#### **Light Scattering**

When light passes through a medium some of it is directed away from its direction of travel. Any photons that are diverted from their direction of propagation are **scattered**.

In the atmosphere both scattering and absorption occur and their combination is known as EXTINCTION.

If we have a medium containing a number of scattering elements we can define a scattering coefficient,  $k_s$ , analogous to an absorption coefficient  $k_a$ =n $\sigma$  where n is the number of absorbers per unit volume and  $\sigma$  is the absorption cross section.

The intensity of light of wavelength  $\lambda$ , after passing through a medium of thickness L,  $I(\lambda)$ , may be given by  $I(\lambda)=I_0(\lambda)\exp(-k_sL)$ , where  $I_0(\lambda)$  is the Intensity of light entering the medium and  $k_sL$  is the **optical depth**,  $\tau$ .

When both scattering and absorption occur the overall extinction can be defined as  $k_e = k_s + k_a$ . And the intensity can be found again using the Beer-Lambert law:

$$I(\lambda)=I_0(\lambda)\exp(-k_eL)$$
.

When the scattering elements are small compared to the wavelength of light being scattered then the treatment is simpler than in other cases. The analysis was first performed by Lord Rayleigh (1871) to explain why the sky is blue.

As the incident light's e-m field varies the molecules and small particles are continuously redistributing their charges. The field causes the charges to oscillate at the frequency of the radiation and these charges then re-radiate an e-m field at the same frequency as the forcing field. However the emitted radiation will not be propagated in the same direction as the forcing field and will not necessarily have the same polarization.

When equal positive and negative charges are displaced in opposite directions the charge distribution is called a dipole. The dipole moment p is the product of the quantity of displaced charge and the displaced distance. The vector **p** points in the direction of the charge displacement.

If the incident field is  $E_0 \exp(i\omega t)$ , the magnitude of the magnitude of the polarisation is

$$p = \alpha t E_0 \exp(i\omega t),$$

where  $\alpha$  is the polarisability of the particle,  $\omega$  is the angular velocity of the particle and t is time

Light interaction with gas molecules and small particles can be modelled using dipoles.

Let **r** be the unit vector in the direction from the dipole to the observation point and **p** the vector in the direction of the electric field or dipole moment.

The scattered field at a distance r from a dipole is:  $E = \omega \alpha t E_0 \exp(i\omega t)$ ,

$$E = \frac{\omega^2}{c^2 r} \exp\left[i\omega\left(t - \frac{r}{c}\right)\right] r \times (p \times r)$$

Where c is the speed of light and r is large enough to be in the far field of the dipole. The triple product is a vector lying in the plane containing the range vector (NOT  $\mathbf{r}$ ) and the dipole vector  $\mathbf{p}$ . The scattered electric field only has components in this scattered plane.

The vector in the direction of propogation of the incident light and the vector from the dipole to the observation point define a plane. The component of the polarisation vector in this plane is  $\alpha E_{0l} r_l$  and perpendicular  $\alpha E_{0r} r_r$  and  $e_l$  and  $e_r$  are the unit field vectors in these directions.

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If the incident electric field is  $(E_{0r}e_r + E_{0l}e_l)\exp(i\omega t)$ , then the scattered field components at P are

$$\boldsymbol{E_r(r)} = \frac{\alpha\omega^2}{c^2r} \exp\left[i\omega\left(t - \frac{r}{c}\right)\right] E_{0r}\boldsymbol{e_r} \quad \boldsymbol{E_l(r)} = \frac{\alpha\omega^2}{c^2r} \exp\left[i\omega\left(t - \frac{r}{c}\right)\right] E_{0l} \cos\theta\left(\boldsymbol{e_r} \times \boldsymbol{r}\right)$$

The intensity of the scattered light assuming that the incident light is unpolarised is

$$I = \frac{\alpha^2 \omega^4}{c^4 r^2} \left( 1 + \cos^2 \theta \right) I_0$$

If the scatterer is a sphere of radius a and dielectric constant K, its polarisability is given as  $[(K-1)(K+2)]a^3$ . So we can write:

$$I = \frac{a^6 \omega^4}{c^4 r^2} \left( \frac{m^2 - 1}{m^2 + 2} \right) \left( 1 + \cos^2 \theta \right) I_0$$

