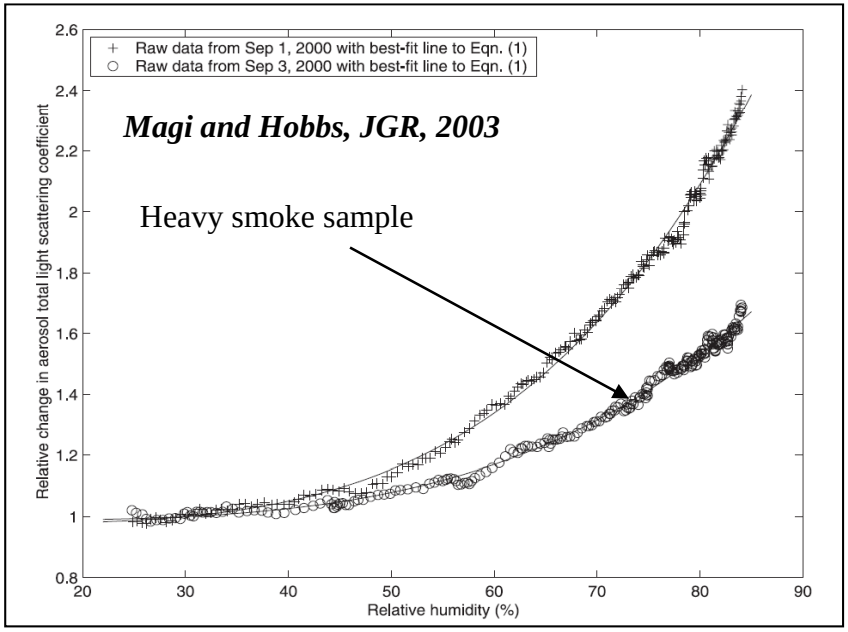
Hydrophobic aerosols

→ « **non aged** » fresh black carbon (BC) & some mineral dust

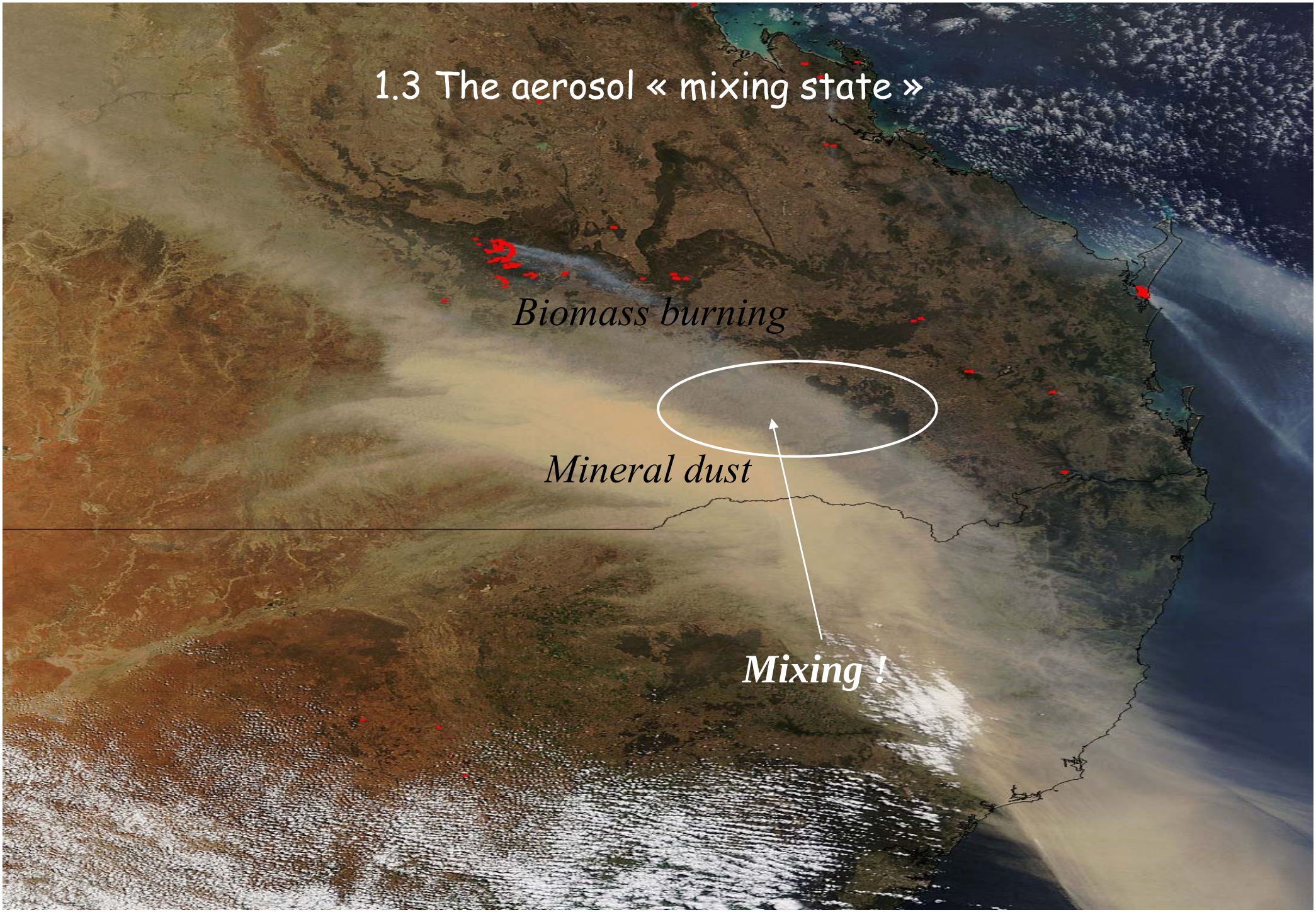
Microphysical properties



Optical properties

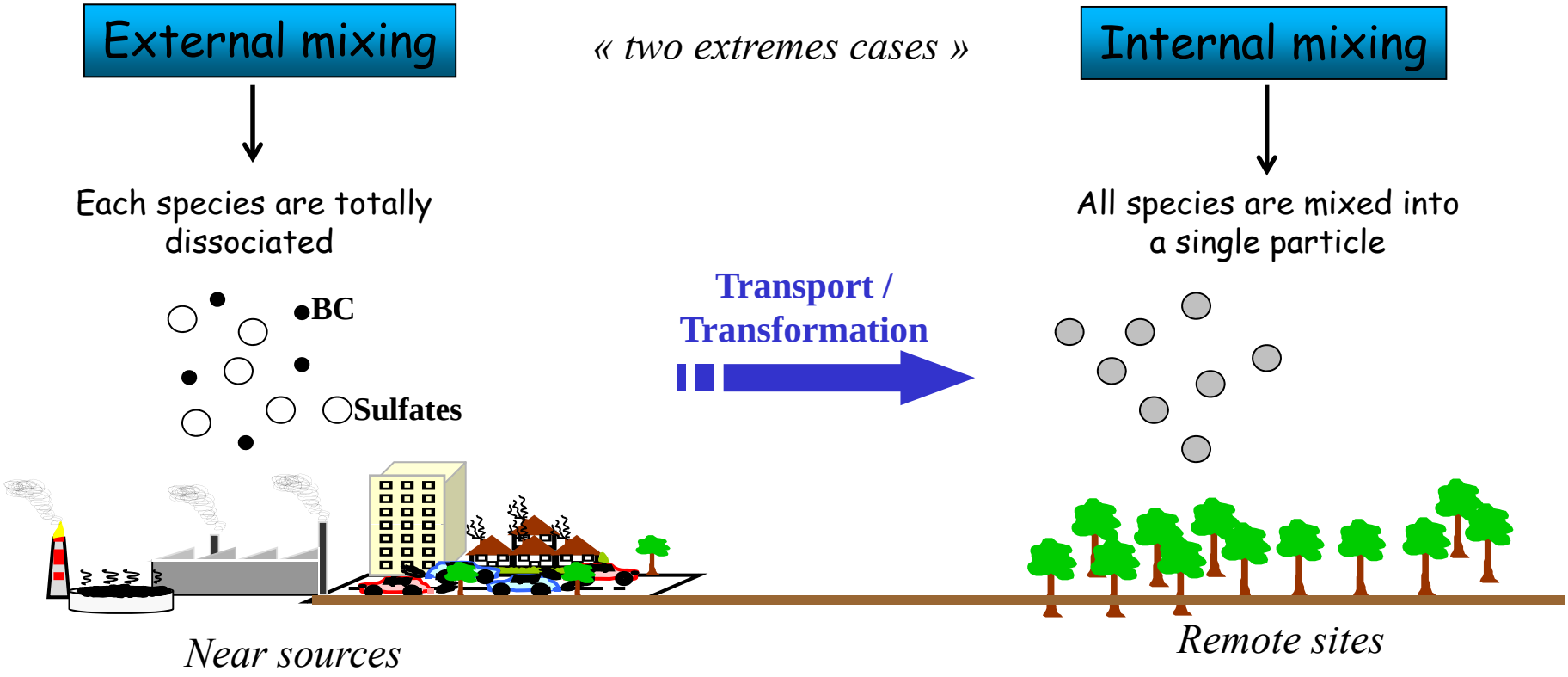


1.3 The aerosol « mixing state »

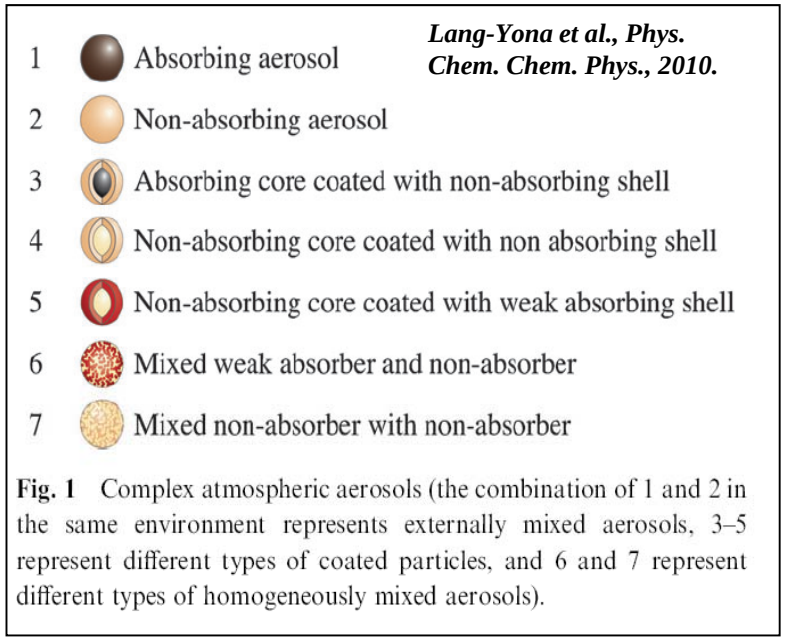


The aerosol mixing state

Context : *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



Lang-Yona et al., Phys. Chem. Chem. Phys., 2010.

