



**aerosols and climate :
uncertainties
and
the need for standardization**

Frank Raes, Jean-Philippe Putaud, Annette Borowiak



From the letter of invitation:

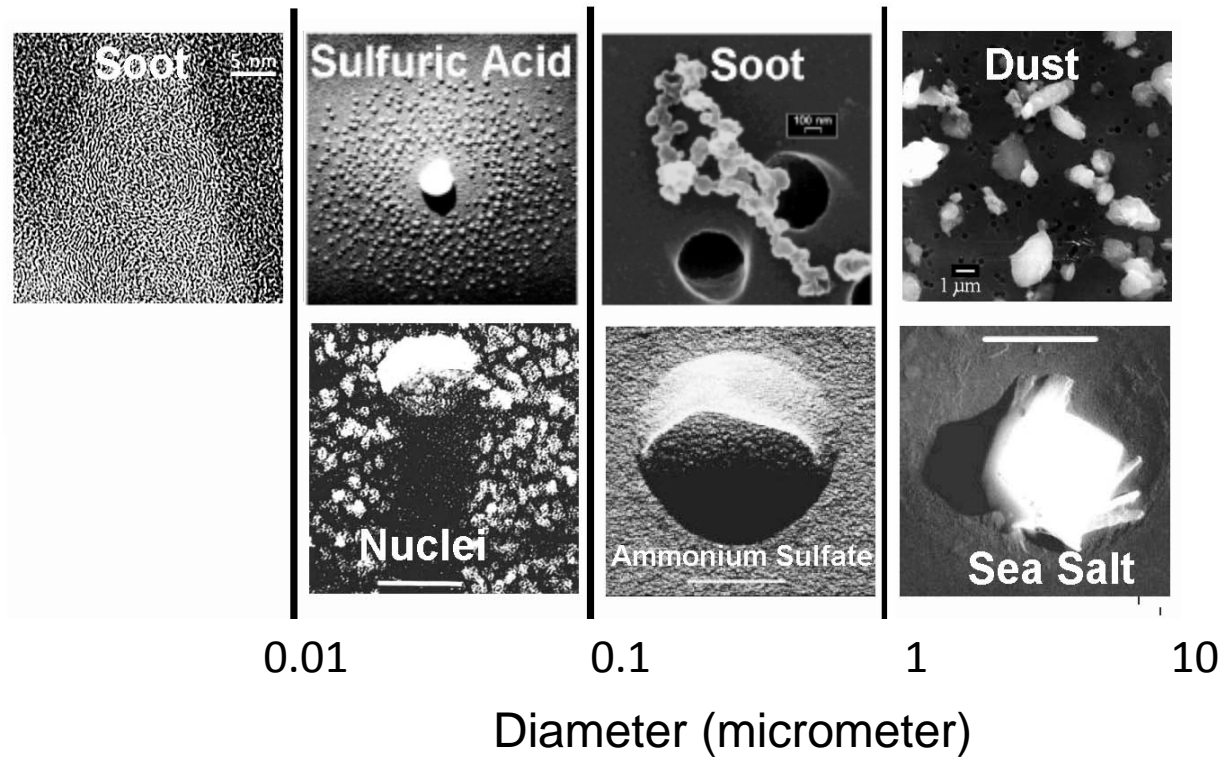
- Set the stage for Workshop that aims at developing a cross academic, industry, public stakeholder and government outlook on key aerosol metrology issues critical to furthering our understanding of aerosols in the climate *theatre*



- We are more aware of the importance of a range of aerosol characteristics and processes in the climate system.
- Quantifying the climate impact of changing the concentration of individual chemical components of the atmospheric aerosol is still very uncertain.
- Quantifying the climate impact of individual human activities , incl. policy measures, is even more uncertain (emissions & their chemical fingerprint are uncertain), but the sign is usually known.
- Deal with uncertainty by adopting a multi-pollutant /multi-effect approach (not just aerosol emissions, not just climate impacts)

- Standardization doesn't reduce uncertainty of measurements or assessments but improves their comparability.
- Policy making and policy implementation *is* about comparing (e.g. comparing climate effects with and without a policy, comparing PM values with target /limit values)
- Policies often affect markets, or are themselves market based: standardization contributes to creating a level playing field. (BC emissions tradeable like CO2 emissions?)

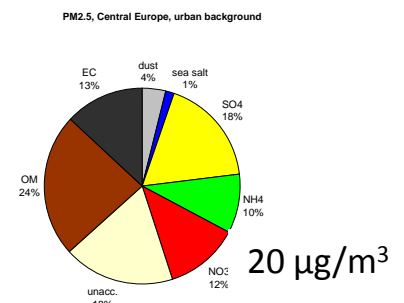
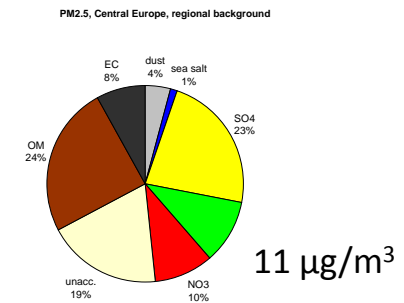
size, chemical composition, morphology



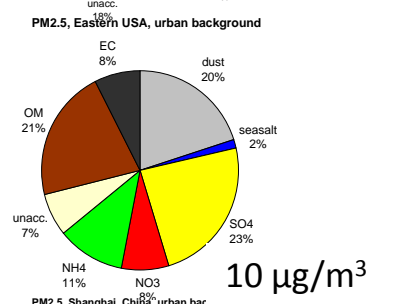
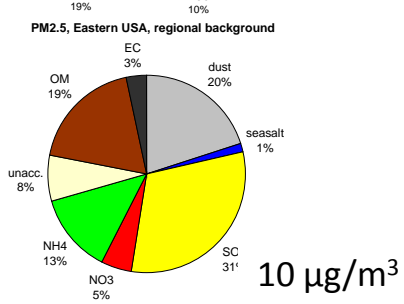
chemical composition of PM2.5, worldwide (2000 – 2010)

Joint Research Centre

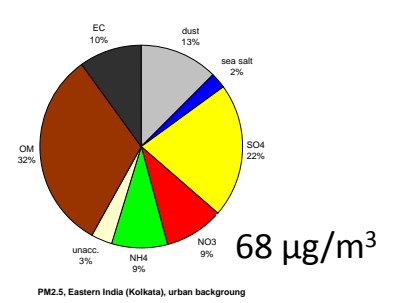
Central Europe



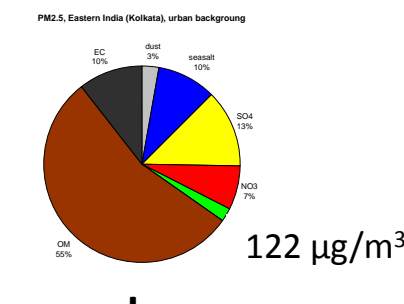
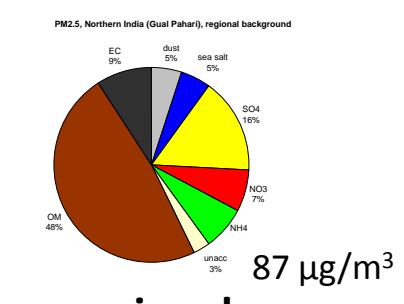
Eastern USA



China Shanghai



Northern India



- Elemental carbon
- Organic Matter
- Unaccounted
- Ammonium
- Nitrate
- Sulfate
- Seasalt
- Dust

regional background

urban background

Source, J.P. Putaud, JRC

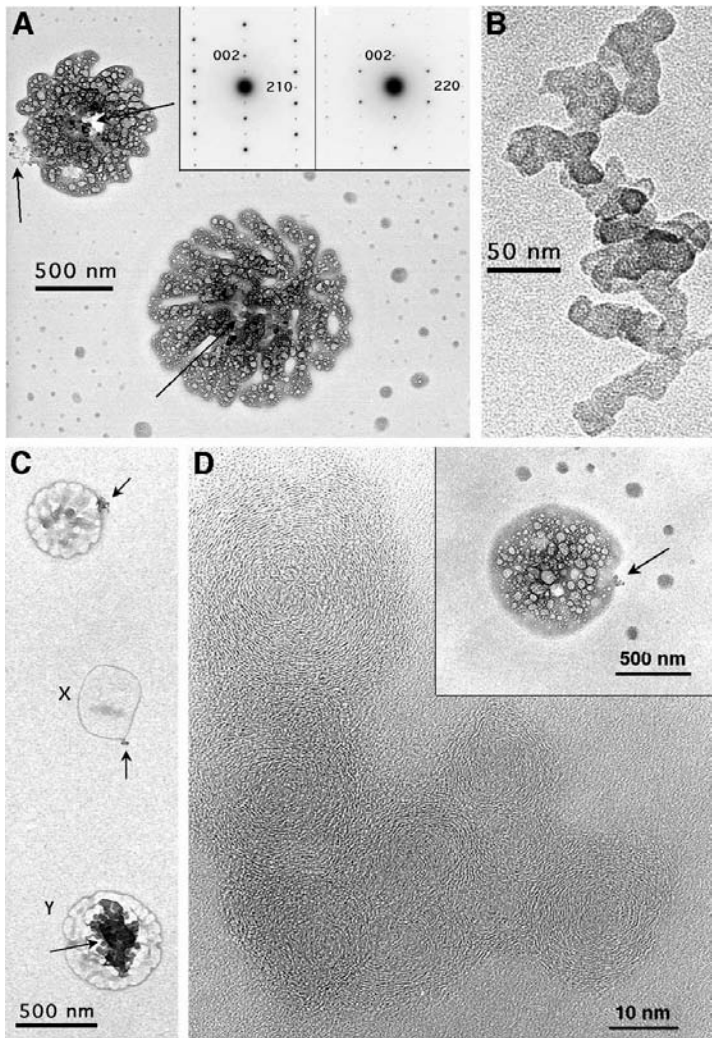


Figure 3. Aerosol particles collected above the Southern Ocean, south of Tasmania. (a) Ammonium sulfate particles with soot inclusions (marked by arrows). The inset displays two different zone-axis SAED patterns that were obtained from a similar particle; spacings are consistent with the structure of ammonium sulfate and are indexed accordingly. (b) A typical, individual soot particle that is not enclosed by sulfate. (c) Residues and soot inclusions (arrowed) that remained after three ammonium sulfate particles were intentionally sublimed with the electron beam. (d) An ammonium sulfate particle with a small soot inclusion (arrowed). Bubbles that resulted from electron-beam damage are visible in the particles in Figures 3a and 3d. The high-resolution image shows the onion-like structure of spherules within the arrowed soot inclusion.

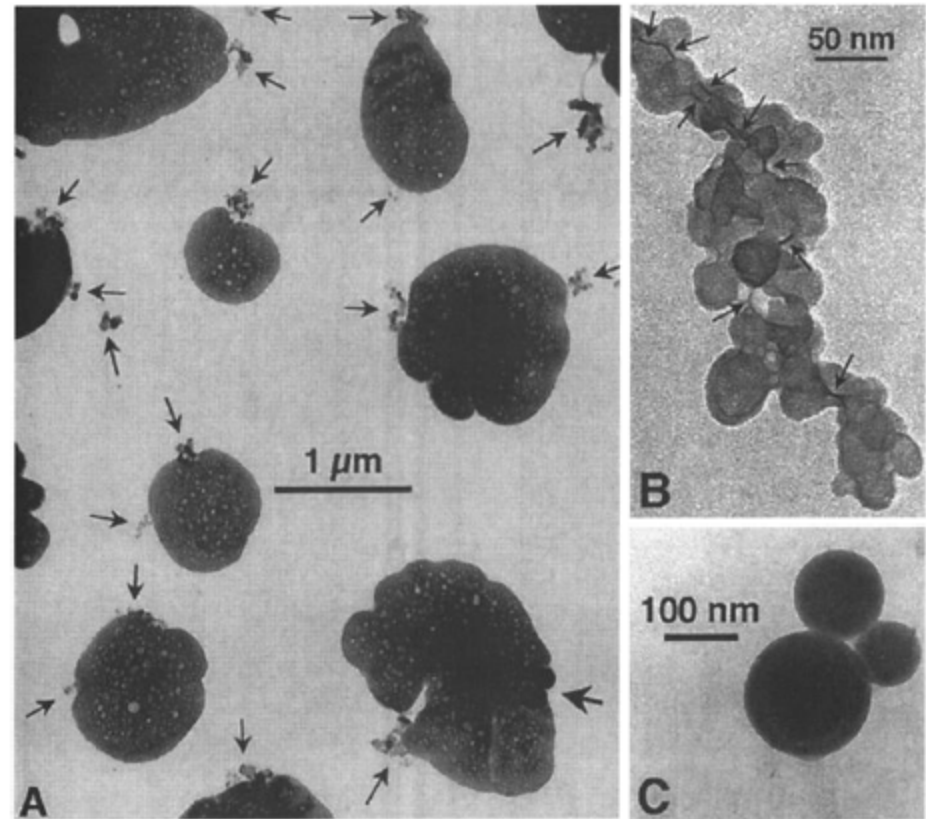
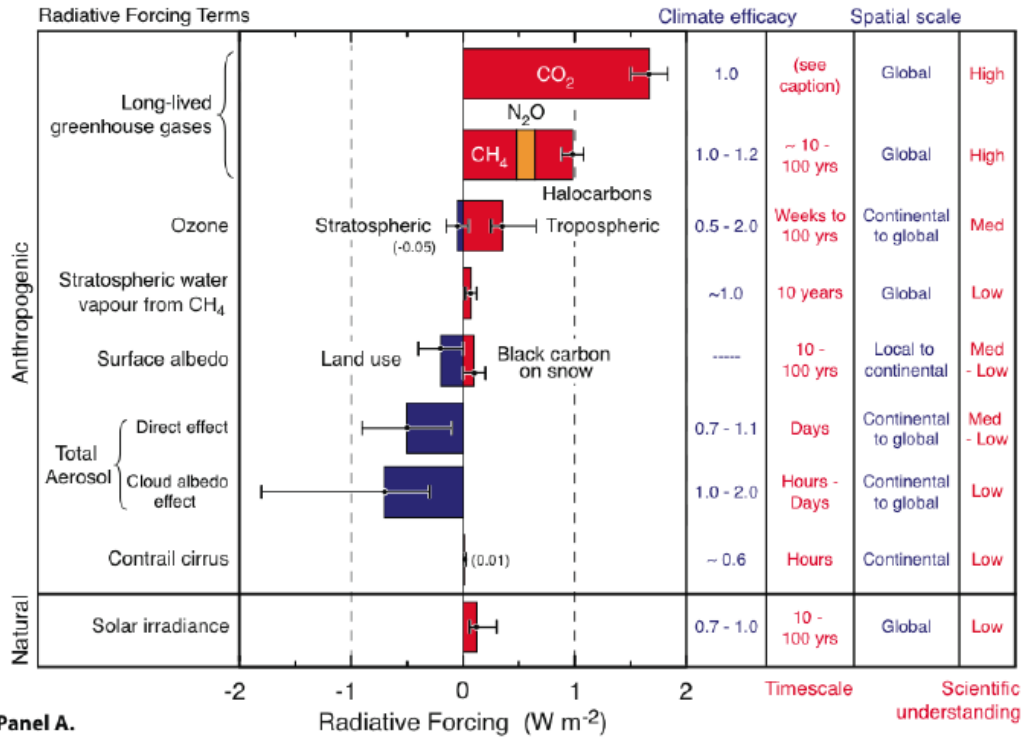
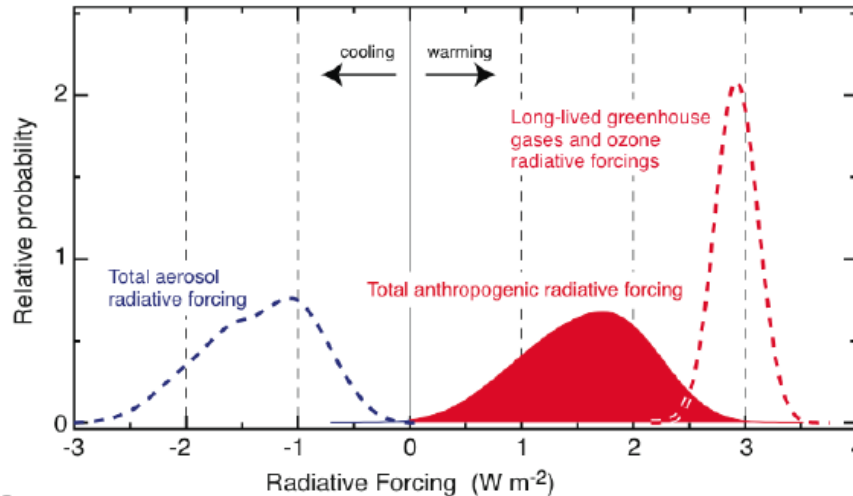


Figure 1. TEM images of aerosol particles collected in the polluted MBL of the North Atlantic Ocean, near the Azores Islands. (a) Ammonium sulfate particles containing soot (marked by the small arrows) and fly-ash spheres (marked by the bold arrow in the lower-right corner). (b) A typical chain-like soot aggregate; the arrows point to a carbon film that connects individual spherules within the aggregate. (c) Fly-ash spheres, consisting of amorphous silica. (d) High-resolution image showing disordered graphitic layers within a soot aggregate. The background of the image is a support film of amorphous carbon onto which the particles were collected. The inset in the upper-left corner is a copy of the boxed area; the graphitic layers are highlighted in the inset.