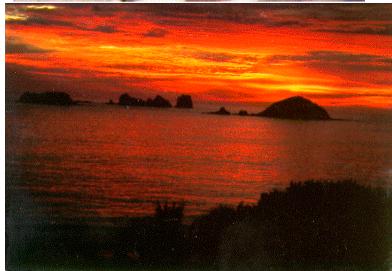
- The angular distribution of the scattered intensity varies from 2 in the forward and backward directions to 1 at 90°, where the 1 component vanishes and the scattered radiation is plane polarised transverse to the scattering plane.
- The scattering intensity and energy removed from the incident beam is proportional to the sixth power of the radius (or for nearly spherical particles to the square of the volume).
- The scattered intensity and energy removed from the incident beam is proportional to  $\omega^4$  or to  $\lambda^{-4}$ .
- The intensity of Rayleigh scattering is proportional to the sixth power of the radius. The absorption, however, is proportional to the volume. As a result scattering falls off more rapidly than absorption, or put another way the **single scattering albedo** decreases with decreasing particle size.

The sky is blue as blue light is scattered out of the direct solar beam much more efficiently than longer wavelengths.

The horizon appears whiter than the zenith because the increased path leads to increased scattering.

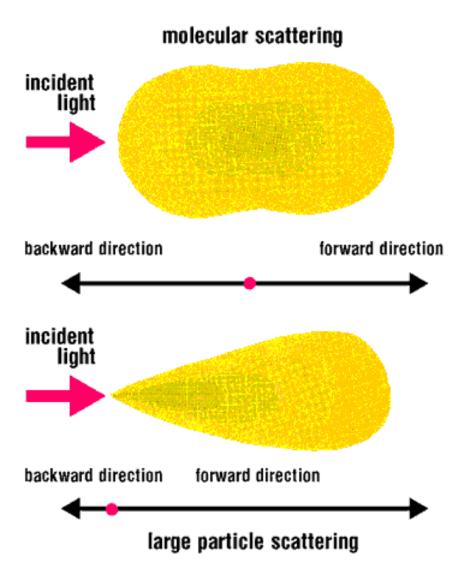
During sunrise and sunset blue and yellow light is efficiently removed from the direct beam over the very long atmospheric paths at the high solar zenith angles leading to spectacular red skies.





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## **The Direction of Scattering**



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#### **Mie Scattering 1**

If the scatterers are large compared to the wavelength of light then geometric optics provide a good approximation. However, in most atmospheric situations aerosols are neither large nor small enough to be treated simply.

The size parameter,  $\alpha = 2\pi a/\lambda$ , is between 1 and 20.

This problem was first solved by Gustav Mie (1908) by determining the wave vector in spherical coordinates for e-m waves, specified by Maxwell's equations. In general particles absorb as well as scatter and Mie gave solutions for the absorption, scattering and extinction cross sections as a function of the scattering angle.

A function for the extinction efficiency factor,  $Q_{\rm ext}$ , for spheres of refractive index n in a medium of unity refractive index can be derived using Mie theory which describe the efficiency with which light is scattered as a function of the size parameter.

#### **Mie Scattering 2**

 $Q_{ext}$  is independent of particle size and can be related to the extinction cross section,  $C_{ext}$  by  $Q_{ext} = C_{ext} / \pi r^2$ , where r is the particle radius.

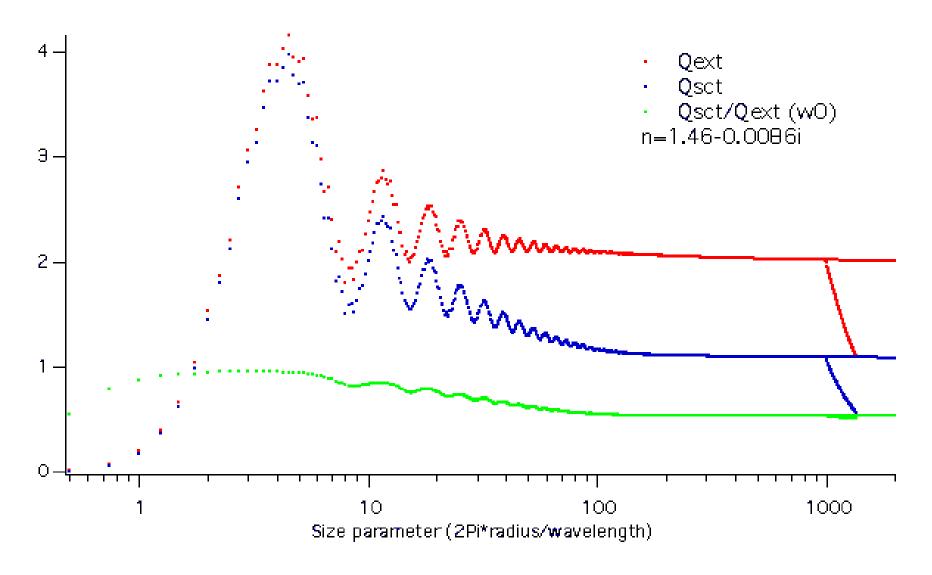
Similar relationships can be formed for  $Q_{scat}$  and  $Q_{abs}$ , the scattering and absorption coefficients.