Fig. 1 Complex atmospheric aerosols (the combination of 1 and 2 in the same environment represents externally mixed aerosols, 3–5 represent different types of coated particles, and 6 and 7 represent different types of homogeneously mixed aerosols).

Aerosols can appear as :

- externally mixed (1 & 2)

- heterogeneously internally mixed (3,4 & 5)

- homogeneously internally mixed (6 & 7)

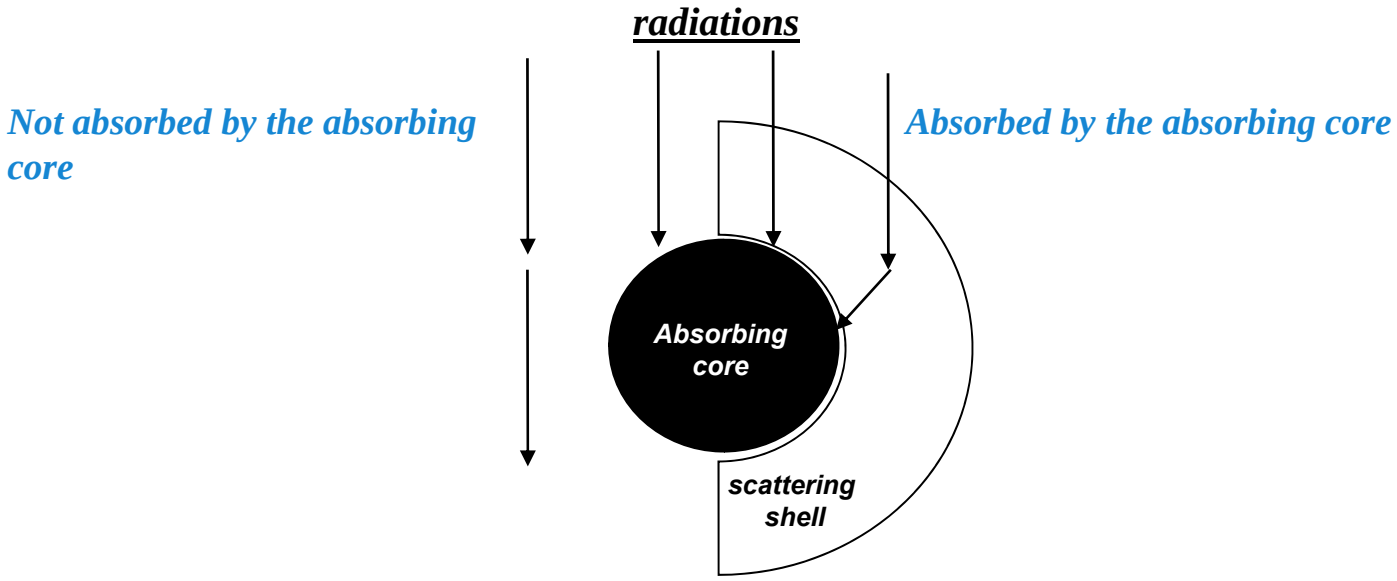
Homogeneous internally mixed form by evaporation of droplets containing several species ***with similar solubility*** *and by* ***simultaneous condensation of semi-volatile species****.*
They can be found in a variety of combinaisons :
(1) Mixtures of several NA components (NaCl & NA organics) → 7
(2) Mixtures of NA comp. (Amm. Sulf.) with weakly or strongly absorbing (Cbrown, BC) → 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 → 4
(2) NA core coated with a weakly absorbing species (Cbrown) → 5
(3) absorbing core (soot or dust) coated with NA or weakly species → 3

Importance of the aerosol mixing on optical properties

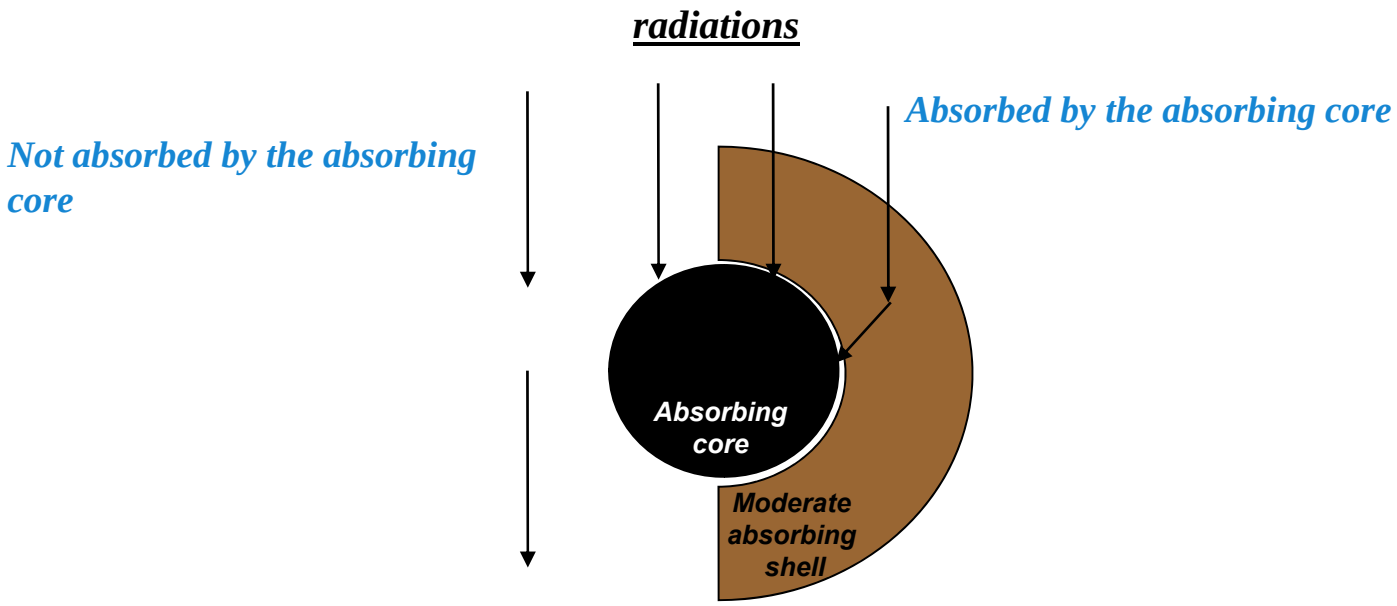
The shell acts as a lens and ***focuses more photons onto the core***

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



Absorbing enhancement due to lensing ***has been observed*** *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) **The « linear » mixing rule**, 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 << λ, 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 nitrate-coated mineral particles collected in regional polluted haze episodes over northern China from transmission electron microscopy (TEM)

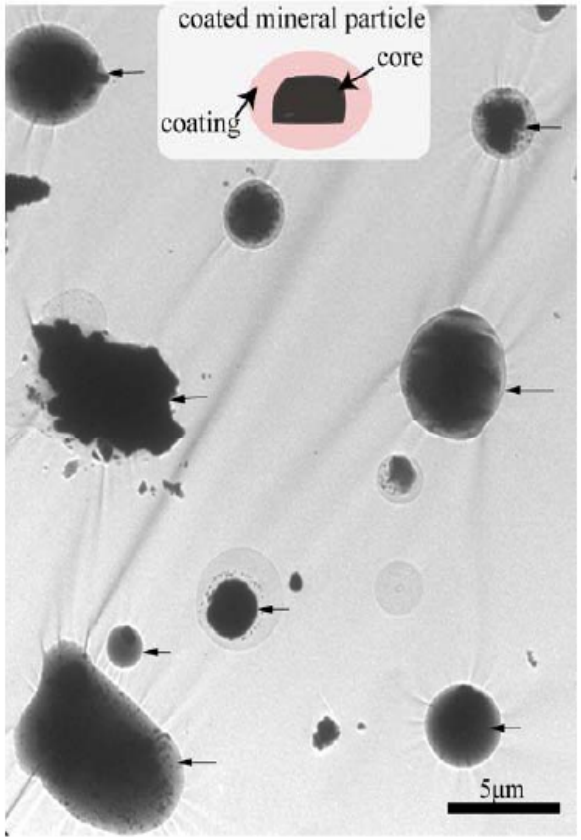


Fig. 2. TEM image of the coated mineral particles. Coated mineral particles include two parts: core and coating. Arrows indicate coated mineral particles.