Radiative forcing of climate between 1750 and 2005



Panel A.



Panel B.



$$\Delta E_{aer,direct} = \frac{1}{2} F_T T^2 (1 - A_c) \left\{ \overline{\omega \beta} (1 - R_s)^2 - 2(1 - \overline{\omega}) R_s \right\} (\delta - \delta_{pre})$$

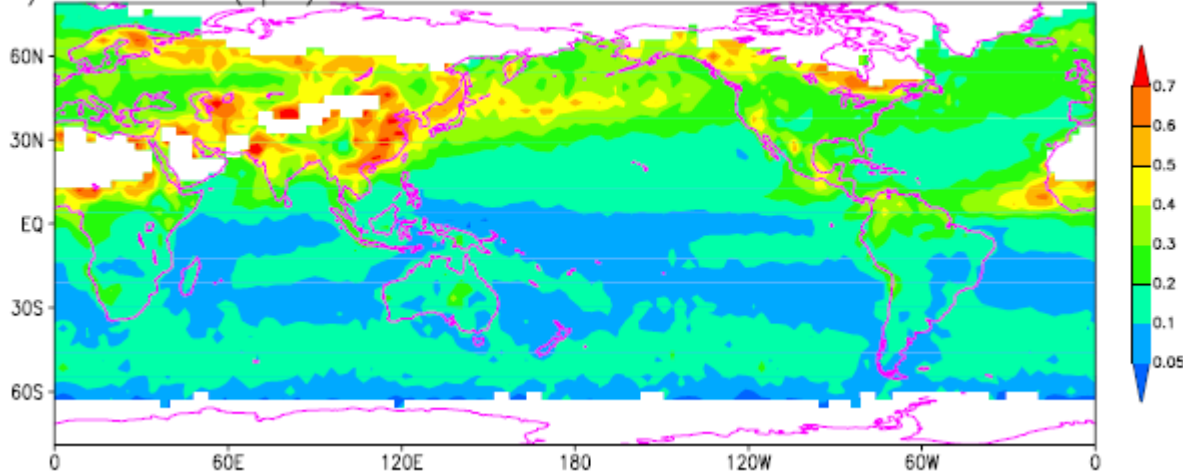
Labels and arrows in the diagram:
 - Solar constant points to F_T
 - Atmosphere transmissivity (w/o aerosols) points to T^2
 - Cloudiness points to $(1 - A_c)$
 - Backscatter fraction points to $\overline{\omega \beta}$
 - Surface Albedo points to R_s
 - Single Scattering Albedo points to $\overline{\omega}$
 - Aerosol Optical Depth points to δ

$$\delta_{pre} \approx \delta_{nat}$$

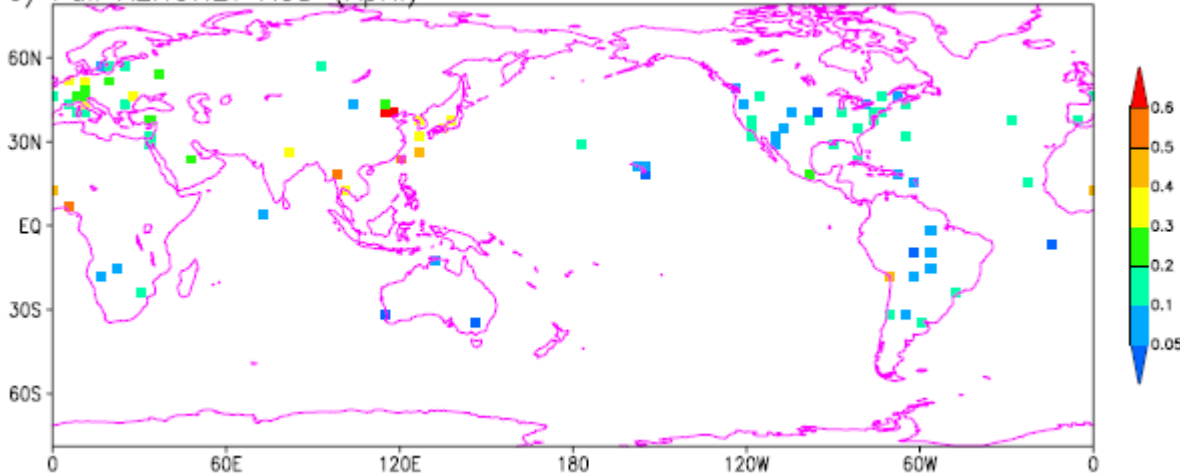
Haywood and Shine, GRL, 1995



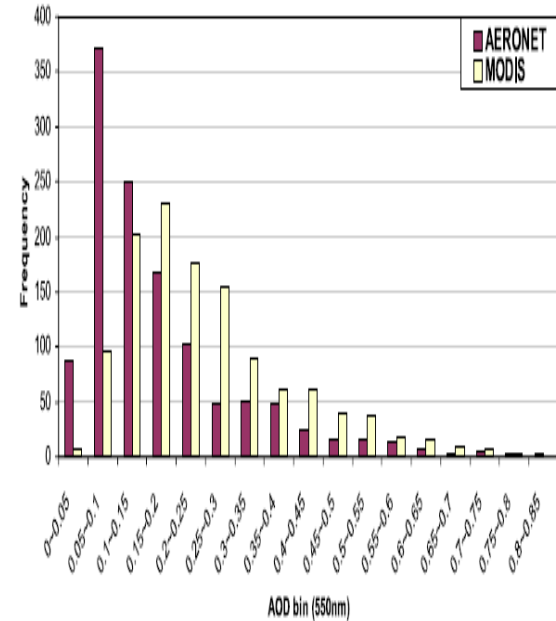
a) MODIS AOD (April)



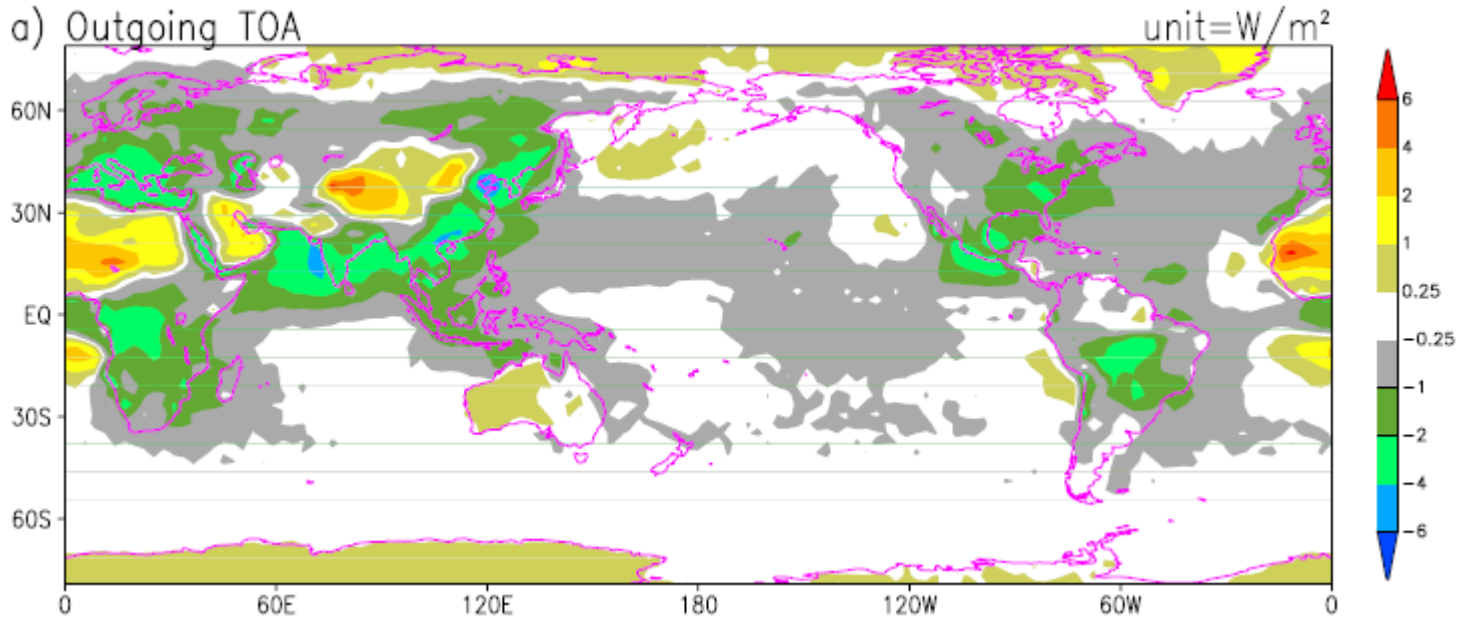
c) Full AERONET AOD (April)



AOD frequency



Chung et al., JGR, 2005

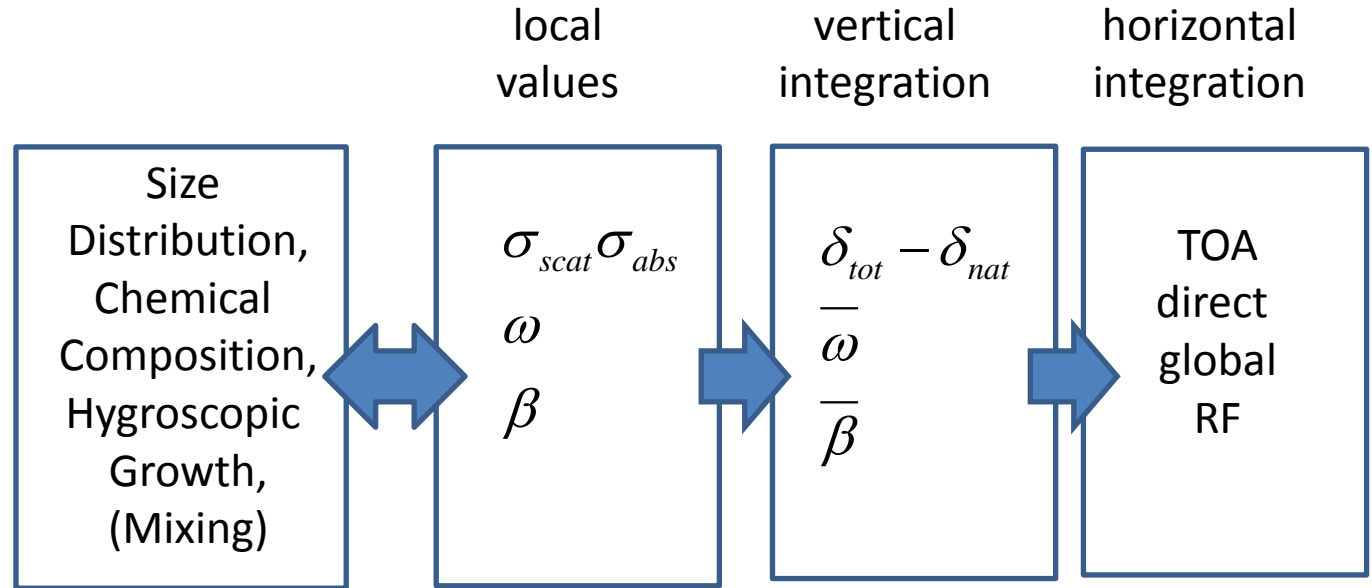


Annual average: - 0,35 Wm-2 (range: - 0,6 to – 0,1 Wm-2)

Uncertainty due to

- absorption properties of the aerosol (single scattering albedo)
- vertical distribution of the aerosol

Chung et al., JGR, 2005



closure experiments

$$\iint Q_{sca}(\lambda, D_p, m) \frac{\pi D_p^2}{4} n(D_p, m) dm dD_p = \sigma_{sp}(\lambda)$$



WORLD METEOROLOGICAL ORGANIZATION
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GAW No.

WMO/GAW Standard Operating Procedures for
In-Situ Measurements of Aerosol Mass
Concentration, Light Scattering
and Light Absorption

AEROSOL
GUIDE



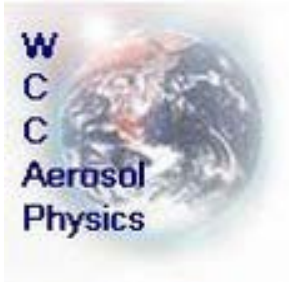
**World
Meteorological
Organization**
Weather • Climate • Water

WMO/TD-No.
..... 2010

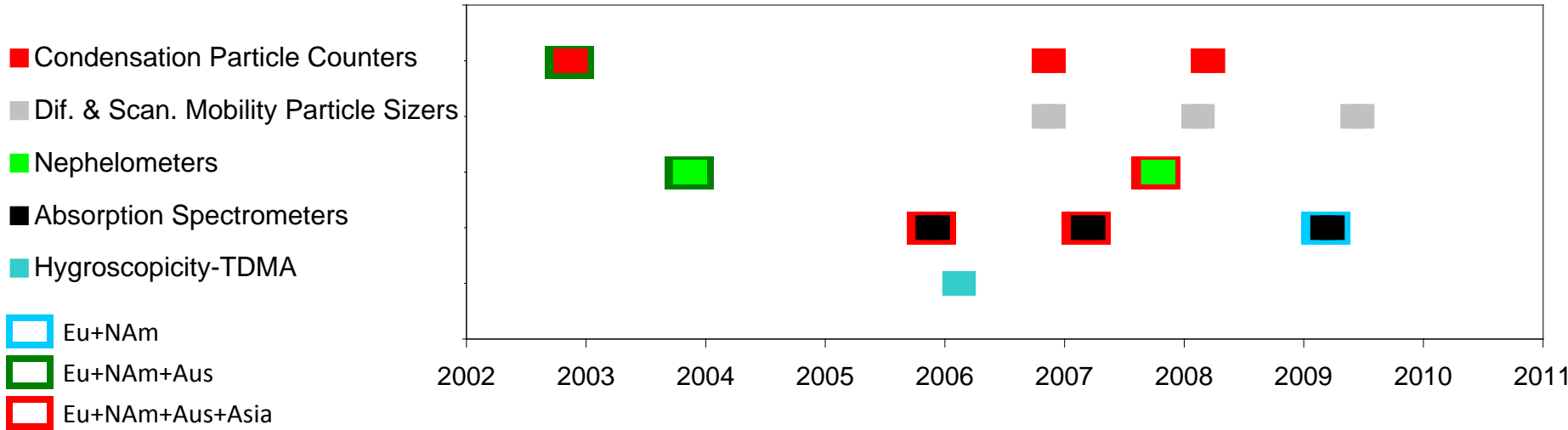
September 2003

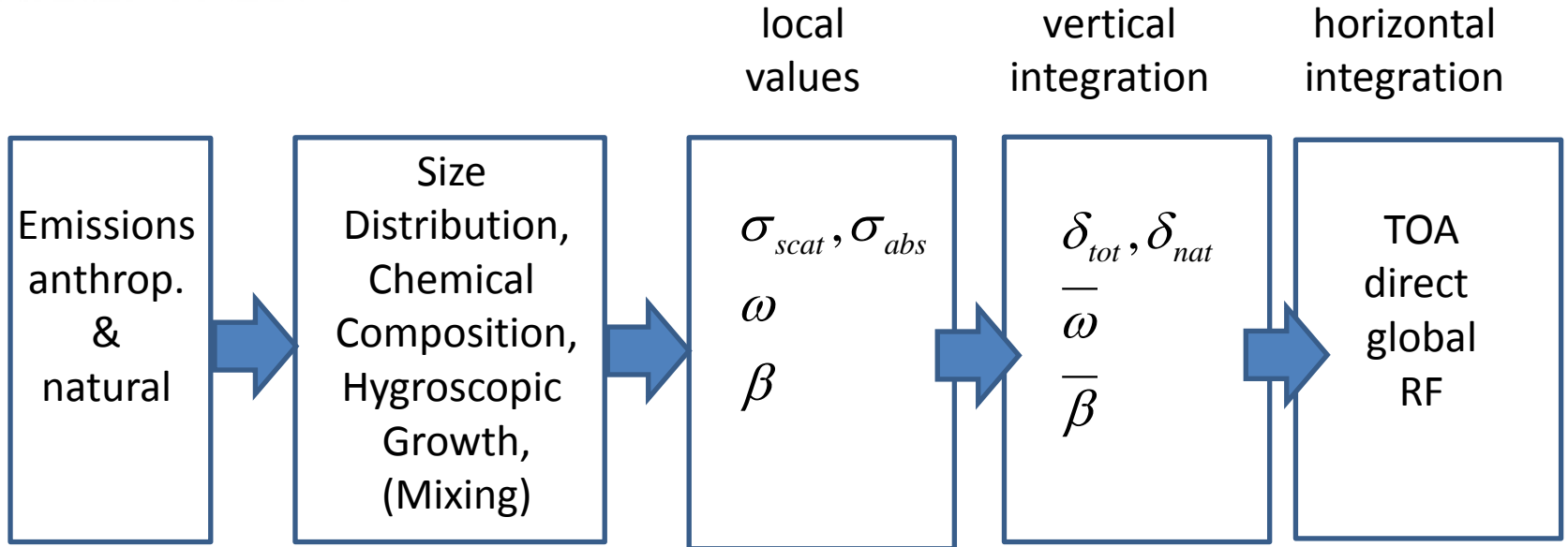
- The GAW – World Calibration Centre for Aerosol Physics