The refractive index, n, is given as a complex number where the real part is the scattering component and the imaginary component is the absorptive component.

We can define the single scattering albedo,  $\omega_0$ , as the fraction of light scattered by a particle to the total extinction caused by it:

$$\omega_0 = Q_{\text{scat}}/Q_{\text{ext}} = Q_{\text{scat}}/(Q_{\text{scat}} + Q_{\text{abs}}) = C_{\text{scat}}/C_{\text{ext}} = C_{\text{scat}}/(C_{\text{scat}} + C_{\text{abs}}).$$

### **Mie Scattering 3**



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So far we have only considered a single particle interacting with radiation in isolation. What we want to know and have to measure is how a bulk air parcel, containing many aerosol particles interacts.

If the aerosol number concentration is very large then the scattering is very complex as radiation undergoes many interactions in a volume. However, in practice the atmosphere, even at its most polluted, has a relatively small number concentration. Even, in highly polluted environments, where the number concentration may reach 10<sup>6</sup> particles cm<sup>-3</sup> the particles only occupy approximately 10<sup>-6</sup> of the volume.

This means that we can approximate the problem by only considering single scattering.

Given this we can assume that the total scattered intensity is the sum of the intensities scattered by each of the particles.

Now the ensemble scattering, absorption and extinction coefficients  $(k_s, k_a, and k_e)$  are functions of particle diameter  $(D_p)$ , refractive index (n), and wavelength of incident light  $(\lambda)$ . If we assume that the entire particle population has the same refractive index, then  $D_p^{max} = D_p^{nax}$ 

 $k_e(\lambda) = \int_{0}^{D_p^{\text{max}}} \frac{\pi D_p^2}{4} Q_{ext}(n, \alpha) N(D_p) dD_p$ 

Where  $N(D_p)$  is the number concentration of particles of diameter  $D_p$ . From here on we assume, unless otherwise stated that  $k_e$  is dependent on wavelength.

Similar expressions can be written for the scattering and absorption coefficients.

And 
$$k_e = k_s + k_a.$$

So at a given wavelength, the intensity, I, at a distance z through a scattering and absorbing layer can be related to the initial intensity,  $I_0$  by the Beer Lambert Law:

$$I=I_0 \exp(-k_e z) = I_0 \exp(-\tau)$$

Where  $\tau$  is the optical depth of the aerosol layer.

Often these coefficients are expressed in terms of the aerosol mass distribution function  $M(D_n)$ :

$$M(D_p) = \rho_p \frac{\pi D_p^3}{6} N(D_p)$$

The result is:

$$k_e = \int_0^{D_p^{\text{max}}} \frac{3}{2\rho_p D_p} Q_{ext}(n, \alpha) M(D_p) dD_p$$

Which can be written:

$$k_e = \int_{0}^{D_p^{\text{max}}} E_{ext}(D_p, n, \lambda) M(D_p) dD_p$$

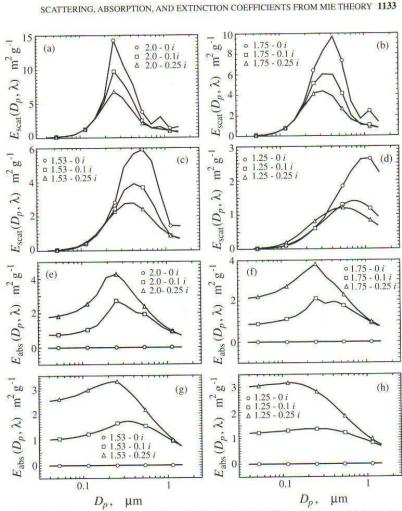
Where  $E_{ext}(D_p, n, \lambda)$  is the **mass extinction coefficient**. Likewise  $E_{scat}$  and  $E_{abs}$ :

$$E_{abs}(D_p, n, \lambda) = \frac{3}{2\rho_p D_p} Q_{abs}(n, \alpha) \qquad E_{scat}(D_p, n, \lambda) = \frac{3}{2\rho_p D_p} Q_{scat}(n, \alpha)$$

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The figure shows how E<sub>scat</sub> and E<sub>abs</sub> vary as a function of particle size over a range of refractive indices. Note that for any real part (scattering) of n, an increase in the imaginary (absorption) part acts to decrease the scattering intensity and shift the peak to smaller sizes. *More of the photon energy is being lost to absorption*. Also at larger n, E<sub>scat</sub> increases over all sizes.