W. J. Li and L. Y. Shao, ACP, 2009

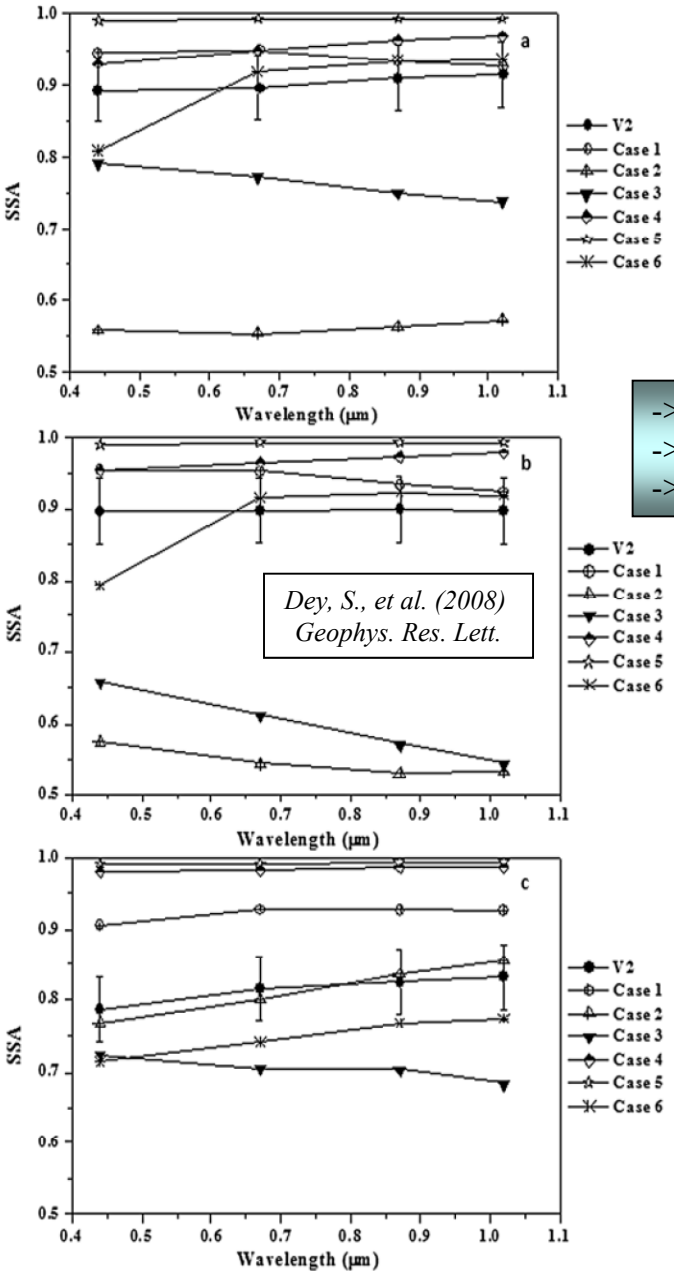
Katrinak et al. [1992, 1993], Posfai et al. [1999], and Clarke et al. [2004] used TEM to identify mixing of soot & sulfate particles,

- Humidified Tandem Differential Mobility Analyzer (HTDMA) system can be used to investigate the mixing state of the hygroscopic and hydrophobic aerosols,

- HTDMA measurements revealed bimodal distributions or more subpopulations in many submicrometer particle size range [e.g., Heintzenberg et al., 2001; Carrico et al., 2005],

- The mixing state can be also inferred from the size-resolved volatility by using a VTDMA (Volatile Tandem Differential Mobility Analyzer) [Clarke et al., 2004] -> core/shell,

Use of the optical (absorbing) closure…

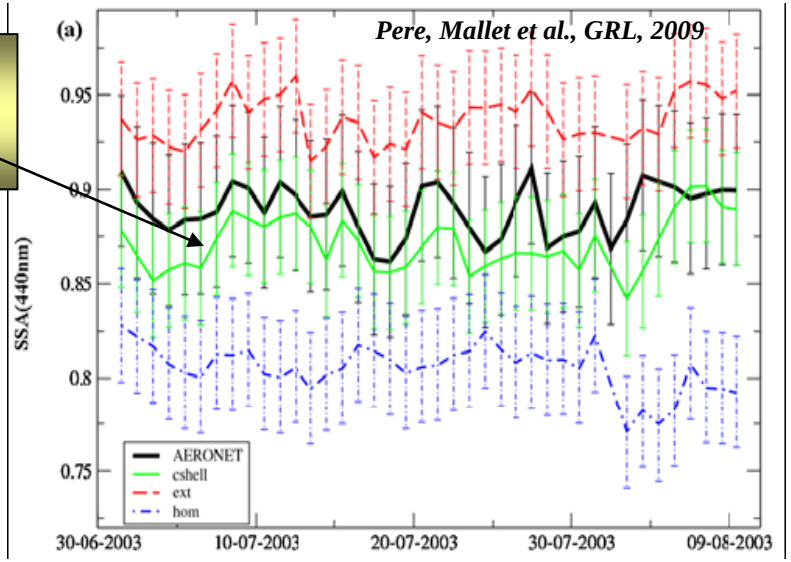


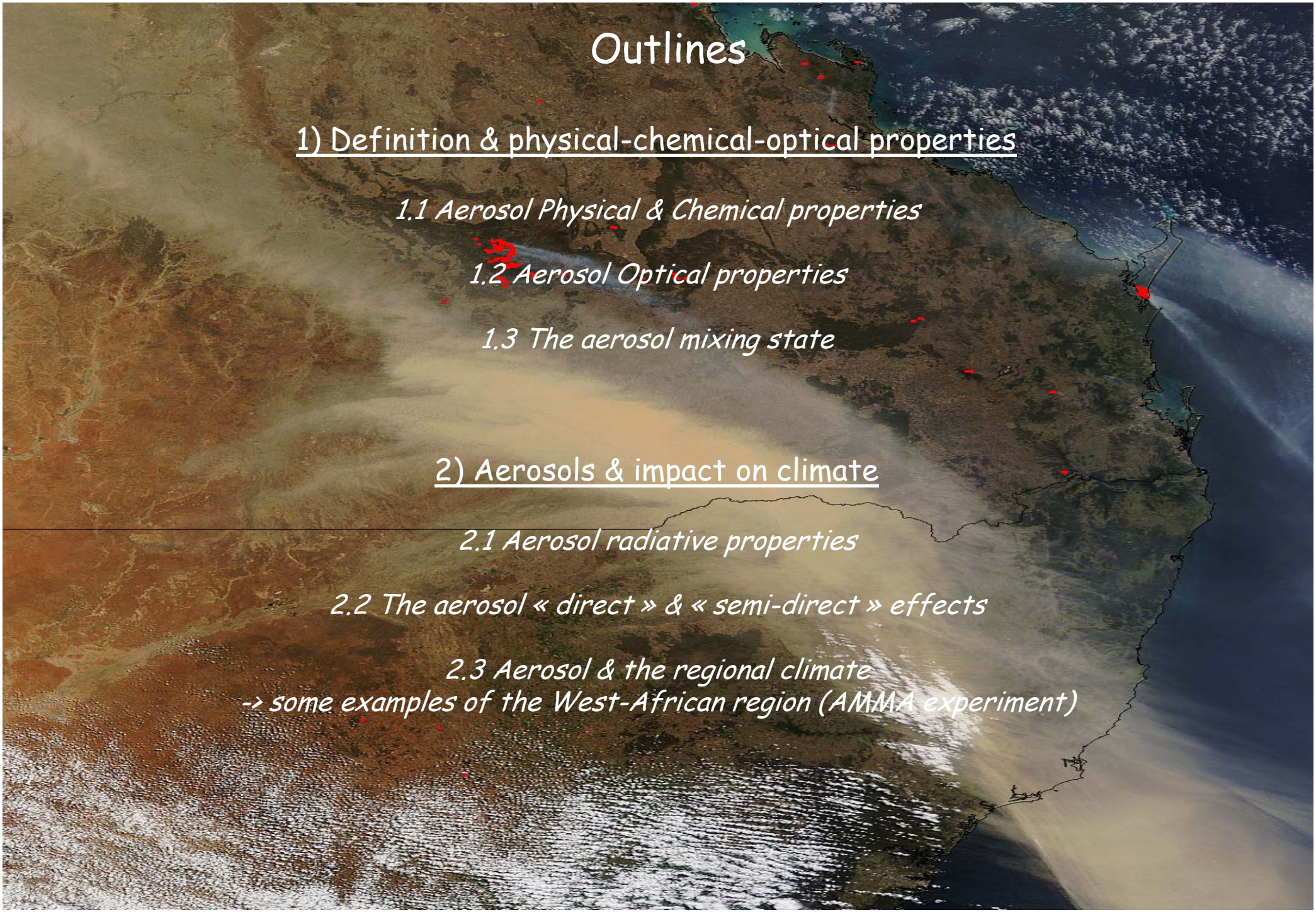
Dey, S., et al. (2008) Geophys. Res. Lett.

Composite SSA for 6 cases in :
(a) October 2005 (post-monsoon),
(b) December 2005 (winter),
(c) March 2006 (pre-monsoon),
along with AERONET-retrieved SSA using version 2 (V2) algorithm..