<http://gaw.tropos.de/WCCAP/>



- Intercomparisons





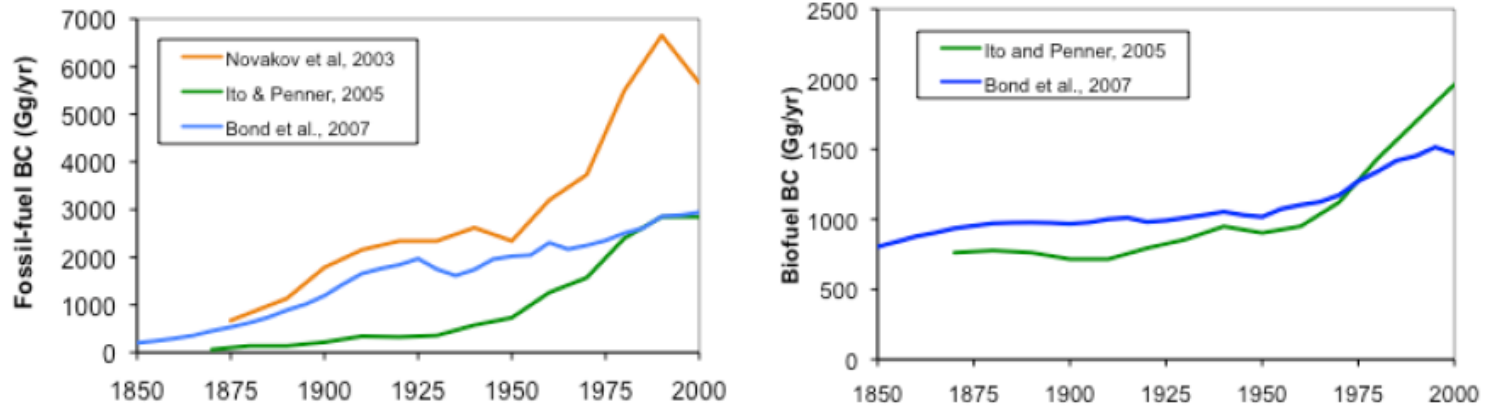
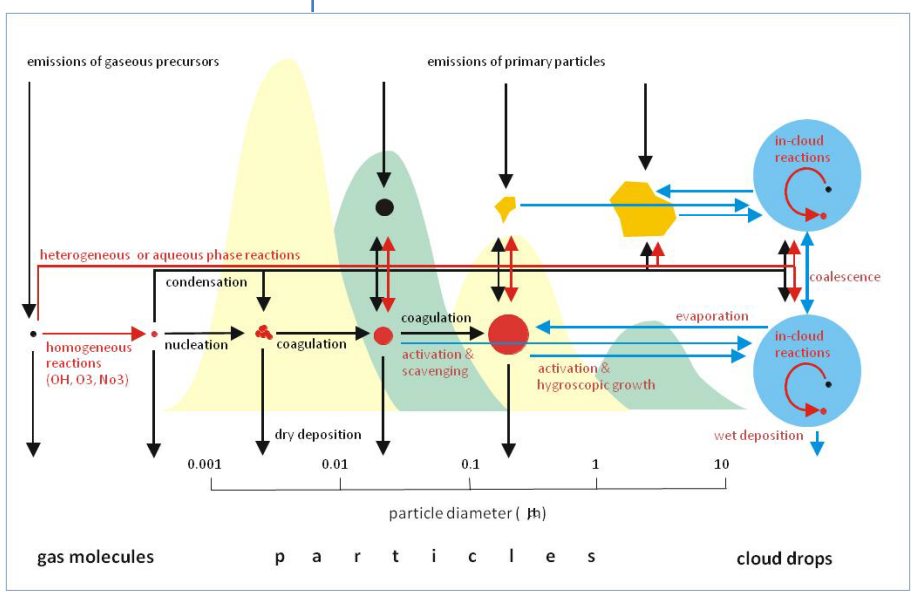
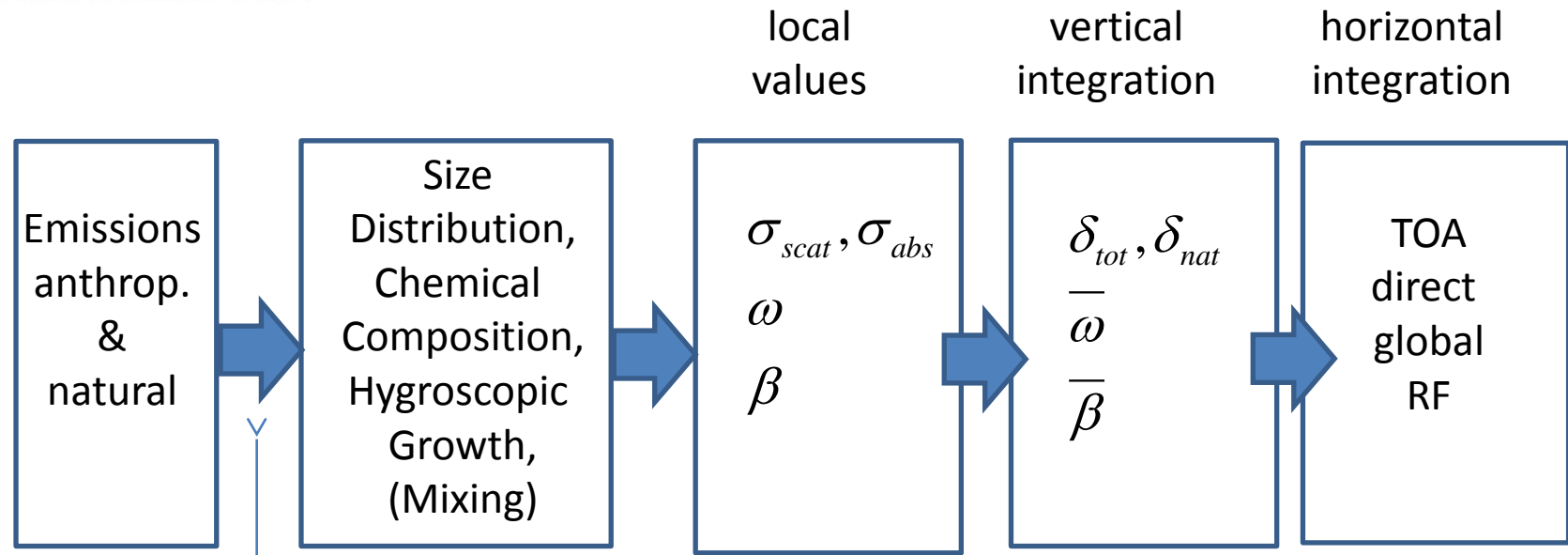
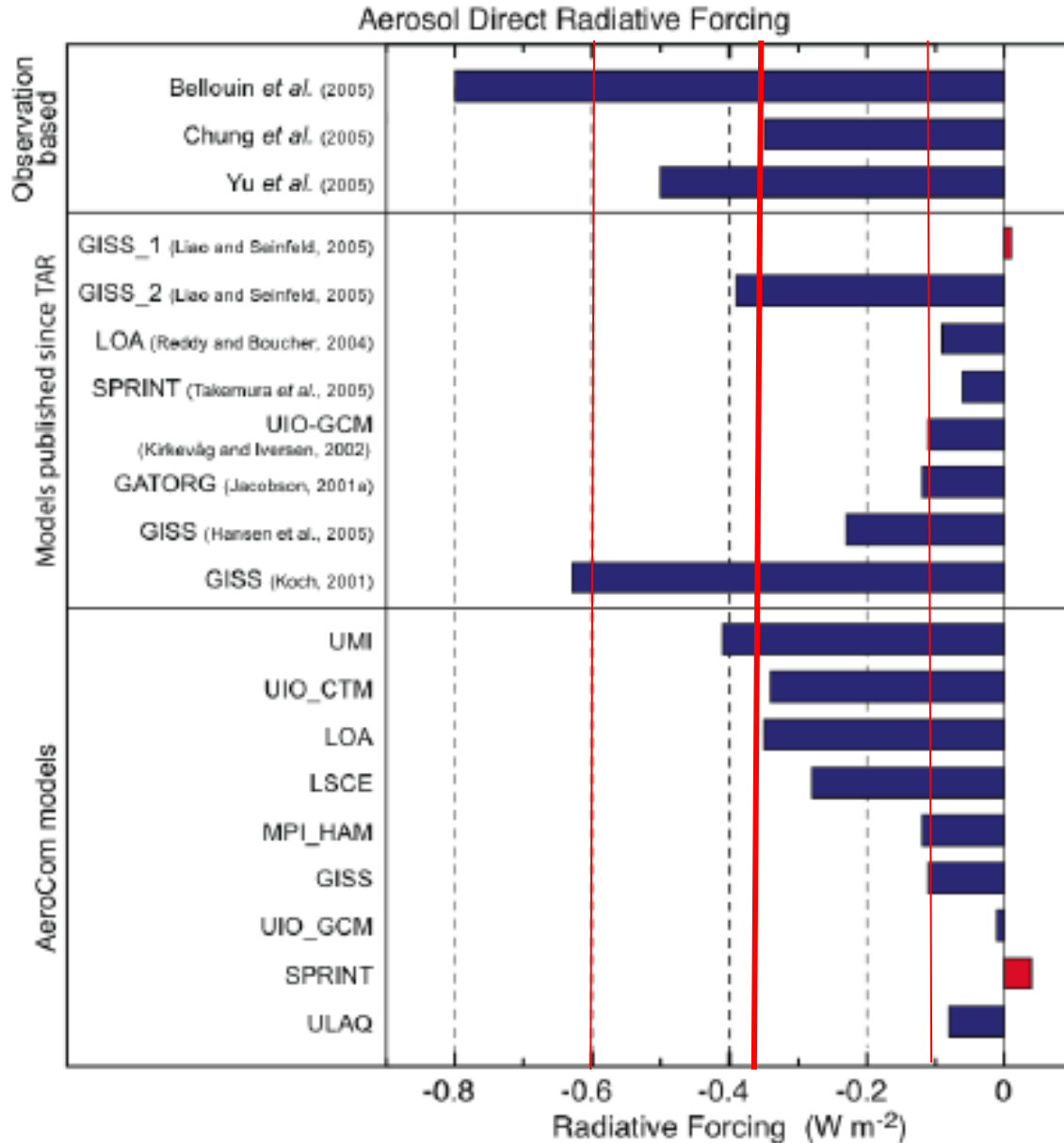


Figure 2.5 Historical BC emission trends from fossil-fuel (left) and biofuel (right) combustion from Novakov et al. (2003), Ito and Penner (2005) and Bond et al. (2007)

BC measured as

- absorbing aerosol
- elemental carbon
- fraction of PM



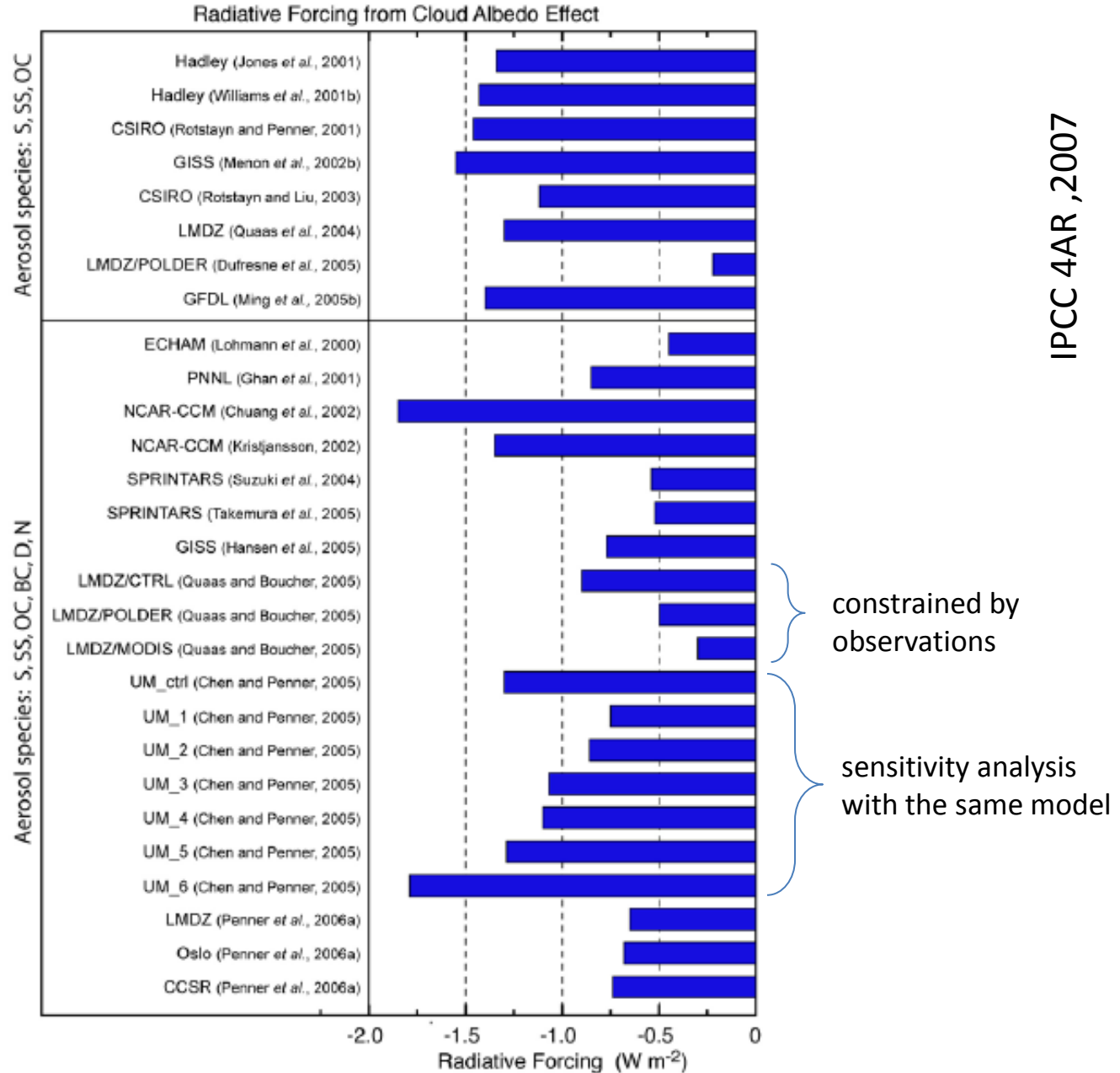


IPCC 4AR, 2007

same emissions



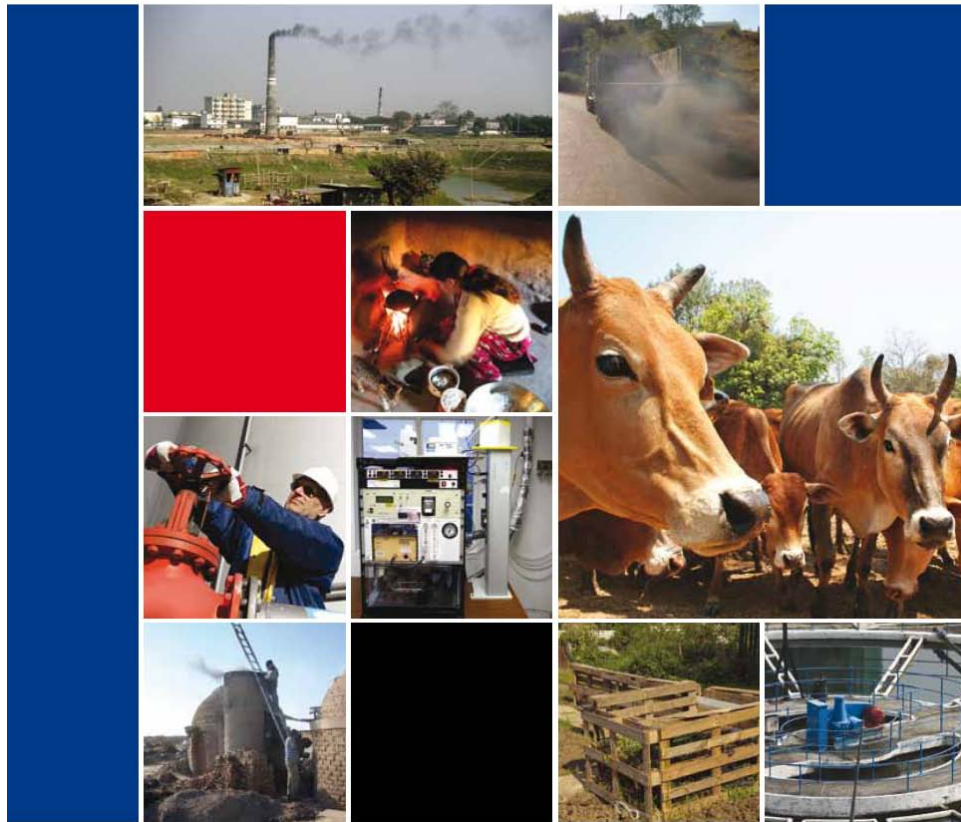
TOA 1st Indirect Radiative Forcing by Aerosols