Increasing the imaginary part of n increases  $E_{abs}$  overall and where the real part of n is small  $E_{abs}$  remains close to its maximum well below  $D_p=100$  nm. i.e.  $E_{ext}$  at low  $D_p$  is entirely due to absorption (e.g. diesel exhaust emissions).



**FIGURE 22.8** Mass scattering (a–d) and absorption (e–h) efficiencies for materials having refractive indices m = n + ik: n = 2.00, 1.75, 1.53, 1.25, and k = -0, -0.1, -0.25 (Note that m = 1.53 - 0k is the refractive index of (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>.)

Consider a direct solar beam impinging on an aerosol layer. Assume that the sun is directly overhead and is at the zenith. The fraction of the incident beam that is transmitted directly through the layer is e<sup>-τ</sup>.

We can define an upscatter fraction,  $\beta$ , which is the fraction of the scattered radiation that is in the upward hemisphere relative to the local horizon. It is the upscatter fraction that is important in this context not the backscatter relative to the incident beam, though the two are the same in our simple model.

The non transmitted fraction is therefore 1-  $e^{-\tau}$ .

Comprising the scattered fraction:  $\omega(1-e^{-\tau})$  and the absorbed fraction  $(1-\omega)(1-e^{-\tau})$ .

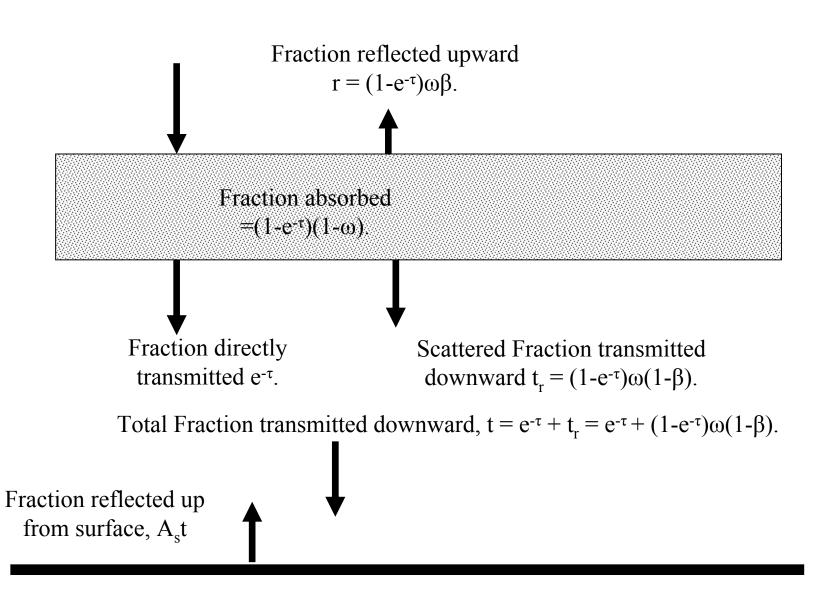
Of the scattered fraction:

 $r = \omega \beta (1 - e^{-\tau})$  is the fraction reflected upwards

 $t_r = \omega(1-\beta)(1-e^{-\tau})$  is the scattered fraction transmitted downwards

The total fraction of radiation transmitted downwards is:  $t = e^{-\tau} + t_r = e^{-\tau} + \omega(1-\beta)(1-e^{-\tau})$ 

If the albedo of the underlying surface is  $A_s$ , then the fraction of the radiation incident on the surface that is reflected is  $tA_s$ .



Now of the fraction of radiation reflected of the surface,  $A_s$ t, back into the aerosol layer, some of the radiation is absorbed, some transmitted and some scattered (in both the up and down directions).

On the first downward pass we saw that the fraction transmitted was t.

So on the first upward pass the fraction transmitted through the aerosol layer is also t, which in terms of the fraction of the original is  $A_s t^2$ 

If we turn the picture upside down in our mind then the fraction of the beam from the Earth reflected back down towards the Earth by the layer is just r.

In terms of the fraction of the incident flux this is rA<sub>s</sub>t.

In turn the downwards reflected beam is itself reflected off the surface, giving an upwards return of  $rA_s^2t^2$ .