-> in the post-monsoon season : external mixing or water-soluble coating over dust,
-> in the winter season, the external mixing seems to be the most probable mixing state,
-> in the pre-monsoon season, BC coating over dust,

Core/shell mixing over Europe during the heatwave event in 2003





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
-> some examples of the West-African region (AMMA experiment)

2.1) Aerosols radiative properties

Key radiative parameters of the RTE

$$\Delta F \approx -DS_0 T_{at}^2 (1-A_c)(1-R_s)^2 \tilde{\omega}_0 \overline{\beta} \delta \left[1 - \frac{2R_s}{(1-R_s)^2} \left(\frac{1-\tilde{\omega}_0}{\tilde{\omega}_0 \overline{\beta}} \right) \right]$$

Geophysical variables — ***Aerosol microphysics (except Rs)***

« 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

ΔF	average aerosol forcing at **top of atmosphere** (TOA)
δ	aerosol optical depth

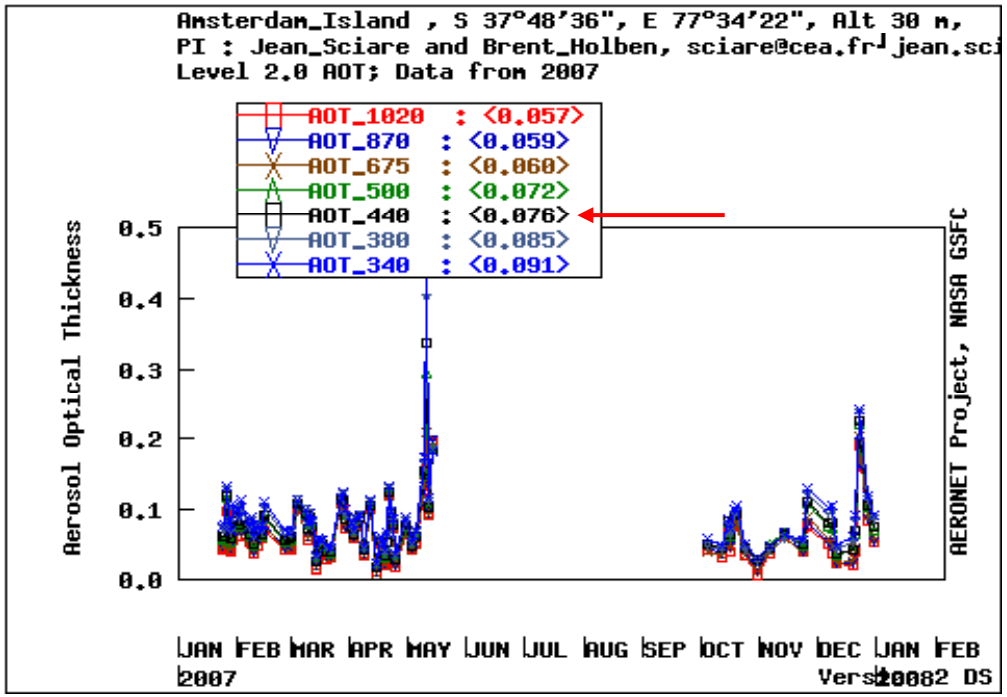
$\tilde{\omega}_0$	aerosol single-scattering albedo
$\overline{\beta}$	average aerosol up-scatter fraction

Aerosol Optical Depth

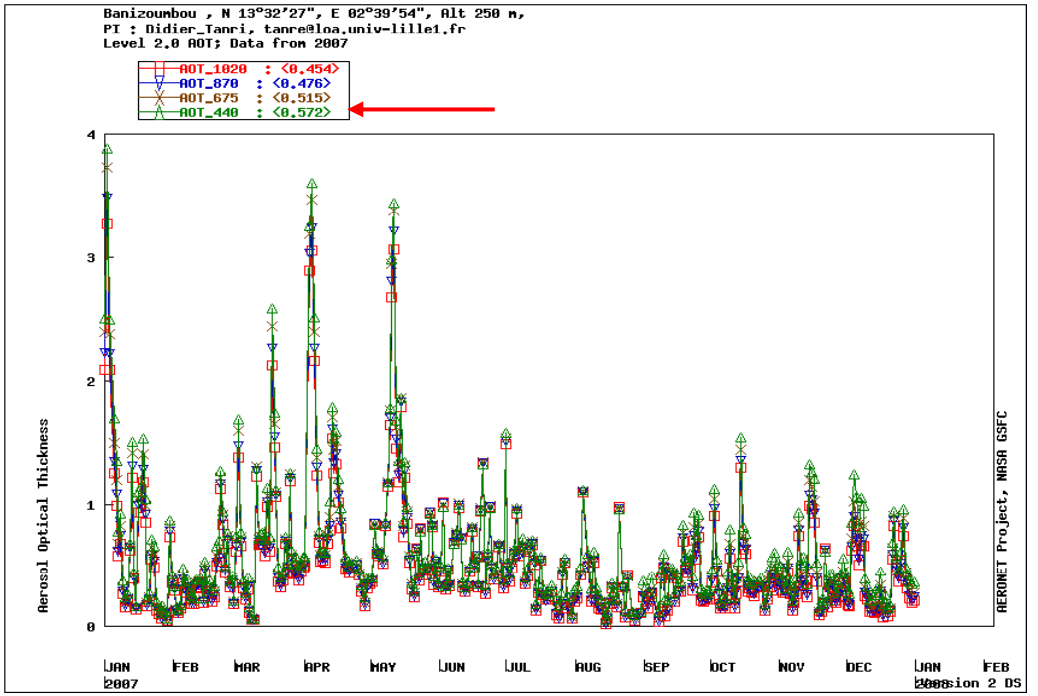
➢ $\delta_a(\lambda)$: extinction of radiations over the whole atmospheric column

$$\delta_a(\lambda) = \int_0^{TOA} K_{ext}(\lambda, \alpha, m)dz \quad (0.05 < \delta_a < 4 \text{ à } 550 \text{ nm})$$

Amsterdam Island
clear atmosphere



Banizoumbou (Niger)
Mineral dust



Aerosol Single Scattering Albedo (SSA)

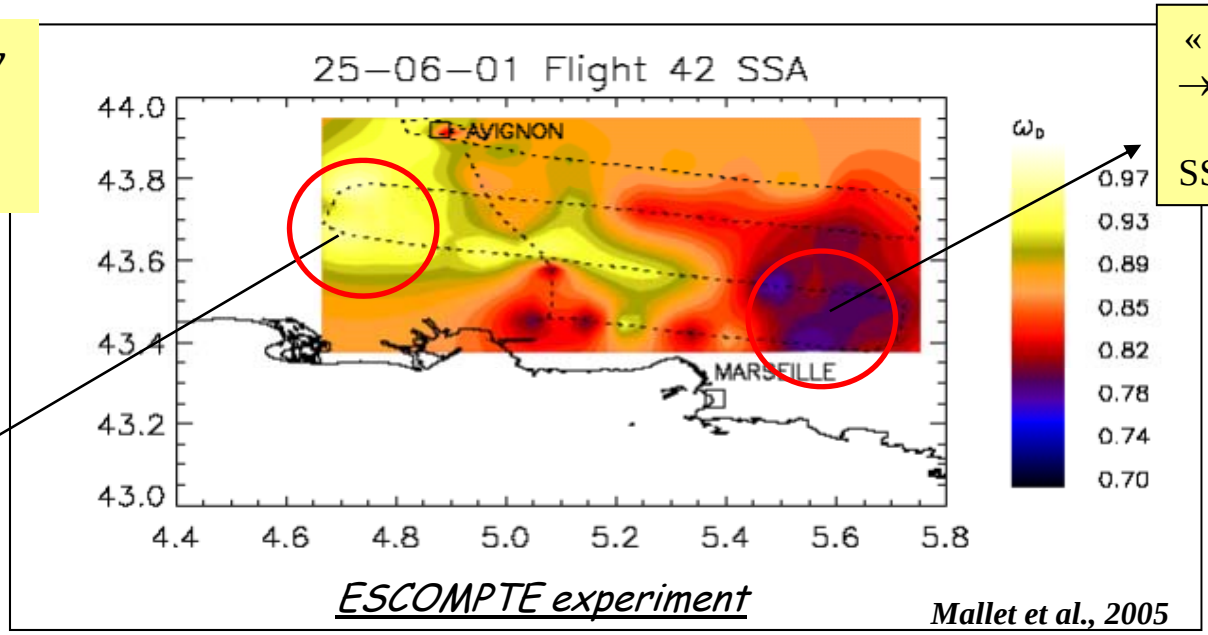
➢ $\omega_o(\lambda)$: ratio of scattering to extinction

$$\omega_o(\lambda) = K_{scat.}(\lambda) / (K_{scat.}(\lambda) + K_{abs}(\lambda))$$

$$(0 < \omega_o(\lambda) < 1)$$

⇒ Black carbon : $\omega_{o,550} = 0.26 - 0.40$
⇒ Sulfate & sea salt : $\omega_{o,550} = 0.99$

*Large SSA **variability** depending on the aerosol types !*



« Urban » atmosphere
→ large concentration of BC particles
SSA ~ 0.80 at 0.50 µm

« Industrial » atmosphere
→ large concentration of sulfates scattering particles
SSA ~ 0.95 at 0.50 µm

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*

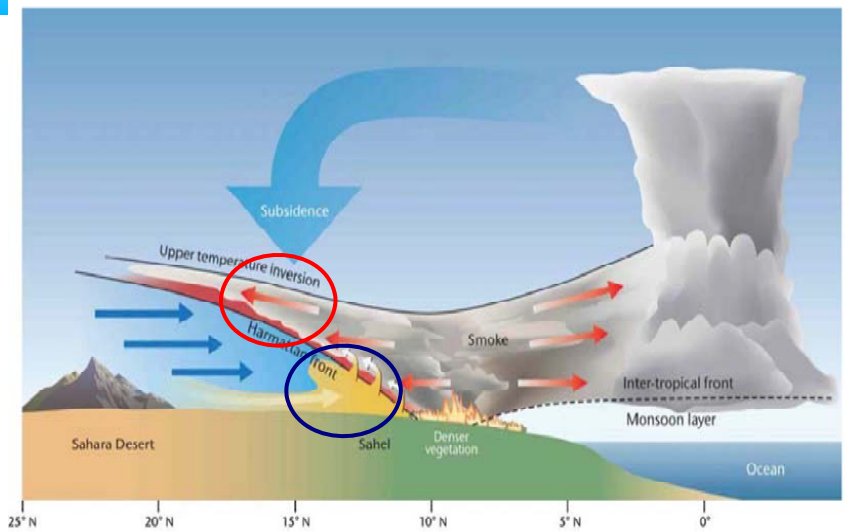
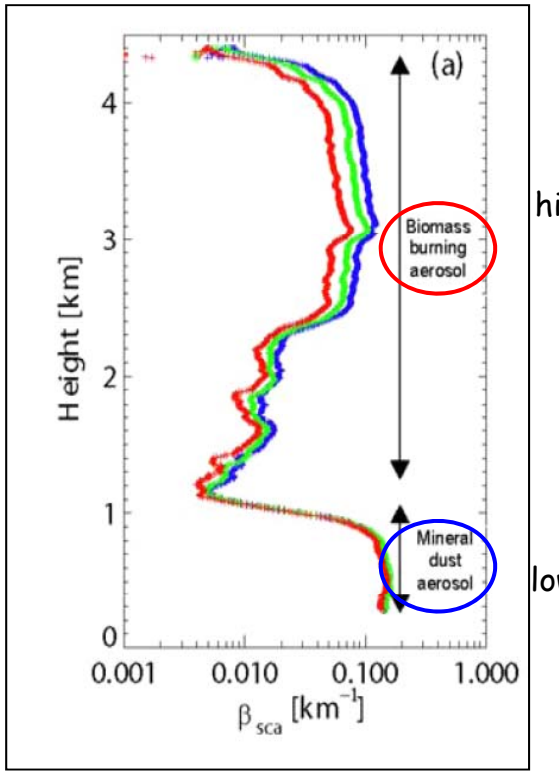


Figure 12. Schematic zonal cross section showing the northward transport of biomass burning aerosol in warm, ascending air (red arrows) and the westward/southward transport of mineral dust in a cooler airflow channeled by the mountainous regions to the north (blue arrows). The "Harmattan front" is shown 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.



high Angs. Expon.

low Angs. Expon.