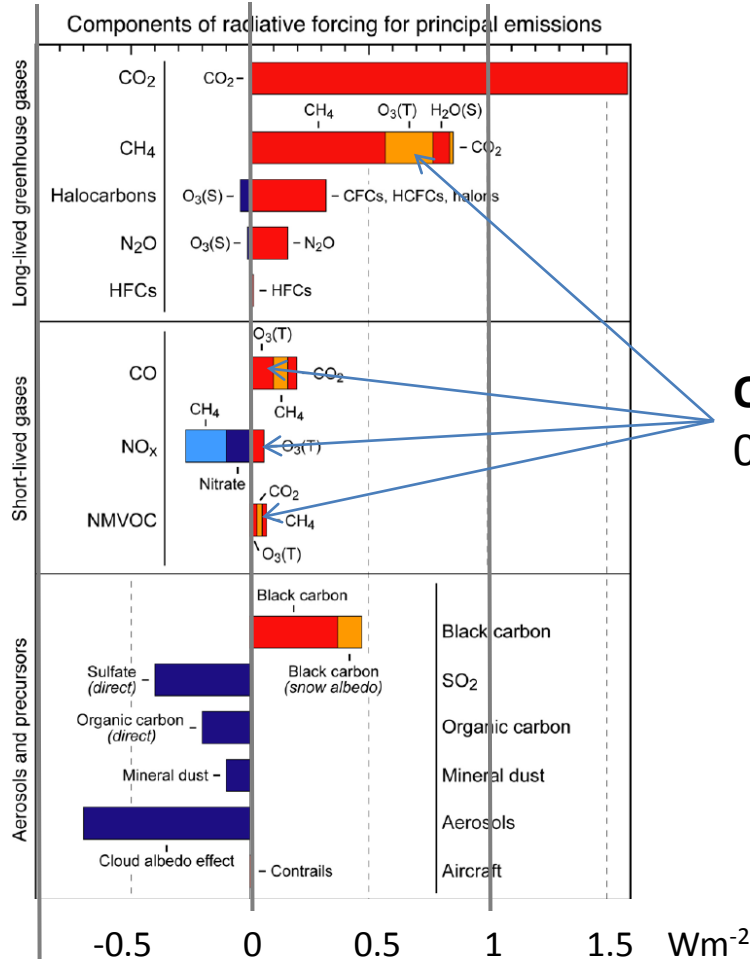
Integrated Assessment of Black Carbon and Tropospheric Ozone

Summary for Decision Makers





TOA Radiative Forcing for emissions



O₃, troposphere
 0.25 - 0.45 Wm⁻²

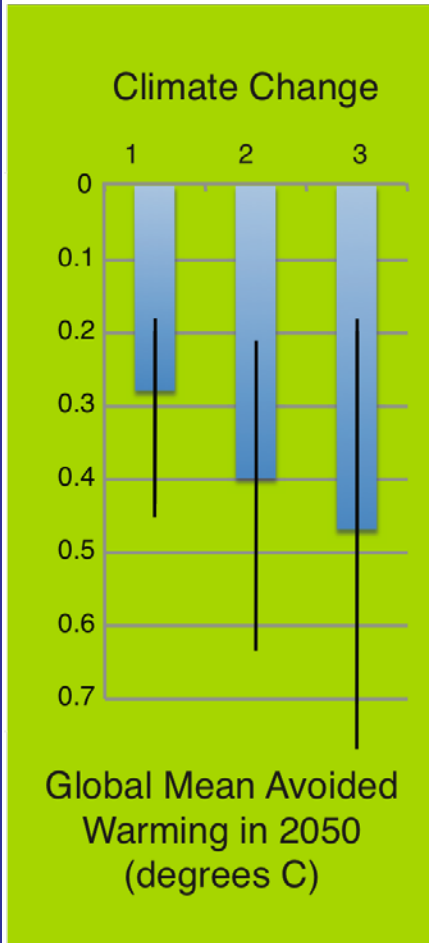
BC direct effect
 BC indirect effects
 BC deposition
BC total

0.0 - 1.0 Wm⁻²



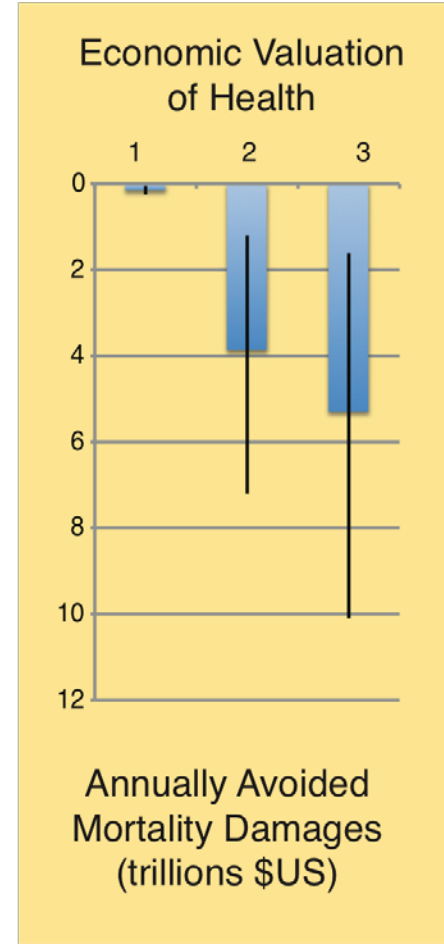
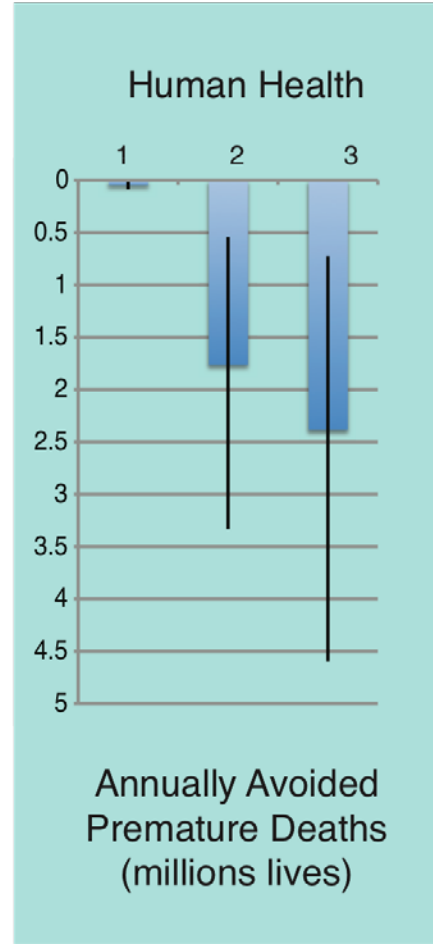
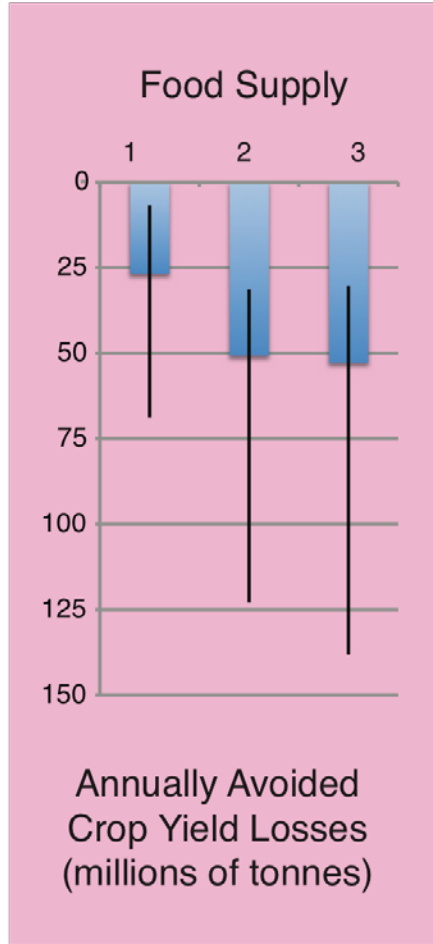
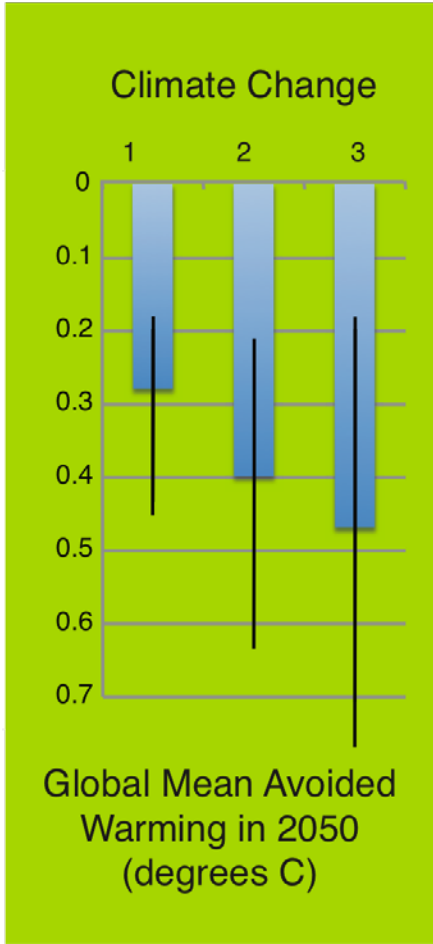
Global Impacts of Additional Emissions Controls on Methane and Products of Incomplete Combustion

1: CH₄ measures, 2: CH₄+BC Group 1 measures, 3: CH₄+ all BC measures



Climate given at 2050, air quality benefits for 2030 and beyond

Global Impacts of Additional Emissions Controls on Methane and Products of Incomplete Combustion
 1: CH₄ measures, 2: CH₄+BC Group 1 measures, 3: CH₄+ all BC measures



Climate given at 2050, air quality benefits for 2030 and beyond



Published standards

EN 12341: PM10 filter sampling & weighing (required by 2008 directive)

EN 14907: PM2.5 filter sampling & weighing (required by 2008 directive)

EN 14902: Cd, As Ni, Pb in PM

EN 15549: BaP in PM

prEN 15980: PAHs in PM

CEN TC 264 – currently active WGs

WG 15: PM2.5 and PM10 (filter sampling & weighing)

WG 21: measurement of PAHs

WG 32: Particle number concentration

WG 35: EC/OC in PM

TC amount measured in NIST RM 8785 by 13 labs in Europe

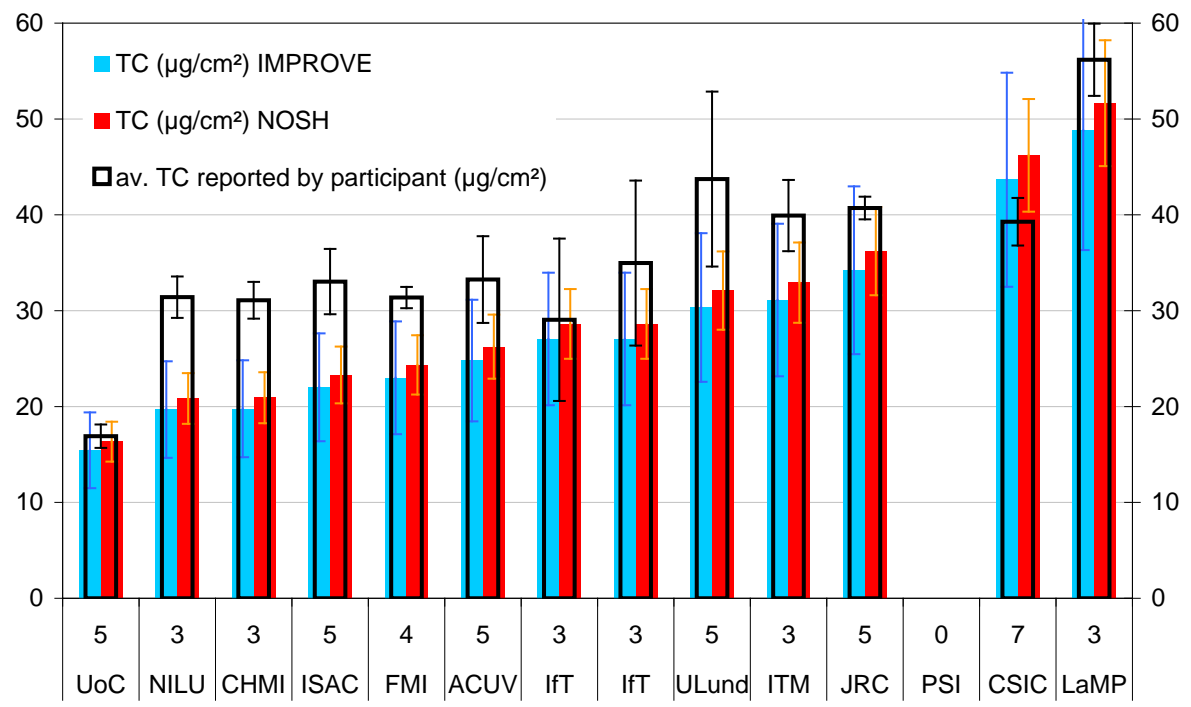


Fig. 1: TC amounts determined by the EUSAAR partners and reference values. Also shown are the uncertainties of these values.

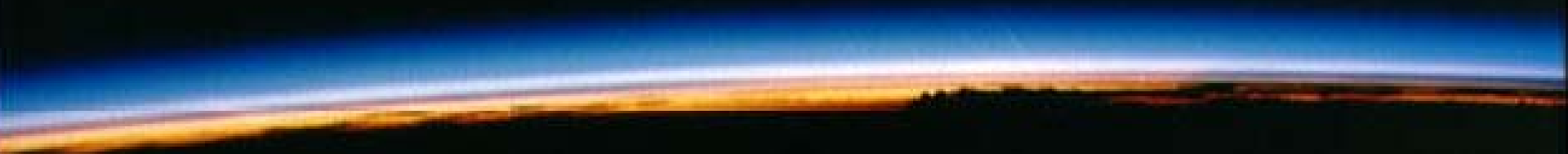
- Deviations to reference cannot be explained by measurement inaccuracy (based on previous inter-comparisons)
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- Policies often affect markets, or are themselves market based: standardization contributes to creating a level playing field. (BC emissions tradeable like CO2 emissions?)
- Aerosol & Climate science has become relevant!

thanks





AeroCom (Schultz et al., 2006)

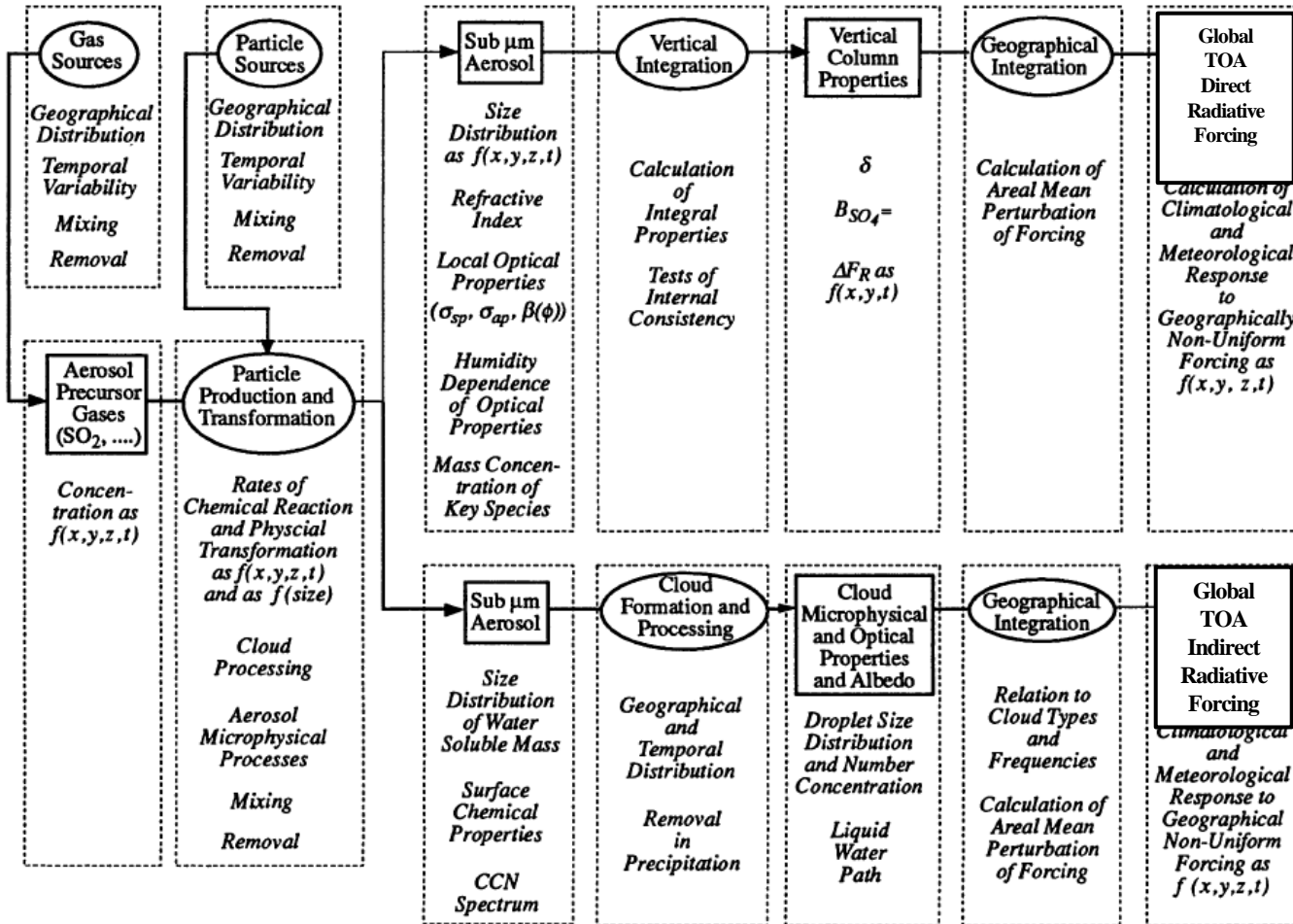
compared instantaneous direct radiative forcing due to anthropogenic aerosols produced by 9 different global models (from Europe, Japan and the USA) with detailed aerosol modules, USING THE SAME EMISSIONS