So for two complete passes the total upward reflected flux,  $F_{r,2}$  is:

$$F_{r,2} = rF_o + A_s t^2 F_0 + rA_s^2 t^2 F_0.$$

This can continue.....

$$F_{r,n} = (r + A_s t^2 + r A_s^2 t^2 \dots) F_0 = [r + A_s t^2 (1 + r A_s \dots) F_0]$$

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The total upward reflected shortwave flux is then:

$$F_r = [r + A_s t^2 (1 + rA_s + r^2 A_s^2 + r^3 A_s^3 + ....) F_0.$$

As  $A_s$  and r are both less than 1, the series can be summed to give:  $1/(1-A_s r)$ 

So 
$$F_r = [r + t^2 A_s/(1 - A_s r)]F_0 = A_t F_0$$
.

Where  $A_t$  is the total reflectance of the aerosol-surface system.

The change in the reflectance as a result of the presence of an aerosol layer is then:

$$\Delta A_a = A_t - A_s = [r + t^2 A_s / (1 - A_s r)] - A_s.$$

We have so far assumed that the atmosphere itself does not scatter or absorb, yet we have already seen that this is not the case.

To a first approximation we can assume that the atmospheric extinction takes place above the aerosol layer, hence the flux on top of the aerosol layer is,  $T_aF_0$ , not  $F_0$ .

The total upward flux can now be written as:  $T_a^2F_r = [r + t^2A_s/(1-A_sr)](T_aF_0)T_a$ 

The term  $(T_aF_0)$  is the fractional transmittance to the top of the layer and the second  $T_a$  on the right hand side is the fractional transmittance of the upwelling flux leaving the layer before exiting the atmosphere.

So in summary

The change in the planetary albedo is:  $\Delta A_a = A_t - A_s = T_a^2[r + t^2A_s/(1-A_sr) - A_s]$ 

And the change in the *outgoing* radiative flux as a result of an aerosol layer underlying an atmospheric layer is:

$$\Delta F = F_0 T_a^2 [r + t^2 A_s / (1 - A_s r) - A_s]$$

The quantity  $\Delta F > 0$  if  $\Delta A_a$  is positive.

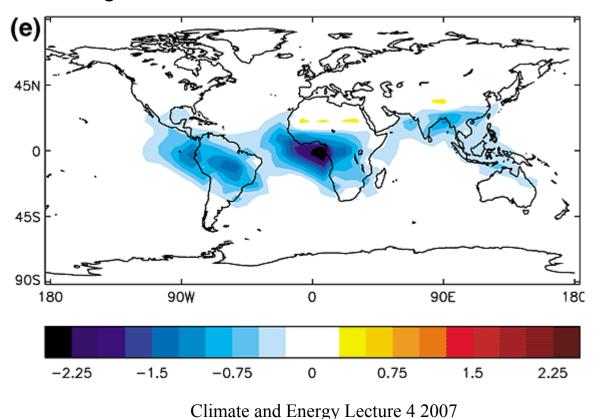
When interpreted in terms of climate forcing (see later) we are concerned with the *incoming* flux so the change in forcing is  $-\Delta F$ .

 $\Delta F$  depends on:

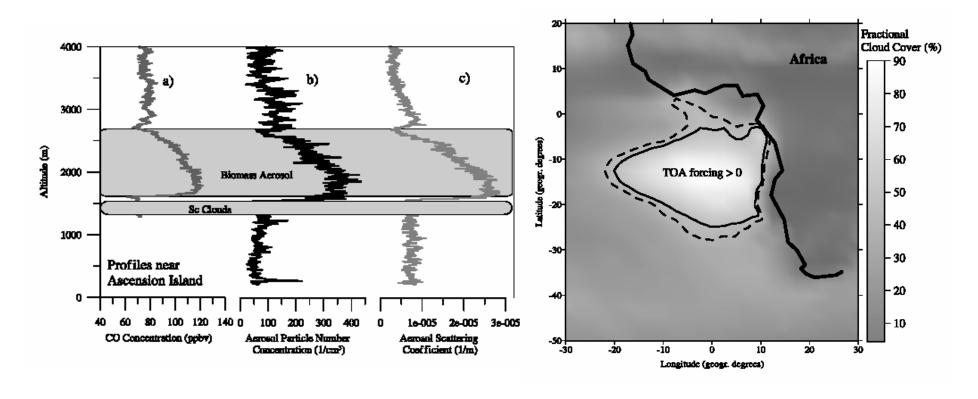
The incident solar flux  $(F_0)$ ; the transmittance of the atmosphere  $(T_a)$ ; the albedo of the underlying surface  $(A_s)$ ; the single scattering albedo of the aerosol  $(\omega)$ ; the upscatter fraction of the layer  $(\beta)$  and its optical depth  $(\tau)$ .