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)…

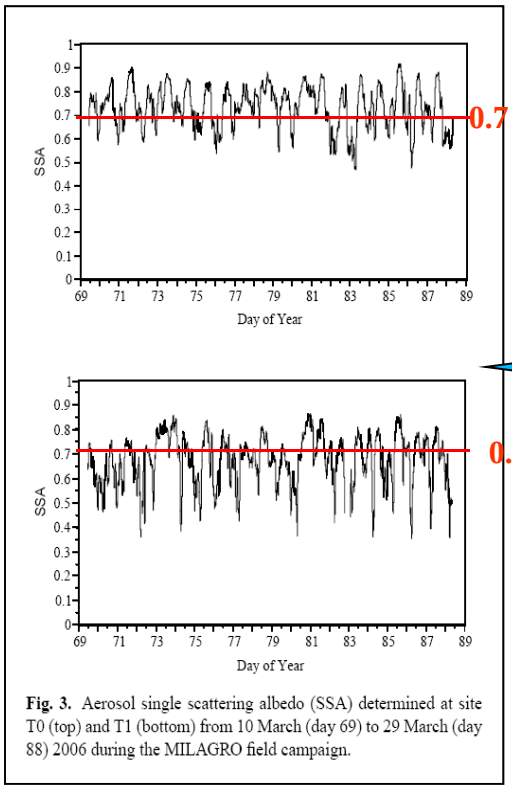
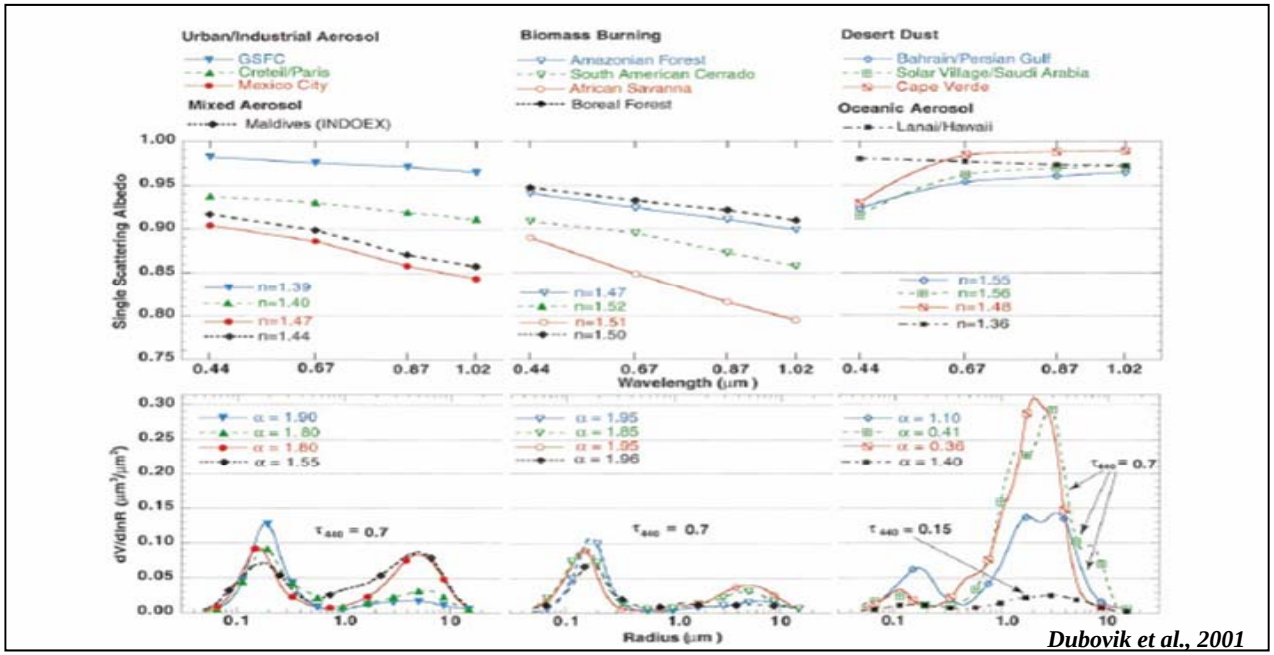


Fig. 3. Aerosol single scattering albedo (SSA) determined at site T0 (top) and T1 (bottom) from 10 March (day 69) to 29 March (day 88) 2006 during the MILAGRO field campaign.

Country	Cities	SSA (visible λ)	References
India	Bangalore Delhi Kanpur New Delhi Goa	0.73 0.67 0.76 0.60-0.80 0.83-0.93	Babu et al. 2002 Singh et al. 2005 Tripatu et al. 2005 Ganguly et al., 2006 Randdriamiarisoa et al. (2004)
Mexico	Mexico	0.47-0.92	Marley et al. (2009)
Europe	Marseille Paris Toulouse	0.85 0.85-0.92 0.70	Mallet et al. (2003) Chazette et al. (2005) Gomes et al. (2009)

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



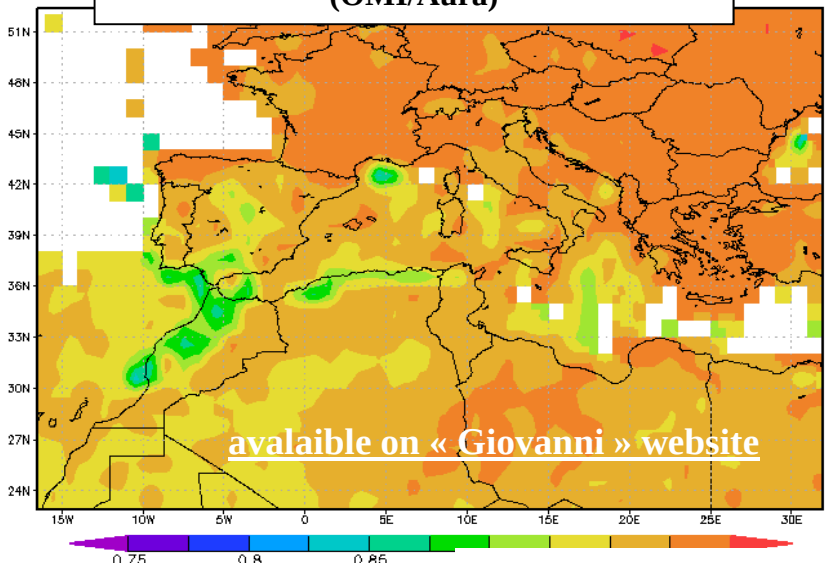
Large variability of urban/Ind and Biom. Burn. SSA at 440 nm,

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

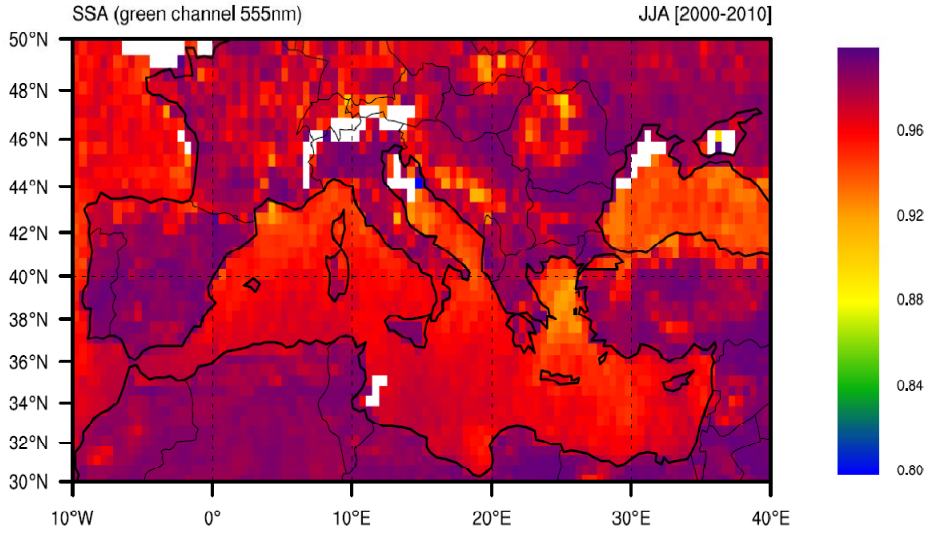
Oceanic aerosols SSA ~ 1.00 for the whole λ : purely scattering species

Some examples of recent SSA aerosol properties retrieved from space…

Aerosol Single Scattering Albedo SSA (OMI/Aura)