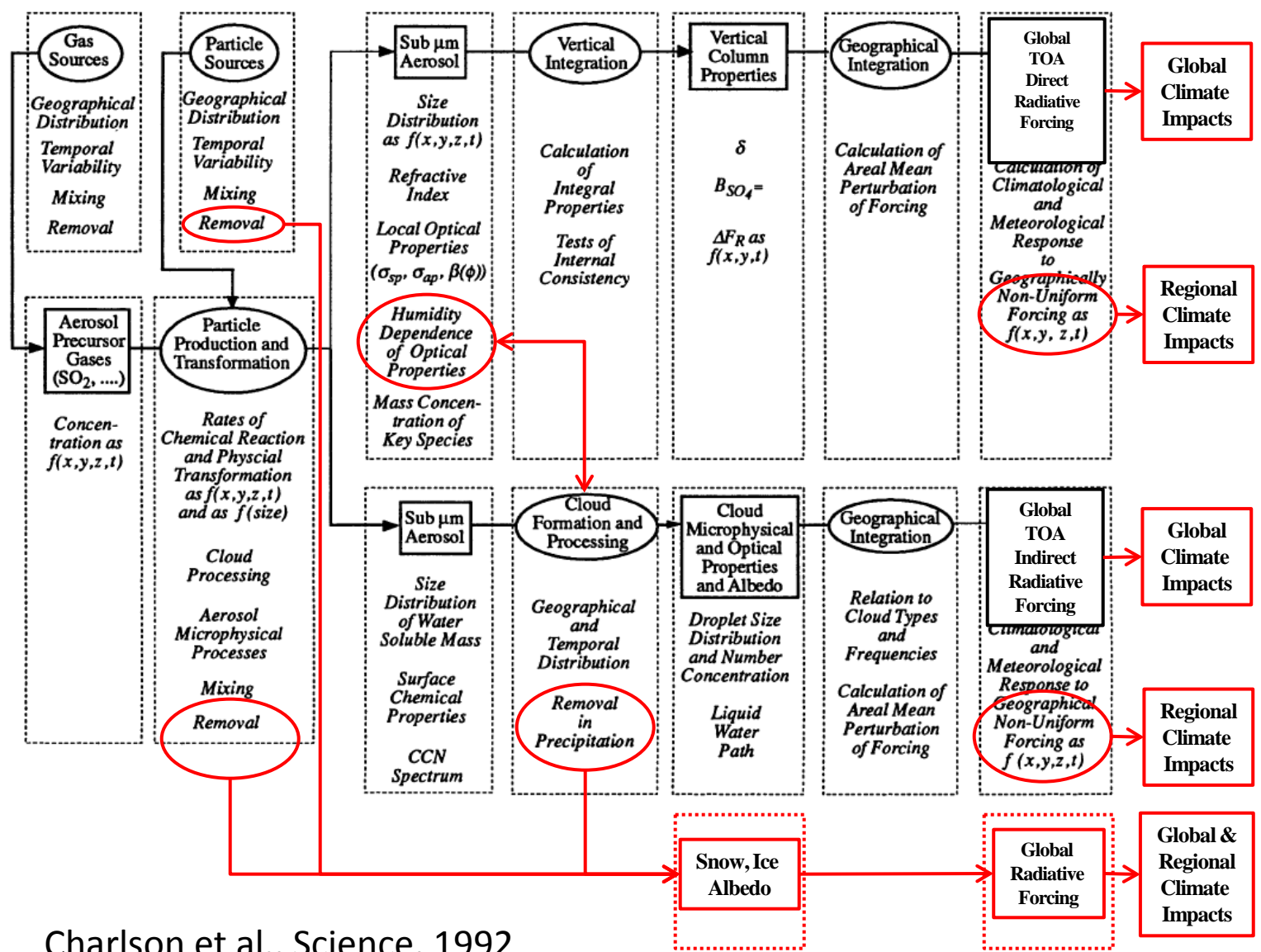
Differences in computed direct aerosol forcing (range $+0.04$ to -0.41Wm^{-2}) are due to:

- diversity in simulated aerosol residence times
- mass extinction coefficients
- **forcing efficiencies** (forcing per unit optical depth)

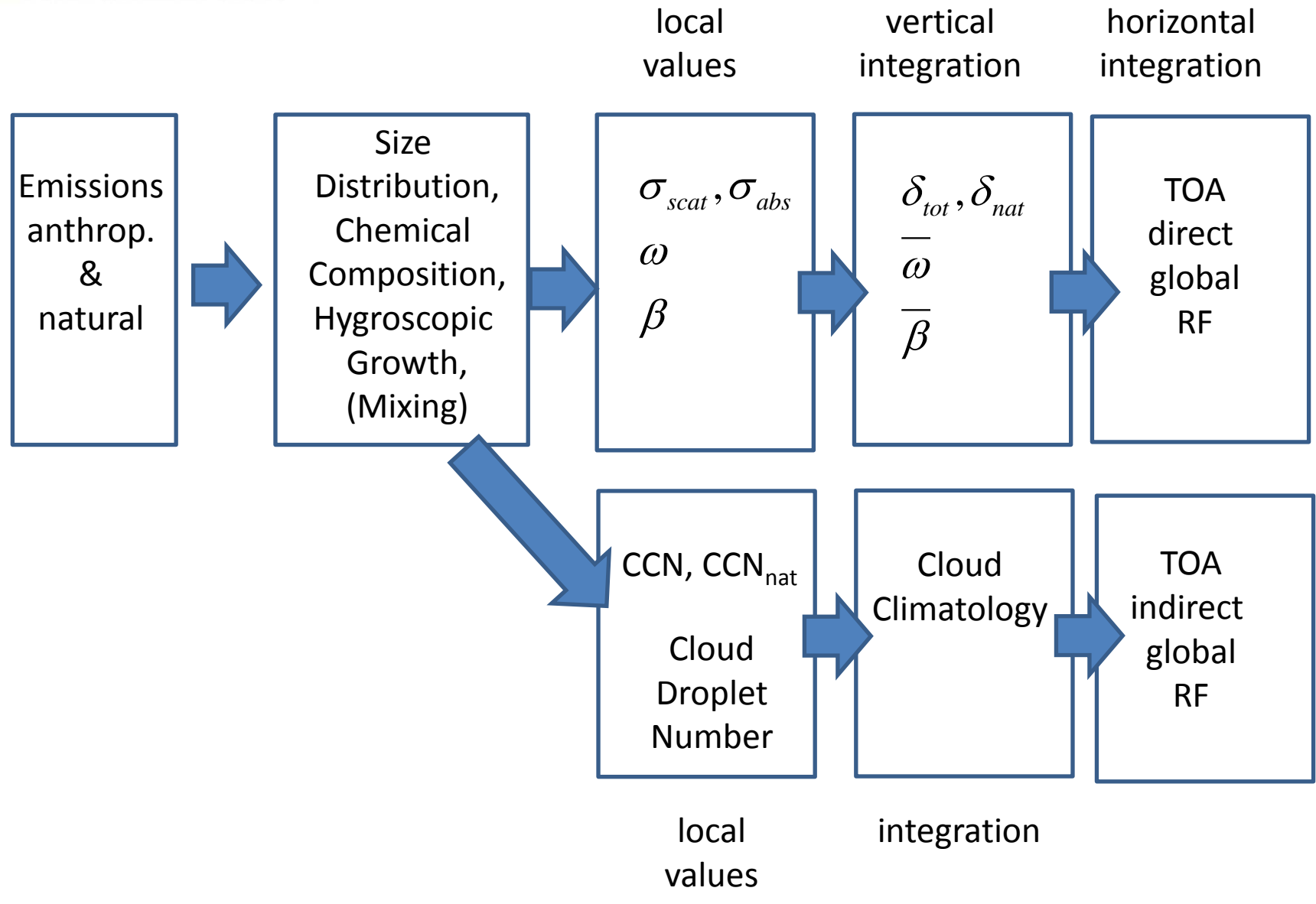
+

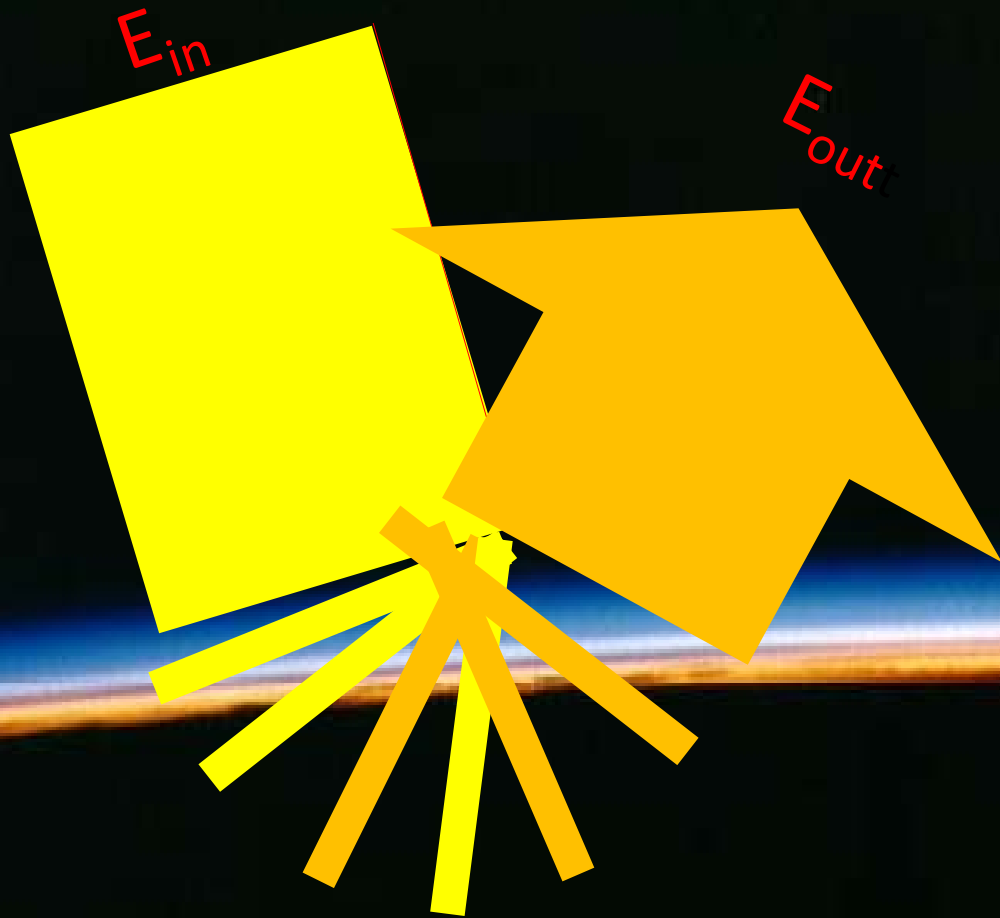
- in clear-sky conditions: aerosol absorption, size, and surface albedo
- in cloud-sky conditions: cloud fields and relative altitudes of absorbing aerosol and clouds





Charlson et al., Science, 1992





$$RF = (E_{in} - E_{out}) - (E_{in}^{pre} - E_{out}^{pre})$$

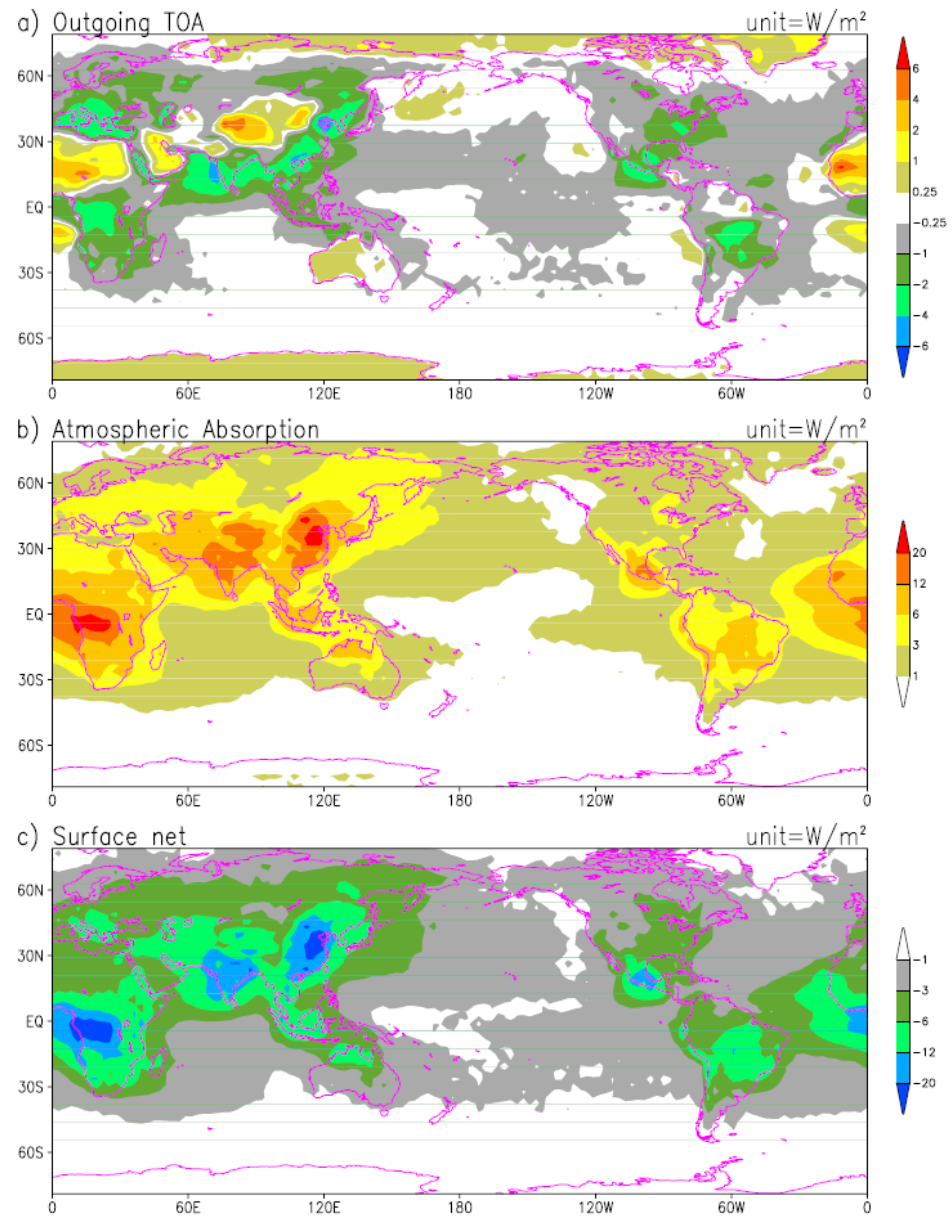


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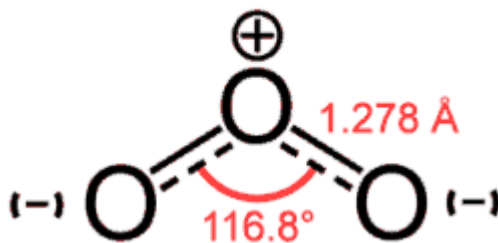
$$RF = \Delta E_{in} - \Delta E_{out}$$

$$RF = \Delta E_{in,solar} - \Delta E_{out,GHG} - \Delta E_{out,aerosols} - \Delta E_{out,surface_albedo}$$





O₃ Ozone Molecule



scenarios → emissions → concentration fields → climate forcing

- Model validation (against measured aerosol constituents' concentrations too)
- Model inter-comparisons

AeroCom (Schultz et al., 2006) compared instantaneous direct radiative forcing due to anthropogenic aerosols produced by 9 different global models (from Europe, Japan and the USA) with detailed aerosol modules.