The problem is that  $\omega$  depends on the aerosol size distribution, its chemical composition and is wavelength dependent; B depends on aerosol size (larger particles scatter more in the forward direction) and composition; and  $\tau$  depends largely on the mass of the aerosol.

Biomass burning is widespread over southern Africa during the dry season. Large quantities of biomass are advected off the west coast of southern Africa over the south Atlantic Ocean. Global models such as Penner et al. 1998 have considered the aerosol as partially absorbing but essentially independent of other features in the GCM so, aerosol over a dark ocean increases scattering back to space. Even though the aerosol is partially absorbing, the sea surface is darker and the net effect is a cooling. (The scale is in Wm<sup>-2</sup>) and is the predicted forcing due to the aerosol layer, a net cooling.



However, in that part of the world, a large sheet of low lying strato cumulus cloud is semi-persistent. Such clouds have high albedos (0.8 is not uncommon). As the biomass burning aerosol are lofted over the continent they have been observed by Haywood et al. 2003 during the SAFARI campaign to exist above the cloud deck (see example aircraft profile data below). The right hand panel shows the expected Top Of the Atmosphere forcing is predicted to be positive (a warming) if such absorbing aerosol is included above the cloud deck.



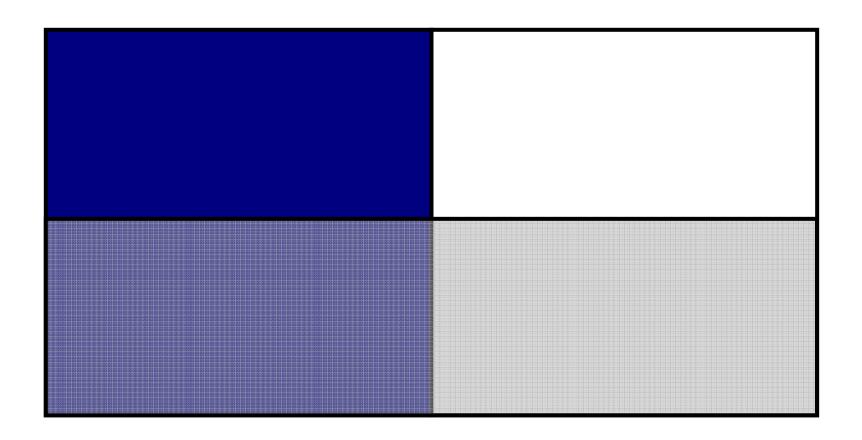
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Even in our simple model we can demonstrate this by including typical values measured during the SAFARI campaign.  $F_0$  is essentially midday sun at 60 degrees Solar zenith angle, the albedo of the sea is 0.1 and cloud is set at 0.8. The single scattering albedo and the optical depth used are from measurements made with the UK C130 aircraft during SAFARI.

	Ocean	Land
Incident solar flux (F <sub>0</sub> ) Wm <sup>-2</sup>	670	670
Transmittance of the atmosphere (T <sub>a</sub> )	0.49	0.49
Albedo of the underlying surface (A <sub>s</sub> )	0.1	0.8
Single scattering albedo of the aerosol ( $\omega$ )	0.89	0.89
Upscatter fraction of the layer (β)	0.2	0.2
Optical depth $(\tau)$	0.45	0.45
$\Delta A$	0.011	-0.014
$\Delta F (Wm^{-2})$	7.3	-9.5

The figures on the left do not include cloud and show that there is a net loss of radiation to space from the TOA. However, once the cloud is included there is a net increase in radiation and a positive forcing. These simple results agree reasonably well with much more complex radiation modelling using satellite information and other constraints by Keil and Haywood (2003).

# Light, high Albedo Cloud



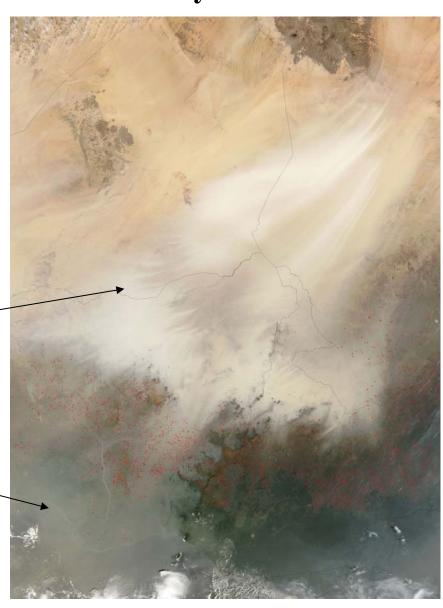
Next January and February a joint UK Met Office (Haywood) and UK university (U of Manchester) experiment are to investigate the interaction between biomass burning and dust in the northern Sahel Savanna. The image is from the MODIS satellite and shows the large areas of dust advected south and west from the Sahara and the main areas of biomass burning in the region and its associated haze.