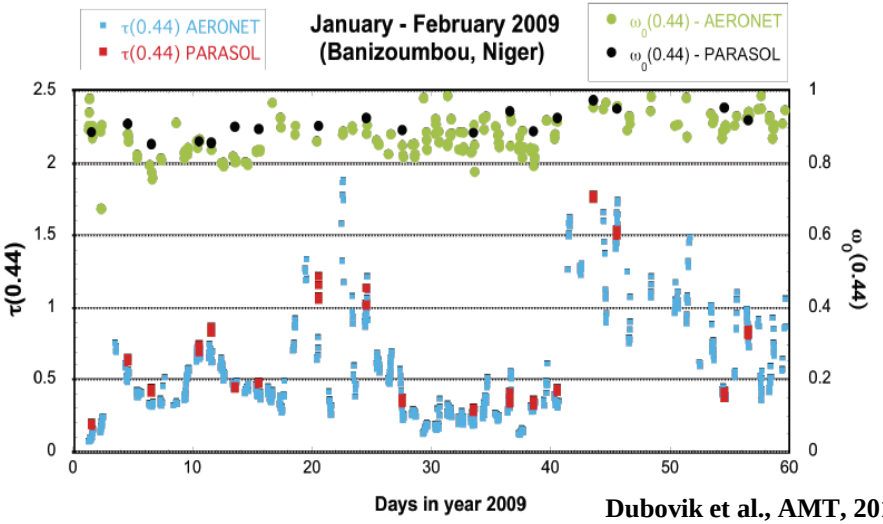


avalaible on « Giovanni » website



Kahn et al., JGR, 2010

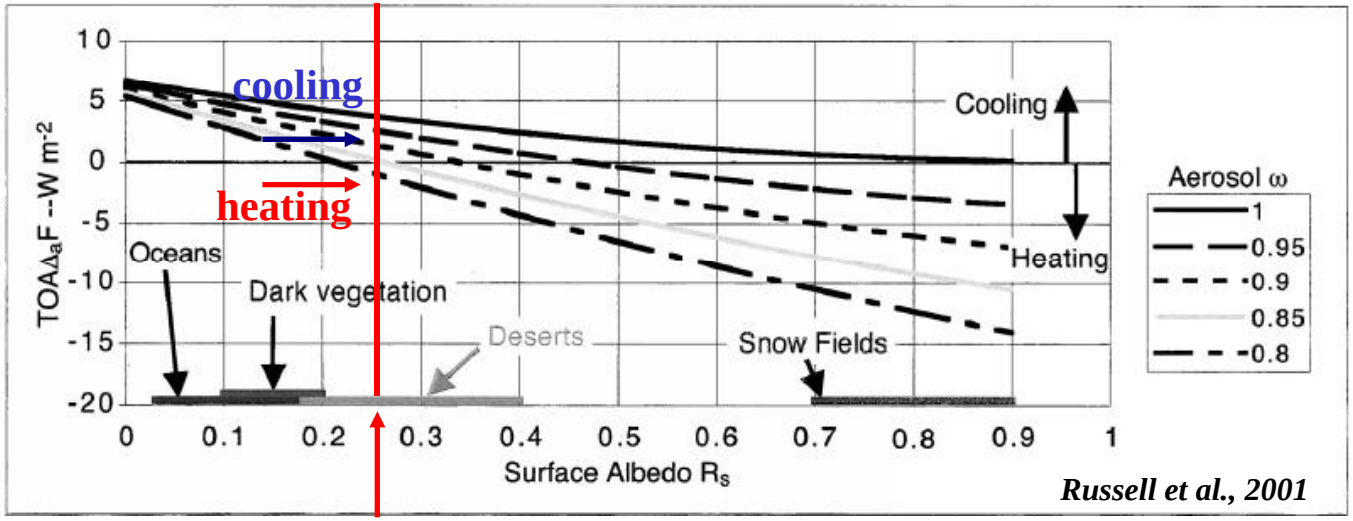
Very useful for comparisons with RCM simulations !



Dubovik et al., AMT, 2011 (see O. Dubovik presentation)

Cooling versus Heating of an aerosol layer

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



Russell et al., 2001

Above a critical surface albedo (R_s ~ 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

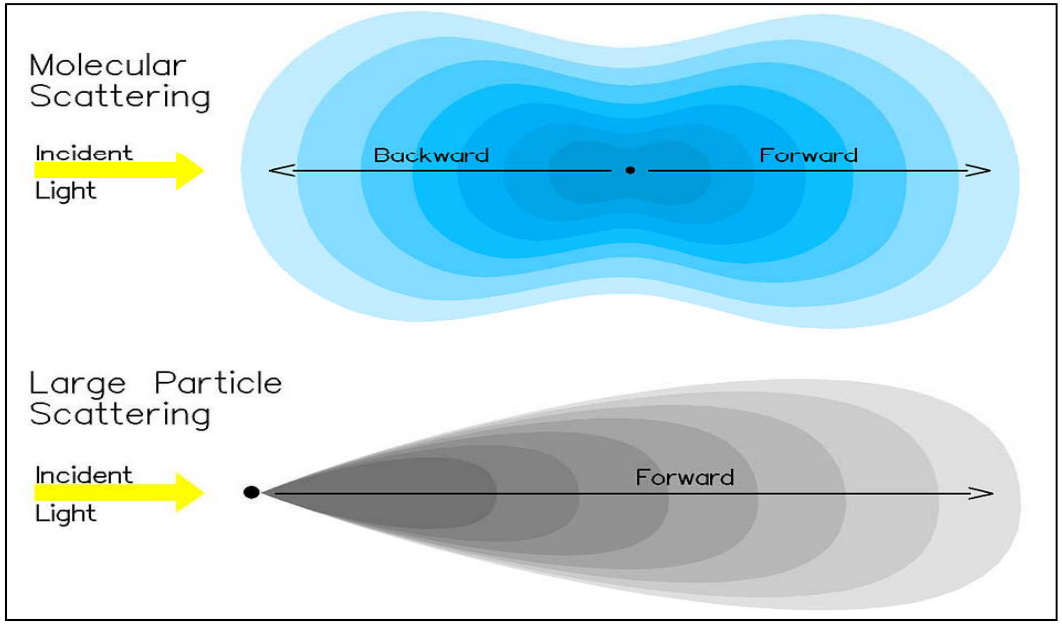
Aerosol asymmetry parameter (g)

- g (λ) : defined as the intensity-weighted average of the cosine of the scattering angle:

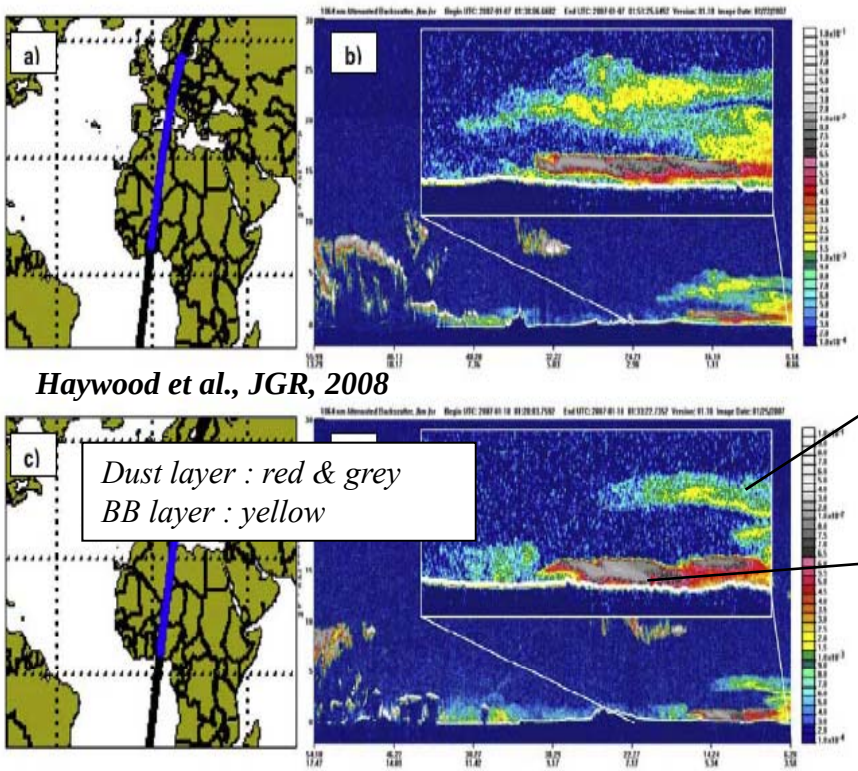
$$g(\lambda) = \frac{1}{2}\int_0^{\pi} \cos\theta P(\lambda,\theta)\sin(\theta)d\theta$$

$(-1 < g(\lambda) < 1)$

⇒ g = 0 (Rayleigh scattering)
⇒ Sulfates : g_{550} = 0.60
⇒ Sea Salt & dust aerosols : g_{550} = 0.77 (more forward scattering)



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



Haywood et al., JGR, 2008

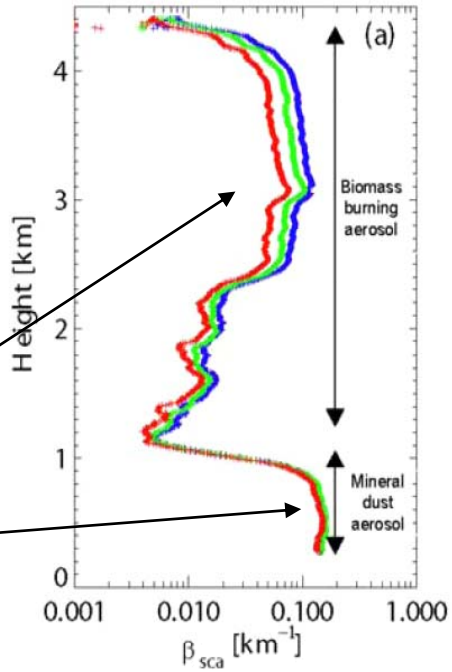


Figure 10. Aerosol backscatter from the CALIPSO lidar (1032 nm) during transects across N Africa: (a, b) 7 January 2007 and (c, d) 18 January 2007. The highlighted boxes in Figures 10b and 10d show close-ups of the area from approximately 22°N to 5°N. The dust layers are shown by red and grey (backscatter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