Differences in computed direct aerosol forcing (range +0.04 to -0.41Wm⁻²) are due to:

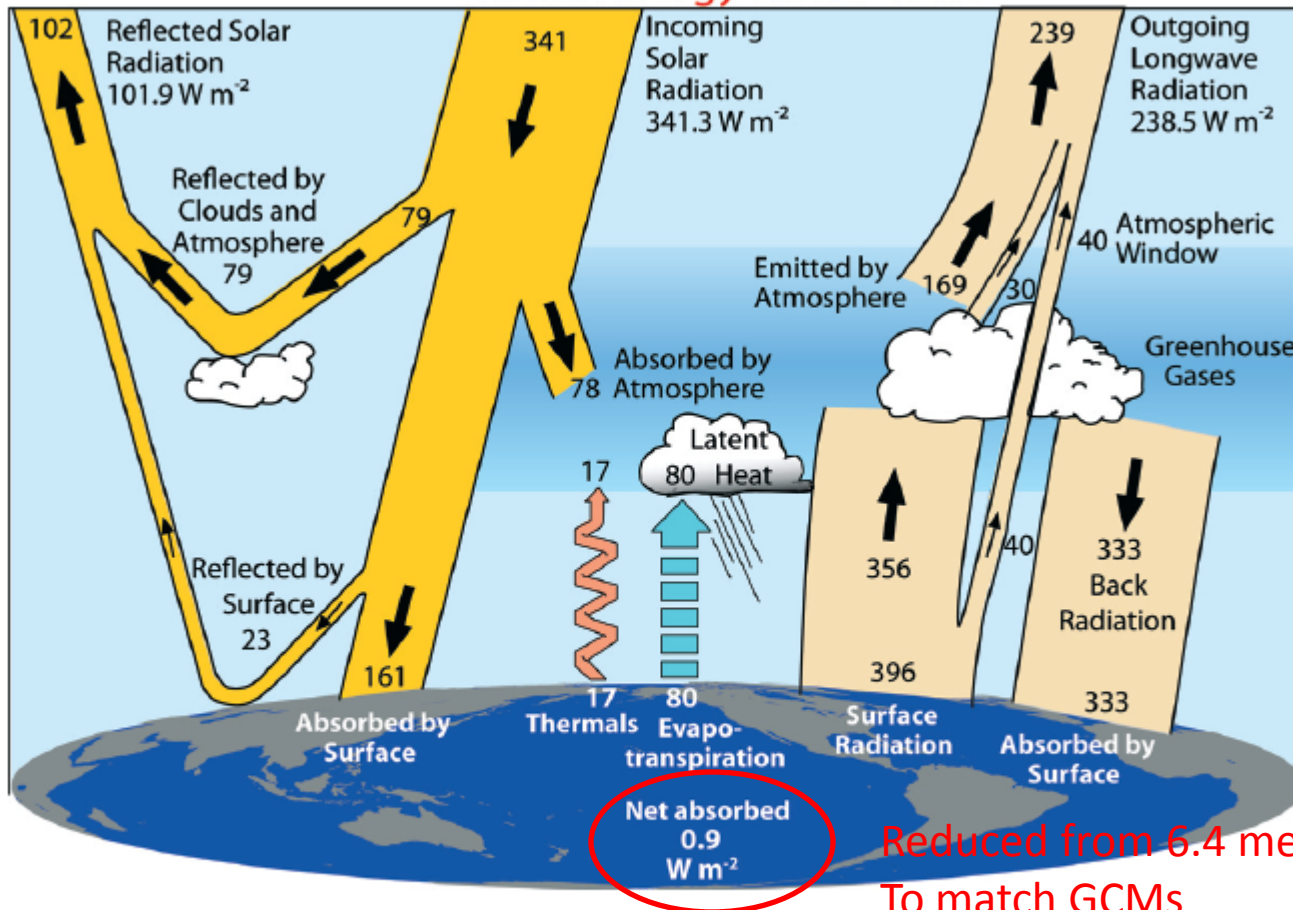
- diversity in simulated aerosol residence times
- mass extinction coefficients
- **forcing efficiencies** (forcing per unit optical depth)

+

- in clear-sky conditions: aerosol absorption, size, and surface albedo
- in cloud-sky conditions: cloud fields and relative altitudes of absorbing aerosol and clouds



Global Energy Flows $W m^{-2}$



Reduced from 6.4 measured to 0,9
To match GCMs

FIG. 1. The global annual mean Earth's energy budget for the Mar 2000 to May 2004 period ($W m^{-2}$). The broad arrows indicate the schematic flow of energy in proportion to their importance.

Computing current aerosol **direct** radiative forcing:

- E.g. Haywood and Shine's approximation (1995) for a given WL:

$$\Delta_a F \uparrow = \frac{1}{2} F_T T^2 (1 - A_c) \{ \omega \overline{\beta_a} (1 - \overline{R_s})^2 - 2(1 - \omega) \overline{R_s} \} \delta_a$$

Is total present forcing, not

Bar = average over solar an

In Charlsons work, optical depth is that of anthro
 If you use measurements you have natural + ant

- Radiative transfer modules (e.g. Modtran) need:
 - extinction vertical profile
 - extinction, absorption, and asymmetry factor WL dependence

Computing current aerosol **indirect** radiative forcing:

- mainly (?) remote sensing
- CCN number ↔ CN number size distribution, hygroscopicity

Measurements of aerosol characteristics relevant to climate forcing

are currently performed for research (rather than regulation) purposes

⇒ they are done with **conscience** and awareness

Different flow rates in Nephelometer give same scattering measurements

Different ΔT in CPCs give same number of particles with $D_p > 50\text{nm}$

go back to a basic physical principle

⇒ they aim at getting true (and not only “comparable”) values

No need for standardisation if several methods can measure the same truth

E.g. - aerosol absorption from **photoacoustic spectrometer** =

- **extinction cell** (extinction – scattering)

In contrast with PM mass concentration (gravimetric methods) which give wrong data because of sampling artifacts and analytical artifacts (aerosol water uptake at 45-50% RH), and then errors have to be the same everywhere.

⇒ continuous closure checks used as stringent data quality check

Still a lot has been done regarding standardisation in the field of aerosol metrology for climate over the past decade (GAW SAG, GAW-WCCAP, ...,EMEP) Very often payed by European Commission

EUROPEAN COMMISSION
DIRECTORATE-GENERAL
Joint Research Centre
2003 WMO-GAW Guidelines (72 p.)
2011 WMO-GAW SOPs (76 p.)

Joint Research Centre

WMO/GAW
AEROSOL MEASUREMENT PR
GUIDELINES AND RECOMMEN



September 2003

No. 153



WORLD METEOROLOGICAL ORGANIZATION
GLOBAL ATMOSPHERE WATCH

WMO/GAW Standard Operating Procedures
In-Situ Measurements of Aerosol Mass
Concentration, Light Scattering
and Light Absorption

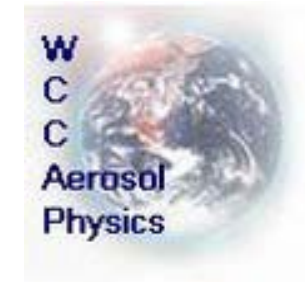
GAW No.



WMO/TD-No.
2010

WORLD METEOROLOGICAL ORGANIZATION
GLOBAL ATMOSPHERE WATCH





- The GAW – World Calibration Centre for Aerosol Physics
<http://gaw.tropos.de/WCCAP/>

- Intercomparisons

- Audits (on-site check of sampling configuration, instruments, etc...)

Joint Research Centre

Tamanrasset/Assekrem	Algeria		2003
Hohenpeissenberg	Germany	2003	
Anmyeon	South-Korea		2004
Pallas	Finland		2004
Danum Valley	Malaysia		2004
Mace Head	Ireland		2004
Waliguan-Beijing	China		2005
Danum Valley	Malaysia		2005
Danum Valley	Malaysia		2006
Cape Point	South Africa		2006
Ny-Alesund	Norway		2006
Beo-Moussala	Bulgaria		2006
Izana	Tenerife, Spain		2006
Point Barrow	Alaska, USA		2007
Monte Cimone	Italy		2007
Harwell	UK		2007
Mauna Loa	Hawaii, USA		2007
Finokalia	Crete, Greece		2007
Cabauw	The Netherlands		2008
Bukit Koto Tabang	Indonesia	2008	
Vavihill	Sweden		2008
Cape Grim	Australia		2008
Shangdianzi	China		2009
Danum Valley	Malaysia		2009
Hyytiälä	Finland		2009
Birkenes	Norway		2009
Finokalia	Crete, Greece		2009
Preila	Lithuania		2009
Aspvreten	Sweden		2009
Melpitz	Germany		2009
Ispra	Italy		2010
JFJ, PdD, Kosetice, K-Puszt			2010 ?

I can make a GANTT-like diagram with this too, highlighting stations from the 5 continents

Computing future aerosol radiative forcing:

scenarios → emissions → concentration fields → climate forcing

- Model validation (against measured aerosol constituents' concentrations too)
 - inorganics: nitrate, sulfate, (ammonium), etc...
 - ∃ reference materials (NIST SRM 2694, IRMM xxx, ...)
 - ∃ yearly or twice yearly regular inter-comparisons
 - GAW - QA/SAC-Americas: 78 labs worldwide
(<http://gasac-americas.org/>)
 - EMEP- CCC: 51 labs worldwide
(<http://tarantula.nilu.no/projects/ccc/>)
 - carbonaceous aerosol
 - reference material (NIST RM 8785) ?
 - yearly inter-comparison with EUSSAR project (18 Eu labs)
 - punctual inter-continental inter-comparison
EnvCan, IMPROVE, EUSAAR

TC amount measured in NIST RM 8785 by 13 labs in Europe

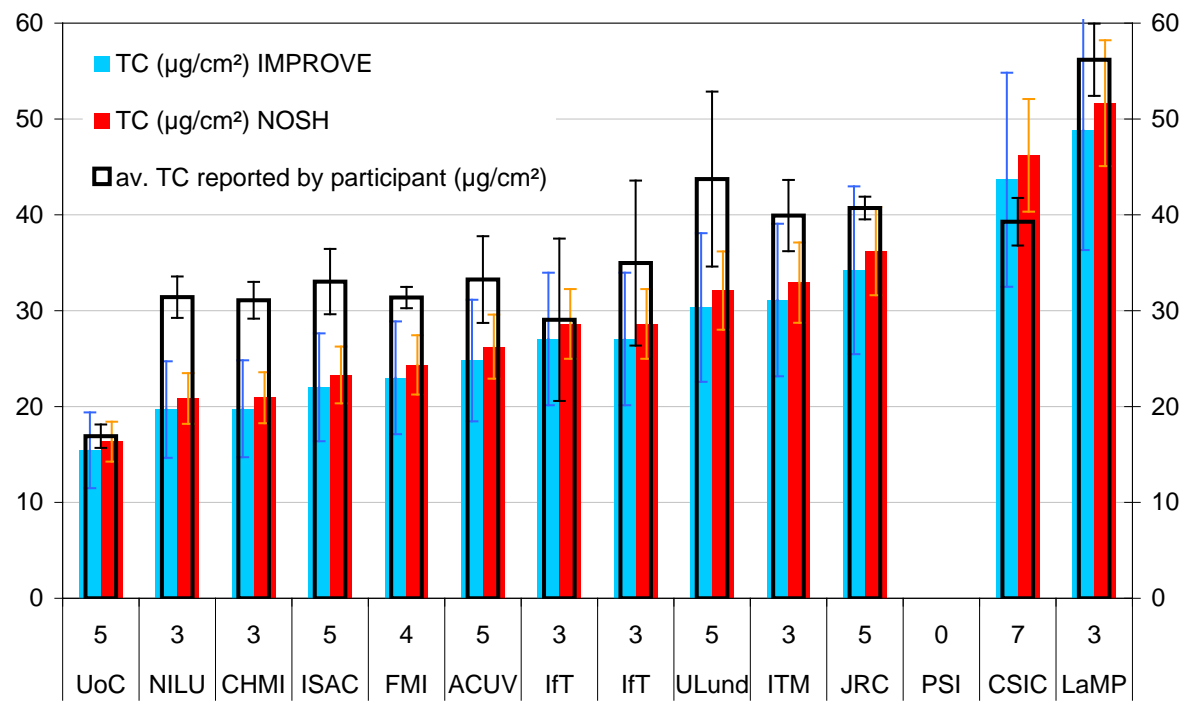
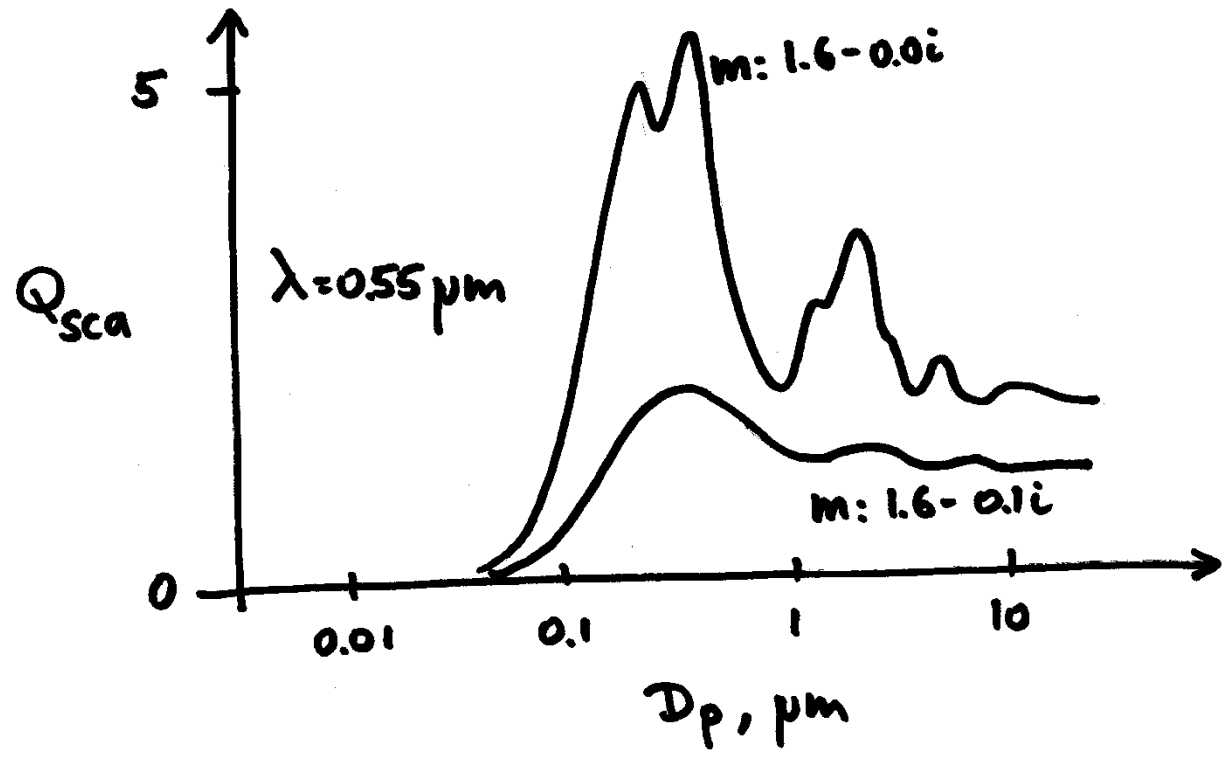


Fig. 1: TC amounts determined by the EUSAAR partners and reference values. Also shown are the uncertainties of these values.