Dust outflow

Regional biomass haze

Red dots indicate main fires

MODIS image January 5<sup>th</sup> 2005



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#### Cooling versus heating of an aerosol layer

The sign of the change in planetary albedo determines whether the forcing is negative (cooling effect) or positive (warming)

The key parameter is the single scattering albedo,  $\omega$ .

The value of  $\omega$  at which  $\Delta A_a = 0$  defines the boundary between cooling and heating. The value of  $\omega$  is critical, over dark surfaces, such as oceans, nearly show negative forcing as they increase the flux out to space above that from the darker surface regardless of how much absorption occurs. However over a high albedo surface such as snow or a desert, absorption by the particles can reduce the solar flux reflected from the surface to result in a net reduction of flux to space, or a positive forcing.

We can use our model to derive an expression for  $\omega$  when  $\Delta A_a = 0$ . We will assume, as is the case for most regions that the aerosol optical depth is about 0.1 and so apply  $\tau << 1$  as an approximation.

So 
$$r = (1-e^{-\tau})\omega\beta \sim \tau\omega\beta$$
 and  $t = e^{-\tau} + \omega(1-\beta)(1-e^{-\tau}) \sim 1 - \tau + \omega(1-\beta)\tau$ 

#### Cooling versus heating of an aerosol layer 2

So 
$$r = (1-e^{-\tau})\omega\beta \sim \tau\omega\beta$$
 and  $t = e^{-\tau} + \omega(1-\beta)(1-e^{-\tau}) \sim 1 - \tau + \omega(1-\beta)\tau$ 

From previously:  $\Delta A_a = A_t - A_s = [r + t^2 A_s/(1 - A_s r)] - A_s$ .

So: 
$$\Delta A_a \approx \tau \omega \beta + \frac{\left[1 - \tau + \omega (1 - \beta)\tau\right]^2 A_s}{1 - A_s \tau \omega \beta} - A_s$$

By rearrangement and neglecting terms involving  $\tau^2$  we get:

$$\Delta A_a \approx \tau \omega \beta + \left[ (1 - A_s)^2 - \frac{A_s}{\beta} \left( \frac{1}{\omega} - 1 \right) \right]$$

We can also neglect  $\tau\omega\beta$  and only treat the second term:

Hence  $\Delta A_a = 0$  when  $\omega$  satisfies:

$$\omega_{crit} = \frac{2A_s}{2A_s + \beta(1 - A_s)^2}$$

Values of  $\omega > \omega_{crit}$  lead to cooling as  $\Delta A_a > 0$ 

### Cooling versus heating of an aerosol layer 3

The figure shows the critical single scattering albedo ( $\omega_{crit}$ ) as a function of the surface albedo ( $A_s$ ) and the upscatter fraction ( $\beta$ ).

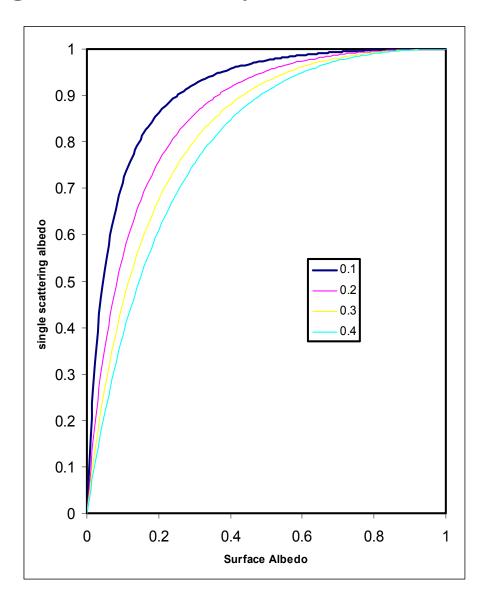
This value is practically independent of the optical depth.

A global mean  $A_a$  is approx. 0.15 and for  $\beta \sim 0.29$ , the  $\omega_{crit} = 0.6$ 

As the surface albedo approaches 1 the critical value of  $\omega$  also approaches 1, independently of the value of  $\beta$ .

At high values of  $A_a$  the total reflectance of the aerosol-surface system is large to begin with and even a small amount of aerosol absorption leads to a heating effect.