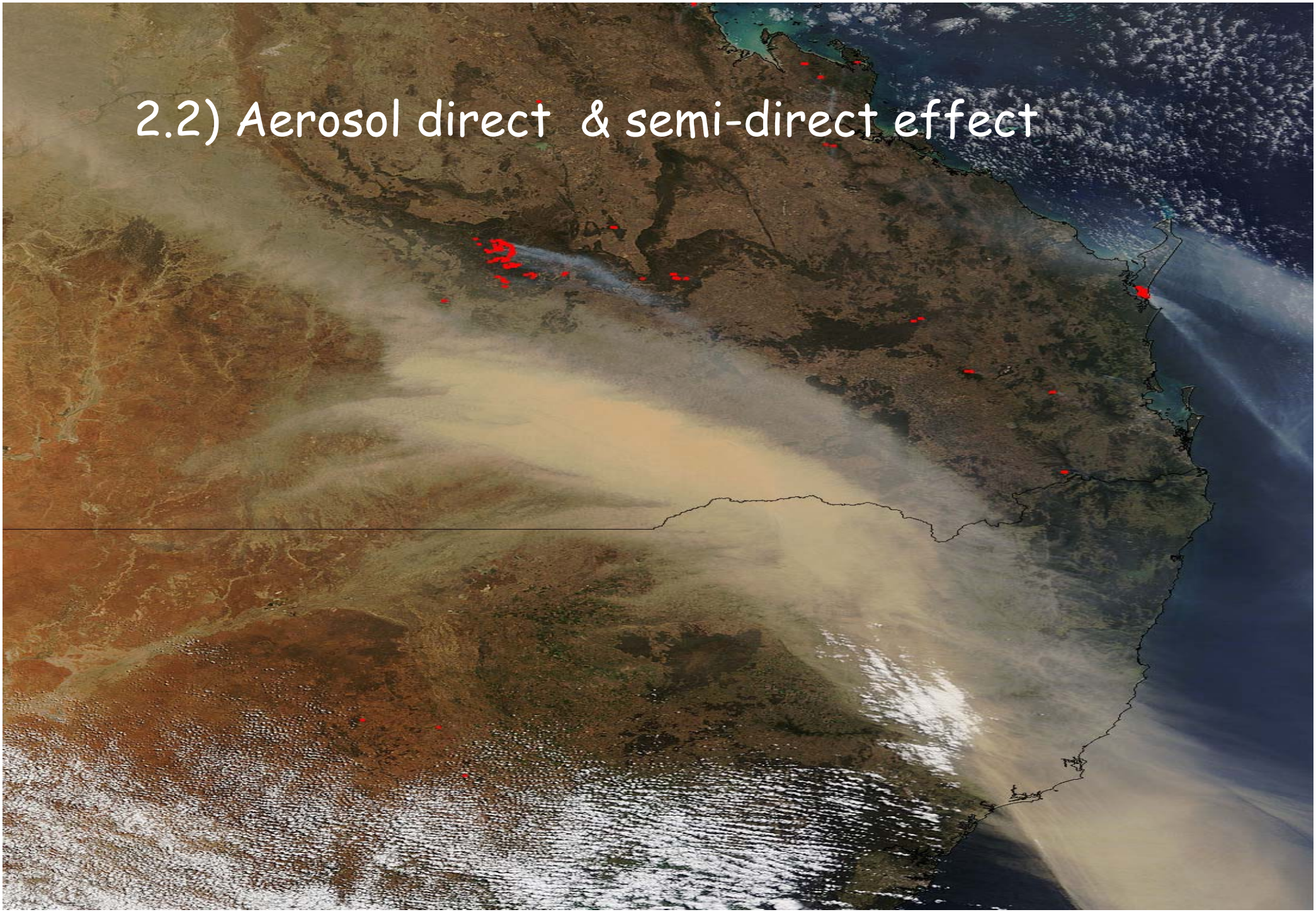
SW RT calculations show that the radiative effect of biomass burning aerosol was sensitive to the vertical distribution of aerosol.

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****.*

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

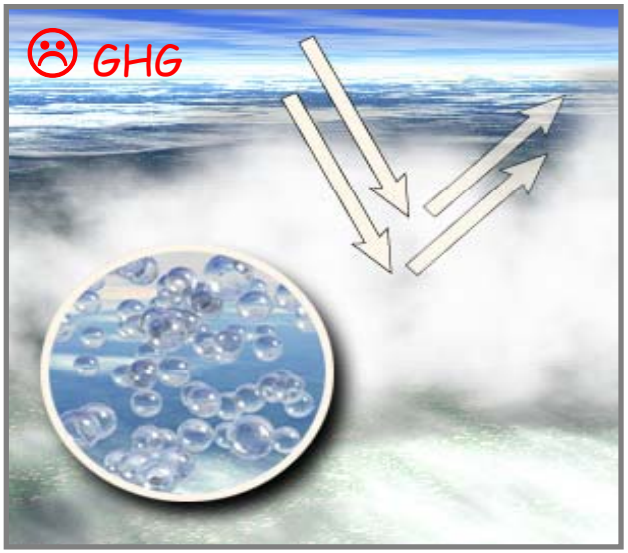
2.2) Aerosol direct & semi-direct effect



Aerosol direct effect

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

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



IPCC, 2007

Scattering Aerosol
("whitehouse" aerosol)
Sulfates ~ -0.4 W m $^{-2}$
Organic Carbon ~ -0.05 W m $^{-2}$

Absorbing particle ("greenhouse" aerosol)

IPCC, 2007 BC ~ + 0.20 W m $^{-2}$

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)**

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.

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}

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

Foster et al., 2007

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} »*

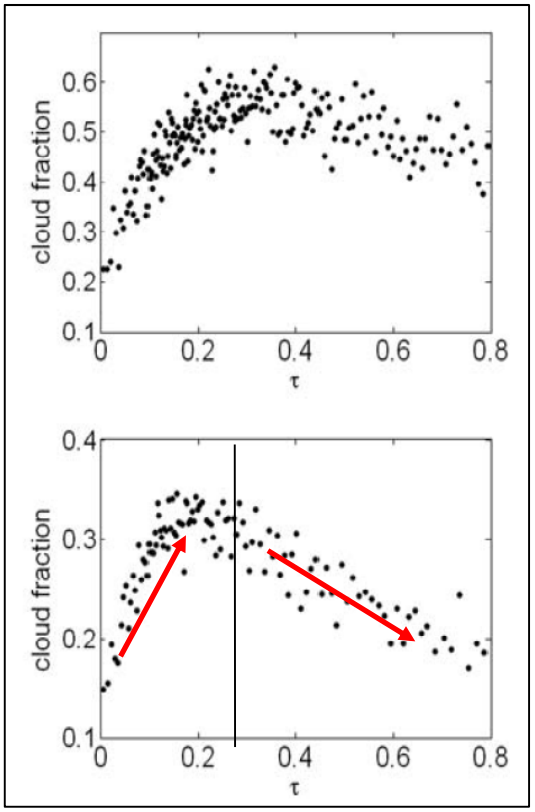
*« The mean and median of the direct RF for **fossil fuel BC** from grouping all these studies together are **+0.19 and +0.16 W m^{-2},** respectively, with a standard deviation of nearly 0.10 W m^{-2} »*

Foster et al., 2007

Aerosol semi-direct effect

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

Absorbing aerosols *are thought to warm the layer in which they are located*
→ decreased relative humidity → less cloud cover *≠ indirect effect,*



Aerosol semi-direct effect from satellite observations

Smoke Invigoration Versus Inhibition of Clouds over the Amazon

Ilan Koren,[1] J. Vanderlei Martins,[2,3] Lorraine A. Remer,[3] Hila Afargan[1]

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.

Science, 2008

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

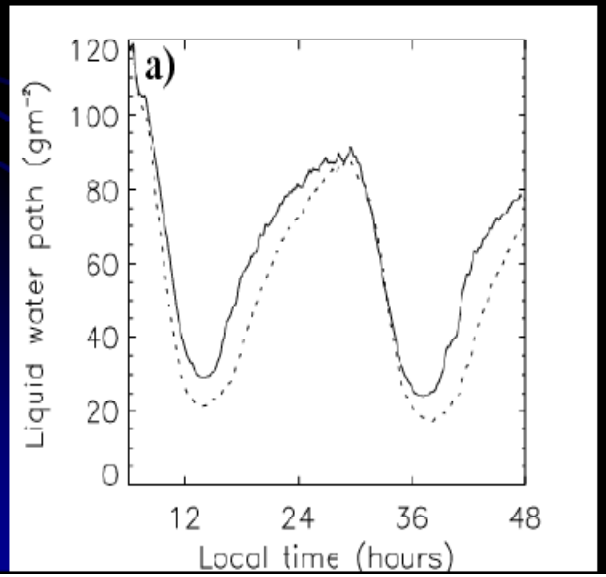
***LES** was used to investigate the SDE for marine stratocumulus & examine the dependency on the vertical distribution (**indirect effect is excluded**)*

Positive SDE

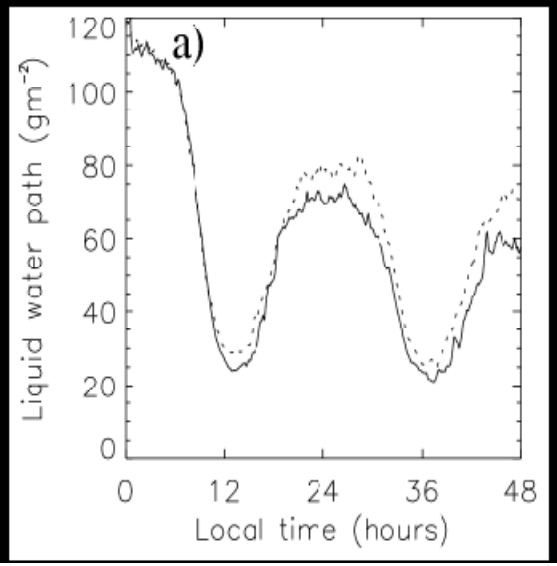
Negative SDE

Johnson et al. (2004) stratocumulus

Absorbing aerosol **in** well-mixed BL:
Reduction in LWP



Absorbing aerosol **above** inversion
Increase in LWP



Johnson,, et al. 2004

Recent work using GCModels indicates a more complex interaction

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

Jan Perlwitz[1,2] and Ron L. Miller[1,2]

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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 → larger diabatic heating and increased warming → decreasing RH,

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

The net effect is more low cloud cover with increasing aerosol absorption

The semi-direct effect remains a challenge !