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effects on radiation balance: scattering and absorption

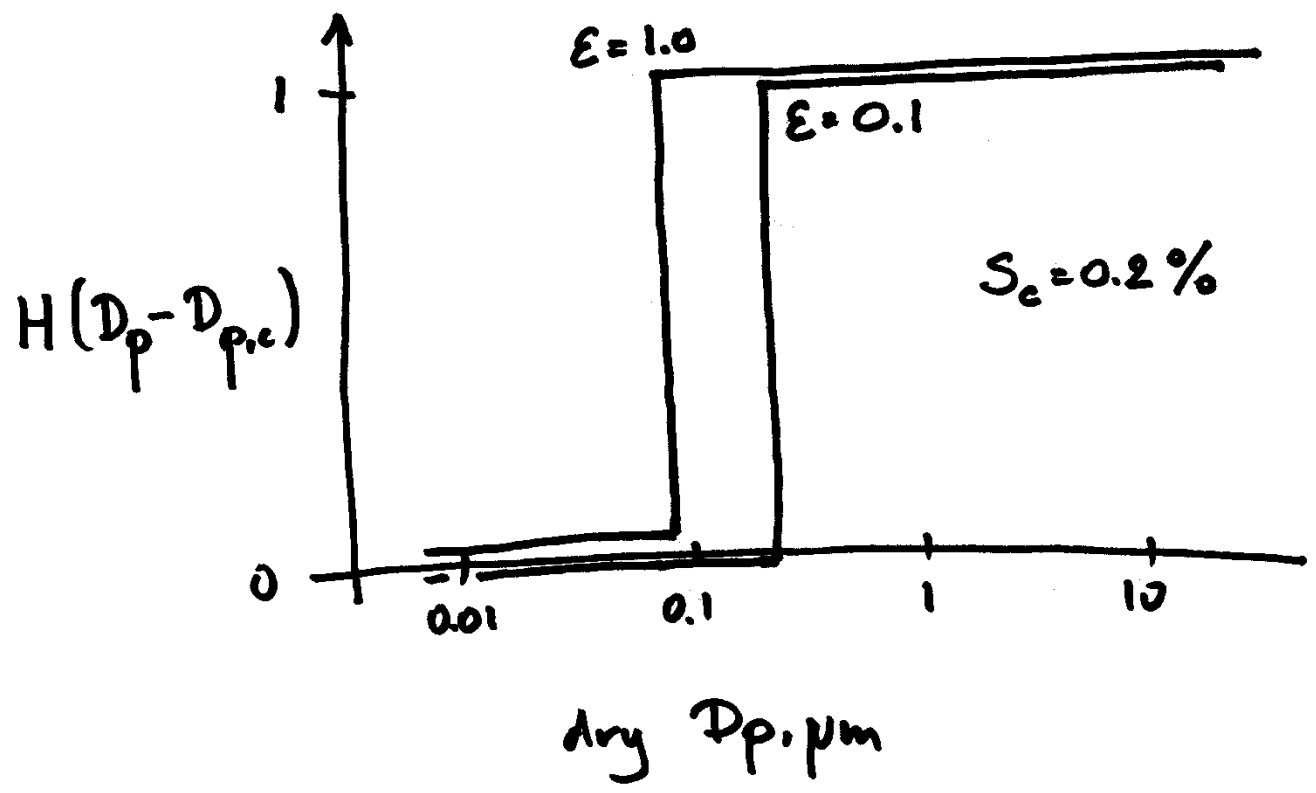
$$\sigma_{sca}(\lambda, \bar{r}, t) = \iint Q_{sca}(\lambda, D_p, m) \frac{\pi D_p^2}{4} N(D_p, m, \bar{r}, t) dm dD_p$$



effects on clouds: cloud condensation nuclei

$$CCN(S_c, \bar{r}, t) = \iint H[D_p - D_{p,c}(S_c, \epsilon)] N(D_p, \epsilon, \bar{r}, t) d\epsilon dD_p$$

Joint Research Centre



The GAW – World Calibration Centre for Aerosol Physics

<http://gaw.tropos.de/WCCAP/>

- Intercomparisons
- Audits
- Travelling reference instruments (for especially remote stations)



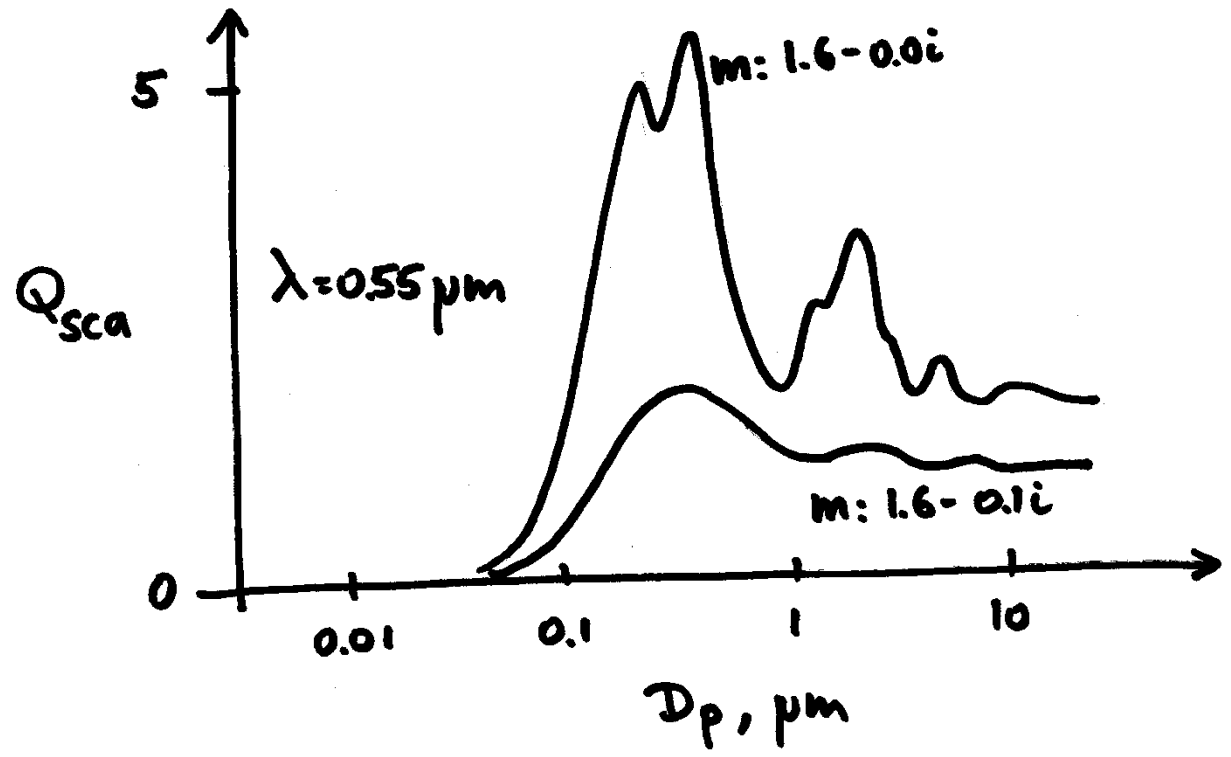
Definitions

What is a Standard?

- ▶ A standard is an established norm or requirement
- ▶ It is usually a formal document that establishes uniform technical criteria, methods, processes and practices
- ▶ It may be developed privately or unilaterally, for example by a corporation, regulatory body, military
- ▶ **Simply** They define how things should be done

effects on radiation balance: scattering and absorption

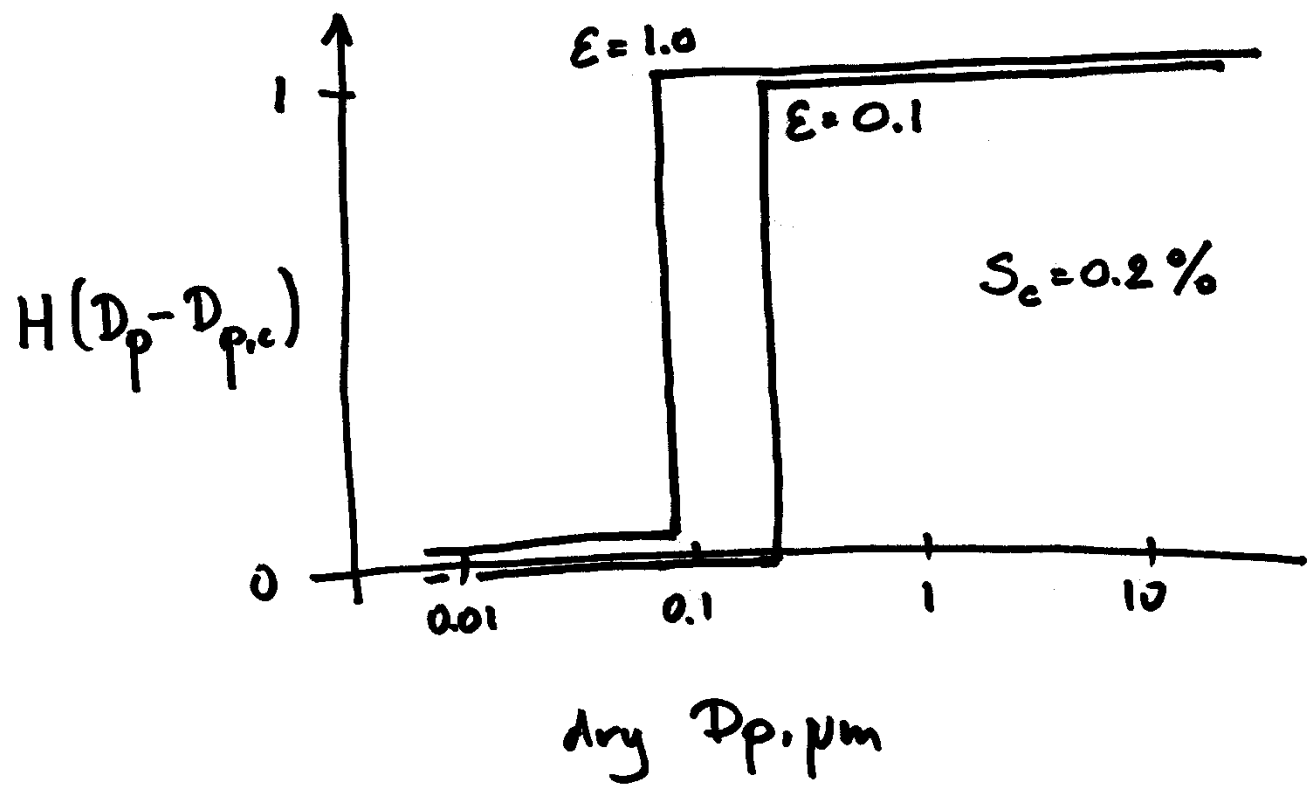
$$\sigma_{sca}(\lambda, \bar{r}, t) = \iint Q_{sca}(\lambda, D_p, m) \frac{\pi D_p^2}{4} N(D_p, m, \bar{r}, t) dm dD_p$$



effects on clouds: cloud condensation nuclei

$$CCN(S_c, \bar{r}, t) = \iint H[D_p - D_{p,c}(S_c, \epsilon)] N(D_p, \epsilon, \bar{r}, t) d\epsilon dD_p$$

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“Black Carbon” or “Elemental Carbon” is:

- carbon-containing particulate matter, resulting from incomplete combustion
 - it withstands high temperatures
 - it is black, hence absorbs light
- emitted together with CO₂, CO, NMHC, organic particulate matter)





Anthropogenic forcing??

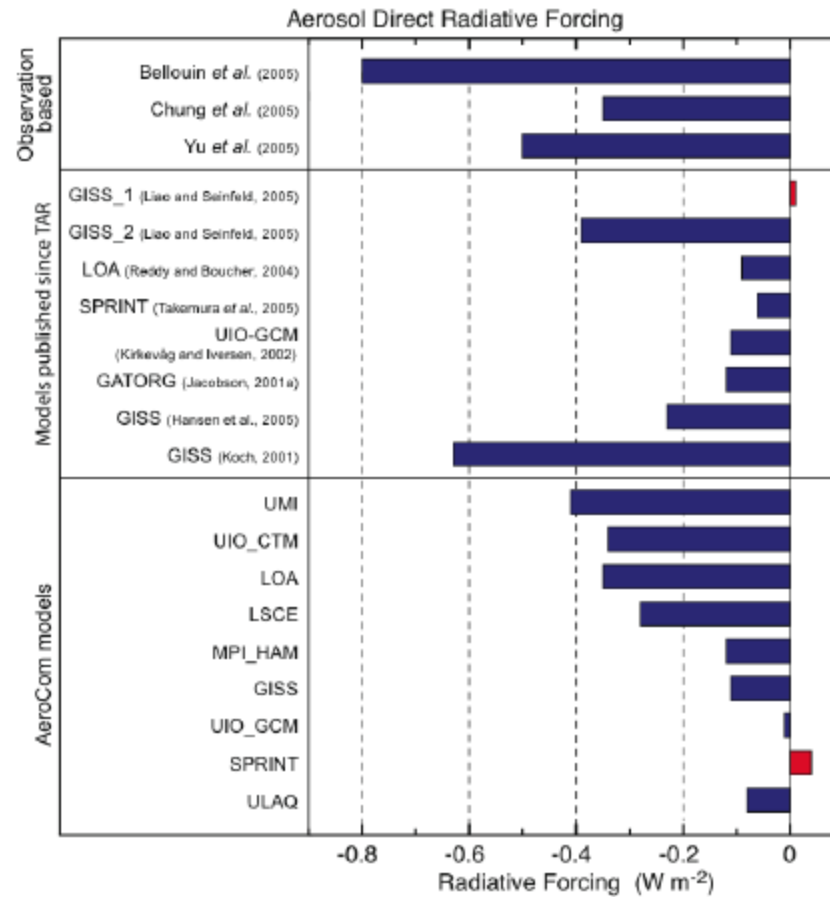


Figure 2.13. Estimates of the direct aerosol RF since the TAR from observational based studies, independent modelling studies, and AEROCOM results with identical aerosol and aerosol precursor emissions. GISS_1 refers to a study employing an internal mixture of aerosol, and GISS_2 to a study employing an external mixture.

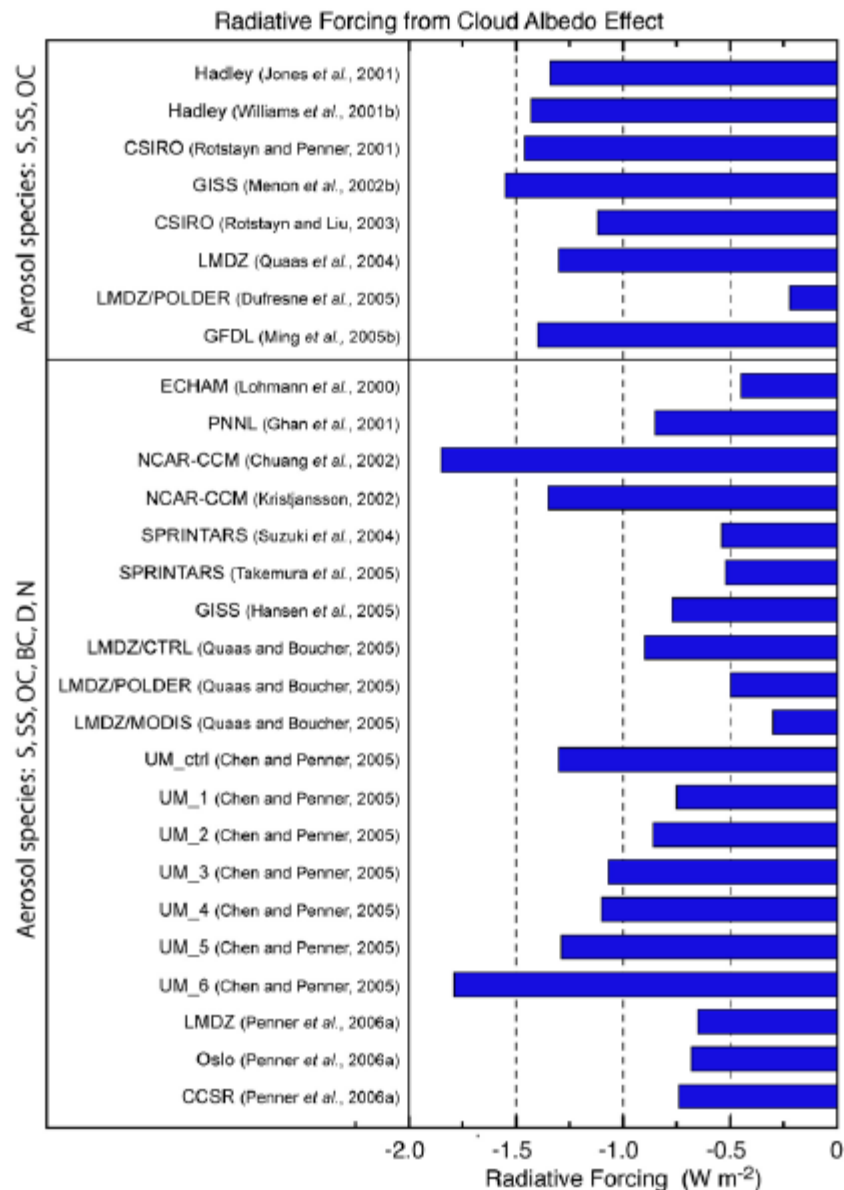
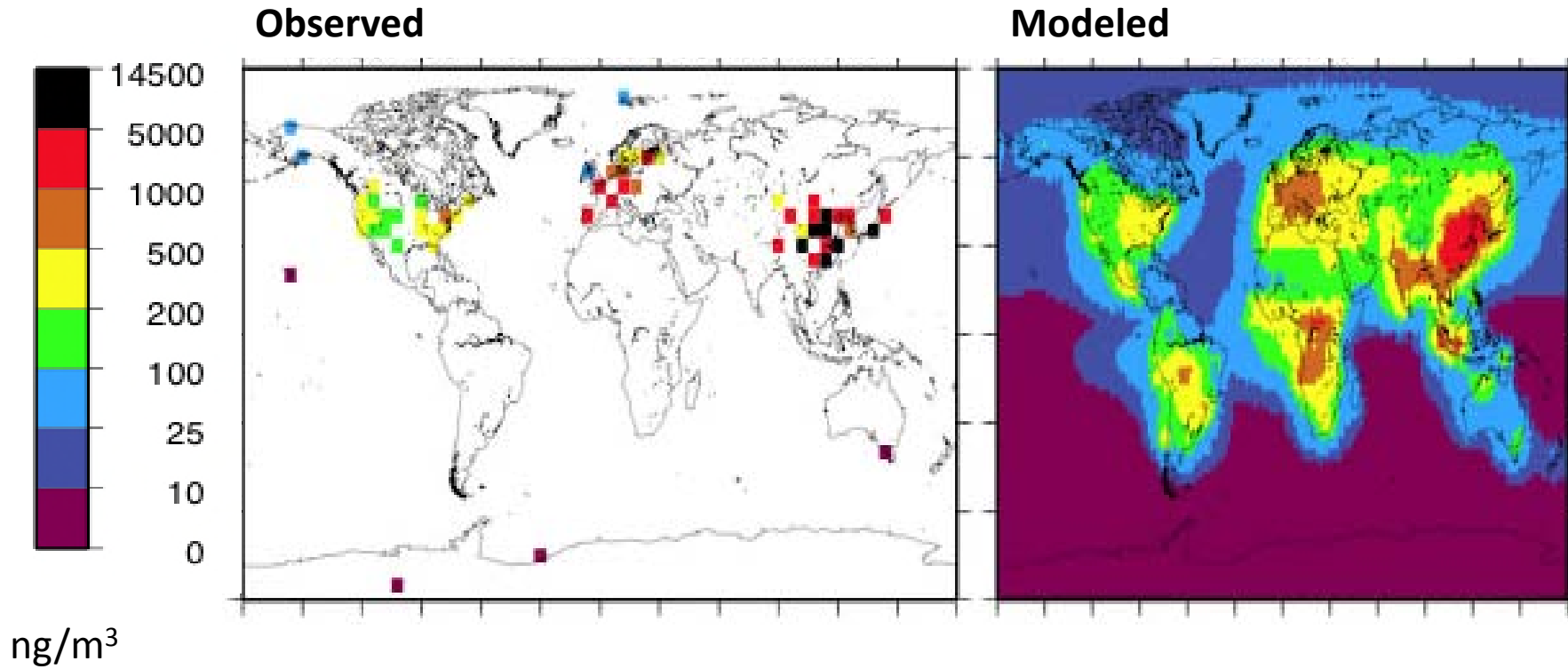


Figure 2.14. Radiative forcing due to the cloud albedo effect, in the context of liquid water clouds, from the global climate models that appear in Table 2.7. The labels next to the bars correspond to the published study; the notes of Table 2.7 explains the species abbreviations listed on the left hand side. Top panel: results correspond to models that consider a limited number of species, primarily anthropogenic sulphate (S). Bottom panel: results correspond to studies that include a variety of aerosol compositions and mixtures; the estimates here cover a larger range than those in the top panel. Chen and Penner (2005) present a sensitivity



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On air quality

Black carbon and ozone in the lower atmosphere are harmful air pollutants affecting health of humans and ecosystems

Black carbon, a component of particulate matter, and ozone both lead to premature deaths worldwide. (PM2.5: ~2 million per year , WHO)

Ozone is also the most important air pollutant, responsible for reducing crop yields, and thus affects food security.