At values of  $A_a$  approaching 0 only a small amount of aerosol scattering is required to produce a cooling effect on the aerosol layer



# Export of Dust

7 April 2001

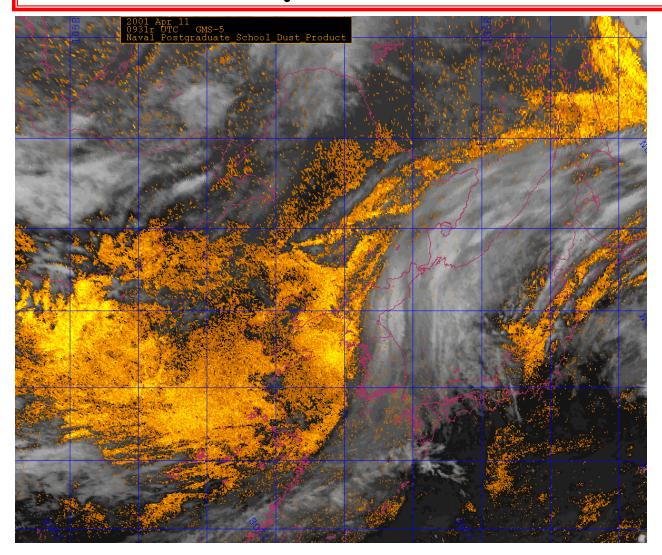
8 April 2001





These photos are reduced-resolution versions of photos taken by Dr. Zev Levin while visiting Baicheng, Jilin Province, China (NE of Beijing) during the dust storm.

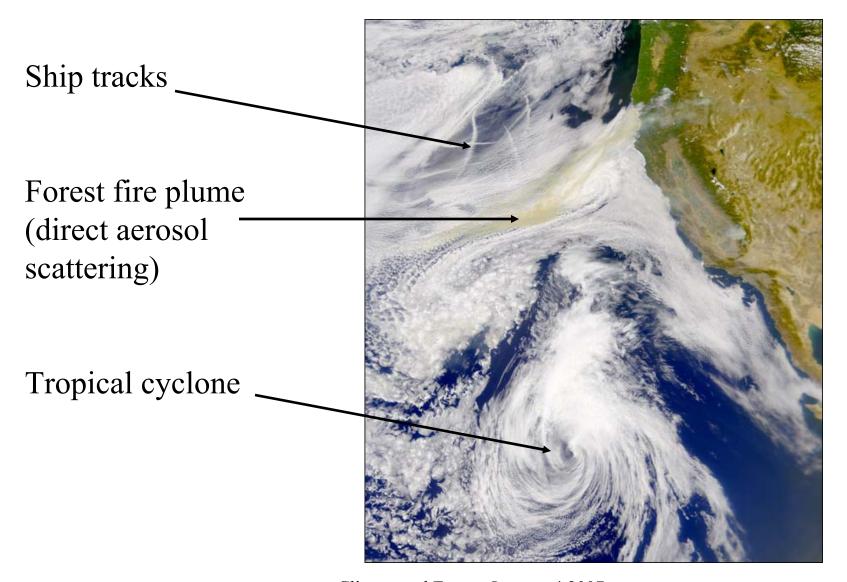
# Export of Dust



Naval Postgraduate School Dust Product, 11 April 2001

Image by Phil Durkee, NPGS

#### The Direct and Indirect Effects



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#### THE INDIRECT EFFECT - "Ship Tracks"

The optical depth of a cloud,  $\tau$ , is the product of the scattering coefficient,  $k_s$ , and the geometric depth of the cloud, h. The larger the scattering coefficient the greater the fraction of incident light scattered per unit depth of cloud.

 $\tau$  can be approximated by  $\tau$ =2πhr<sup>2</sup>N, where r is the droplet radius and N is the number concentration.

The liquid water content of the cloud, L, is  $4/3\pi r^3 N\rho$  where  $\rho$  is the density of water, 1 g cm<sup>-3</sup>)

In the background: 
$$\tau_b = 2\pi h r_b^2 N_b$$
 and  $L_b = 4/3\pi r_b^3 N_b \rho$ 

In the ship plume: 
$$\tau_s = 2\pi h r_s^2 N_s$$
 and  $L_s = 4/3\pi r_s^3 N_s \rho$ 

If we assume that the cloud droplets do not absorb light.

$$L_s = L_b \text{ so } N_b / N_s = r_s^3 / r_b^3 \text{ and } \tau_s / \tau_b = r_b / r_s. \qquad r_s < r_b \text{ so } \tau_s > \tau_b$$

#### **Bottom Line**

To reduce uncertainty in climate change predictions we need to get the errors in estimating aerosol and cloud scattering absorption effects identified and reduced.

Without doing so, we will be unable to assess future temperature changes to the planet