A review from Koch and Del Genio., 2011 gives a review of the effect of BC aerosols and different cloud types

Koch et Del Genio, ACP, 2010

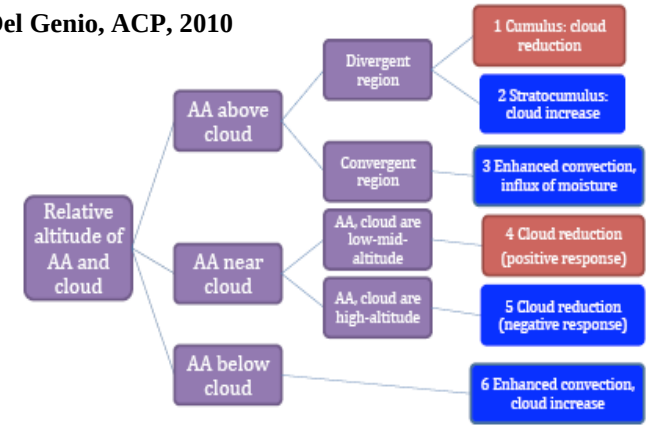
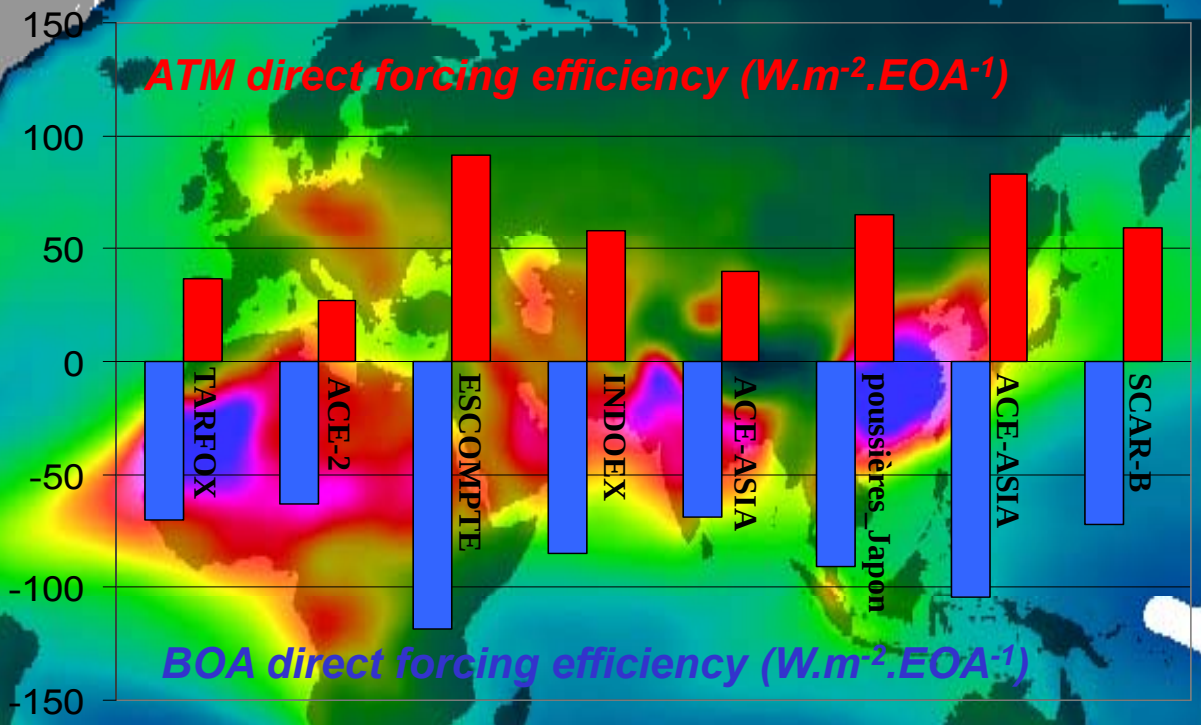


Fig. 1. Suggested framework to organize aerosol absorption effects on cloud cover. Red and blue indicate positive and negative semi-direct effects.

2.3) Aerosol & the regional scale

-> Example over the WA region (AMMA experiment)



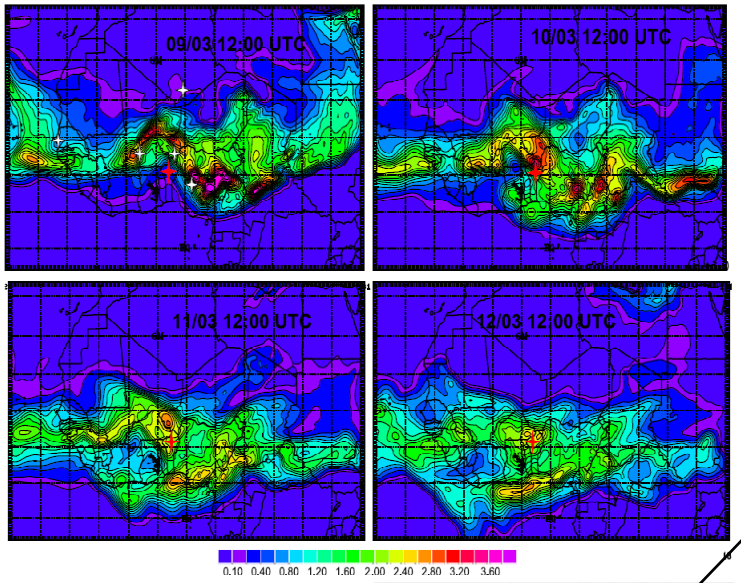
Due to the direct radiative forcing at the regional scale, more and more studies are now focused on the effect of particles on the regional climate

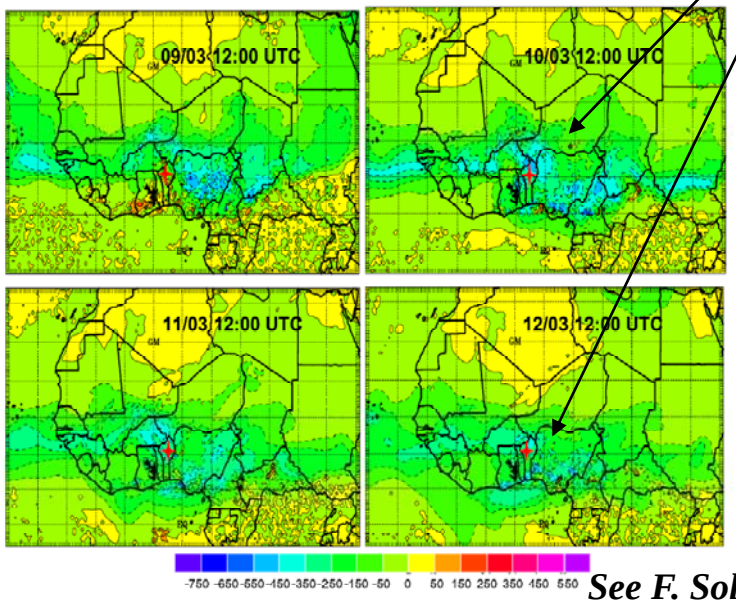
*In most of cases, the **regional DRF** due to aerosols is clearly larger than GHG !*

*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



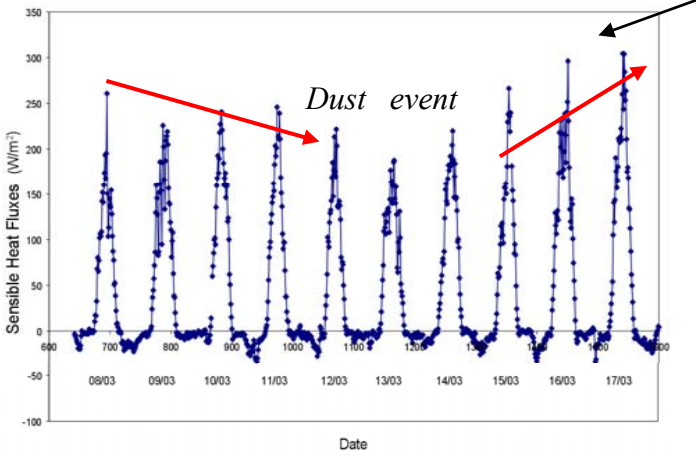


*Simulations conducted with the meso-scale model (MEsoNh) : 3*3 km resolution, duration ~ 1 week*

Large simulated Dust Optical depth during this specific event, with values reaching up to 3 (at 0.55 µm)

Large aerosol « dimming » at the surface

Compensated by a decrease in surface T° & sensible fluxes



Cooling at the surface and heating of the atmospher is able to modify the atmospheric dynamic and hydrological cycle over WA

See F. Solmon presentation for long term climatic simulations…

Mallet el al., ACP, 2009
Tulet, Mallet et al., JGR, 2008


Thank you for your attention !