On global and regional climate ?

On global and regional climate

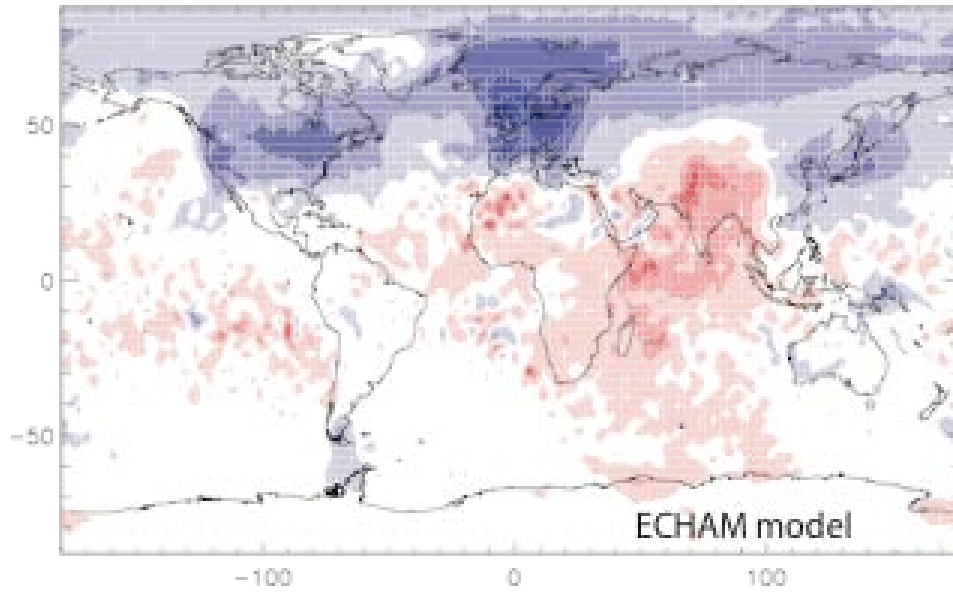
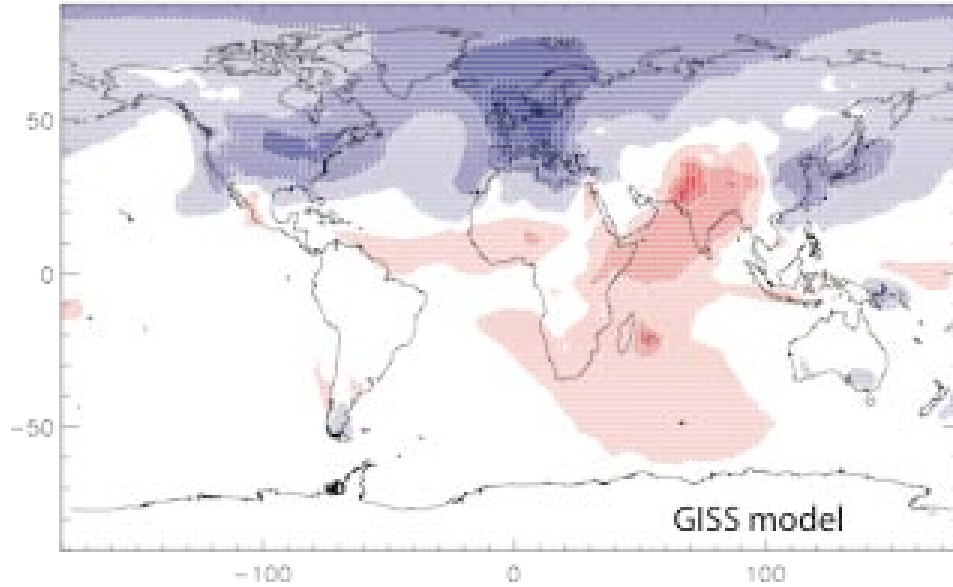
Changes in their burdens over the 20th Century result in an expected equilibrium global warming of 0.0-0.8 C due to BC and 0.1-0.4 C due to O₃. (The equilibrium warming expected from CO₂ is about 1.3 C.)

Atmospheric heating by BC disturbs tropical rainfall and regional circulation patterns such as the Asian monsoon.

Black carbon's darkening of snow and ice surfaces increases their absorption of sunlight, which, along with atmospheric heating, exacerbates melting of snow and ice around the world, including in the Arctic, the Himalayas.



change in BC deposition: 2005 to 2030 reference scenario



%

Avoidance of 0.2-1.8 million premature deaths per year , in North America, East Asia, SE Asia & Pacific, While in increase by 0.1-2.0 million in South, West & Central Asia and Africa..

Decreasing production of wheat, rice corn and soy by 7 to 120 million tonnes across Asia with an associated economic loss of US\$ 1 – 20 billion. Increasing crop yields in US and Europe.

The compensating warming and cooling impacts of changes in BC, ozone and other aerosols lead to a small net warming of less than 0.1 C globally

Black carbon's darkening of snow and ice and atmospheric heating, keeps exacerbating melting of snow and ice in the Himalayas

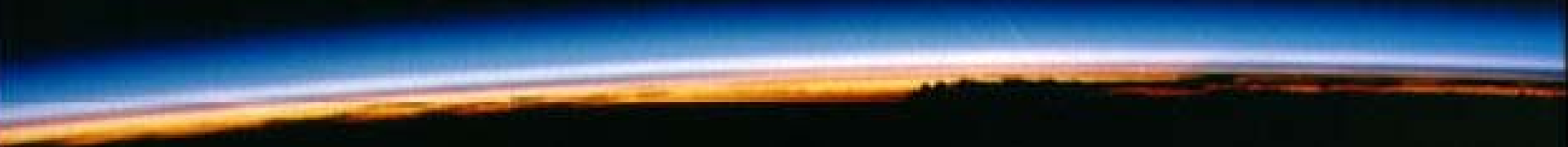


Conclusions so far:

based on our knowledge of the behaviour of BC and ozone in the atmosphere, and of their impacts on air quality, And on lobal and regional climate in teh near term

it makes a lot of sense to look for measures that specifically target the emissions of BC and ozone precursors in order to solve a range of important problems at once.

thanks





scattering, absorption

$$\sigma_{sp}(\lambda) = \iint Q_{sca}(\lambda, D_p, m) \frac{\pi D_p^2}{4} n(D_p, m) dm dD_p$$

cloud activation:

$$CCN(S_c) = \iint H[D_p - D_{p,c}(S_c, \epsilon)] n(D_p, \epsilon) d\epsilon dD_p$$

particulate matter

$$PM_x = \int_0^x \int \rho(D_p) \frac{\pi D_p^3}{6} n(D_p, \rho) d\rho dD_p$$

assuming internal mixing of chemical species into an average one

$$n(D_p, \epsilon) = n(D_p) \delta(\epsilon - \bar{\epsilon})$$

with δ the Kronecker delta function

assuming external mixing of i chemical species

$$n(D_p, \epsilon) = \sum_1^k n_i(D_p) \delta(\epsilon - \epsilon_i)$$

- We are more aware of the importance of a range of aerosol processes in the climate system.
- Quantifying the climate impact of changing the concentration of individual chemical components of the atmospheric aerosol is still very uncertain.
- Quantifying the climate impact of individual human activities, incl. policy measures, is even more uncertain (emissions & their chemical fingerprint are uncertain) but the sign is usually known.
- Deal with uncertainty by adopting a multi-pollutant /multi-effect approach (not just aerosol emissions, not just climate impacts)

- Standardization doesn't reduce uncertainty of measurements or assessments but improves their comparability.
- Policy making and policy implementation about comparing (e.g. climate effect with and without a policy, comparing PM values with target /limit values)
- Policies often affect markets, or are themselves market based: standardization contributes to creating a level playing field. (BC emissions tradeable like CO2 emissions?)
- Aerosol & Climate science has become relevant!

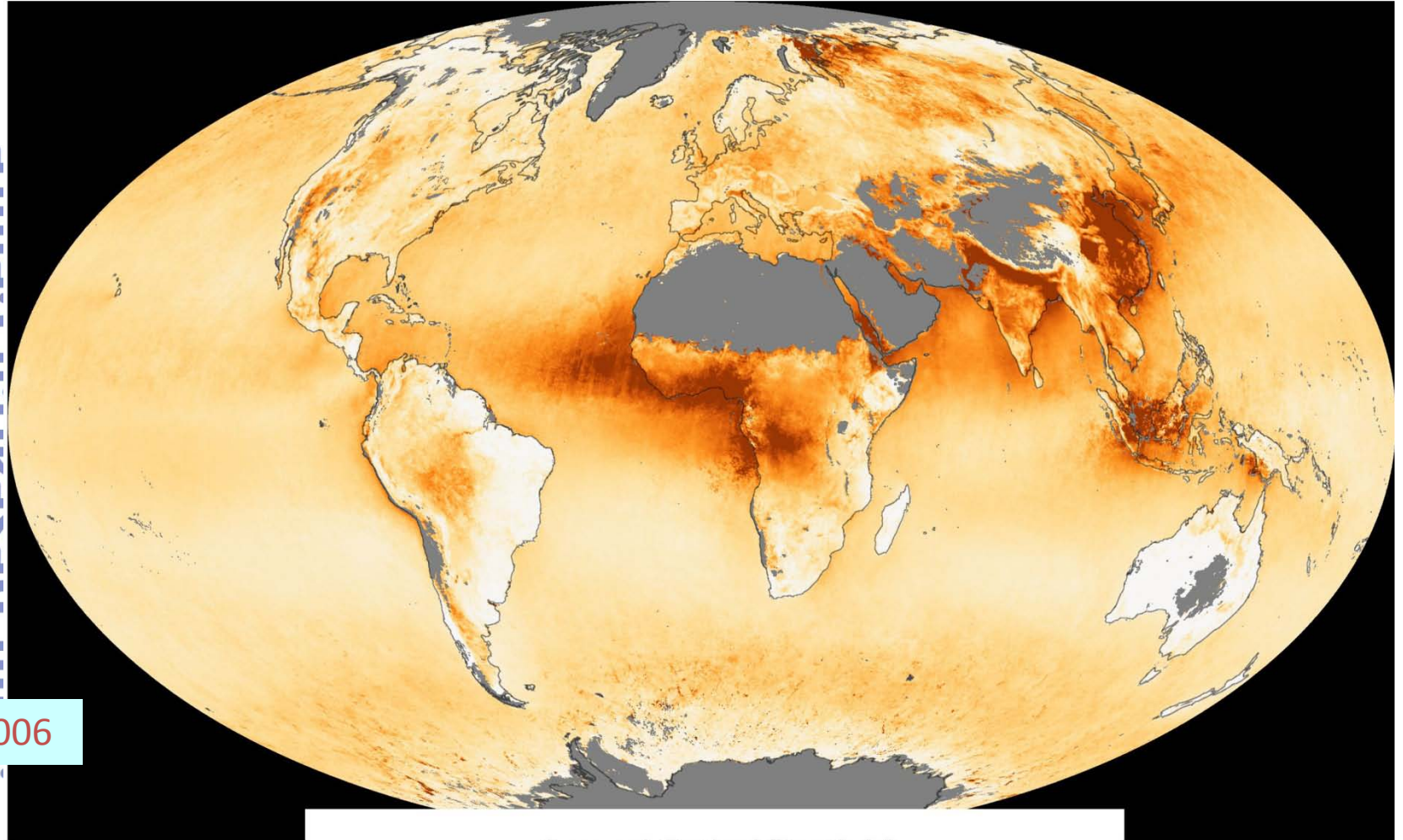


$$RF = (E_{in} - E_{out}) - (E_{in}^{ref} - E_{out}^{ref})$$

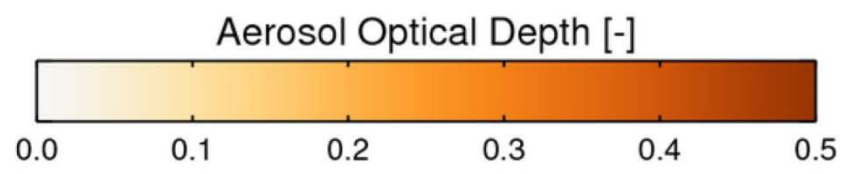
reference = pre-industrial era

hence $(E_{in}^{ref} - E_{out}^{ref}) \approx 0$

and $RF = (E_{in} - E_{out})$



2006



MISR

AeroCom (Schultz et al., 2006) compared instantaneous direct radiative forcing due to anthropogenic aerosols produced by 9 different global models with detailed aerosol modules, using identical emissions.

Differences in computed direct aerosol forcing (range $+0.04$ to -0.41 Wm^{-2}) are due to:

- diversity in simulated aerosol residence times
 - mass extinction coefficients
 - **forcing efficiencies** (forcing per unit optical depth)
- +
- in clear-sky conditions: aerosol absorption, size, and surface albedo
 - in cloud-sky conditions: cloud fields and relative altitudes of absorbing aerosol and clouds



- For any statement that I going to make there is an expert in the room that can go much mre n depth, or even challenge it
- Steve: father if not grand-father: without a beard mor like a father
- When interest of standardization organizations > relevance!
- PM/healt already relevant: legislation > lavel playing field money (even though this community knows much less aerosol, but there is epidemiological evidence!) (do we havedirect evidence of impacts of aerosols on climate:, and that it is important?)
- **How can we go from microscopic compexity to values regional or glonal impacts on radiation balece and climate?**
- **Do we need to consider all that complexity, to come to usefull values?**
- Not yet legislation for aerosols and climate, scientific need (but e.g. BC in CDM is discussed ?) anyway relevance
- Ozone vs PM, PM external internal mixing: onc you ave O3, RF easy to calculate. once you have PM dot easy to calculate
- TOA forcing, RF = delta(present) – delta (pre-industrial) > role of knowing pre-industrial values
 - Overall Earth radiation balance
 - Delta aerosols, delta GHG.delat alsbedl
- - IPCC usual graph (not policy relevant), immediately show other IPCC (more relevant) > Shindell
- At this point usefull to make a distinction between – assessement of present day effects of PM on climate (relevant to monitor compliamce with legilation: standardization) – predicting future impacts of PM on climate (policy development)
- Paper Charlson et al. / Haywood & Shine
 - based on secondary parameters: problem of pre-industrial
 - Based on primar parameters related to emisioins
- What did we learn? Include TOA > TOA+ surface+atmospheric forcing >regional impacts
- We know a lot about aerosols, compared to what health people know or want to consider about PM. Still the latter made it to policy and hence standardization: research becomes different ...
- lbe exampels of standardization work in air quality work
- Two communities are any way mergibg: e.g. UNEP assessement.
 - RF of BC emissions (IGAC, telephone conference India, US, Nairobi, Europe)
 - Effect on Climate, Health Crops. Integrated > **uncertainties are less problematic.**
- Conclusion?