# The urban / background mix Fates, lifetimes, aging, climatic impact

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**NEW ZEALAND BRANCH SEMINAR**
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# Urban outflow, transport and global climate impacts of aerosol

- How aerosol ages (the basic model)
- How background & urban interact – what do we see?
- Heterogeneous Chemistry (gas – aerosol reactions)
- Aerosol-Cloud Interaction (CCN, ship tracks)
- Effects on climate and Earth system (IPCC)

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Marine - **Mike**

Mechanical

Biogenic etc

…up to the urban edge

The aging process - **Mike**
LRT and aging
Effects on climate (and other atmospheric process)
Heterogeneous chemistry, clouds and other processes

# The aging process - **Mike**

# The aging process - **Mike**

- What's involved?
- **Mingling and mixing**
- **Physical aging** – Evolution of size spectrum
- **Chemical aging**
  - Oxidation $\dot{O}H, O_3$ reactions especially for biogenic carbon aerosols
  - Heterogeneous reactions (gas – aerosol processes)
  - Aerosol-Cloud interaction

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Mixing and mingling -

# Particle life cycles

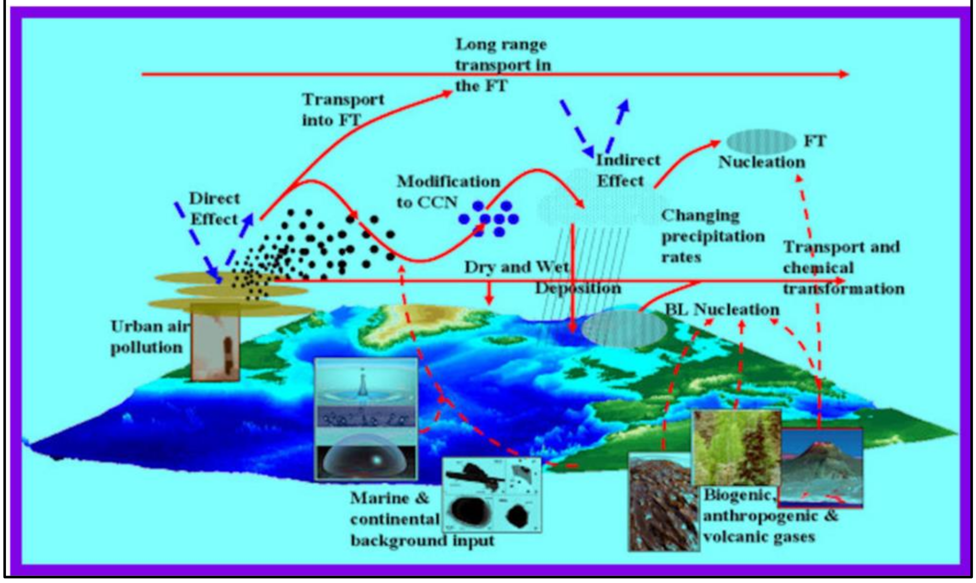
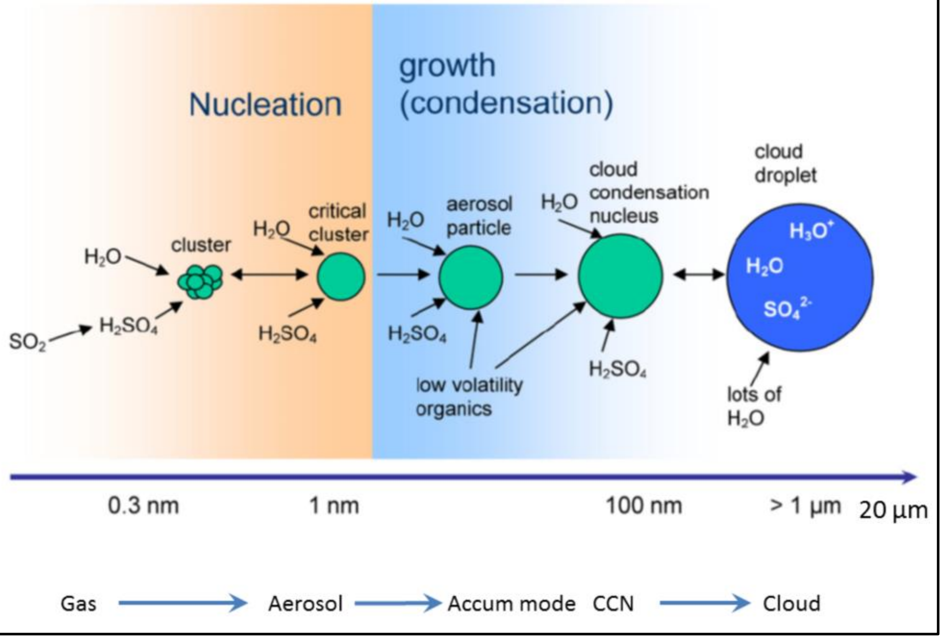
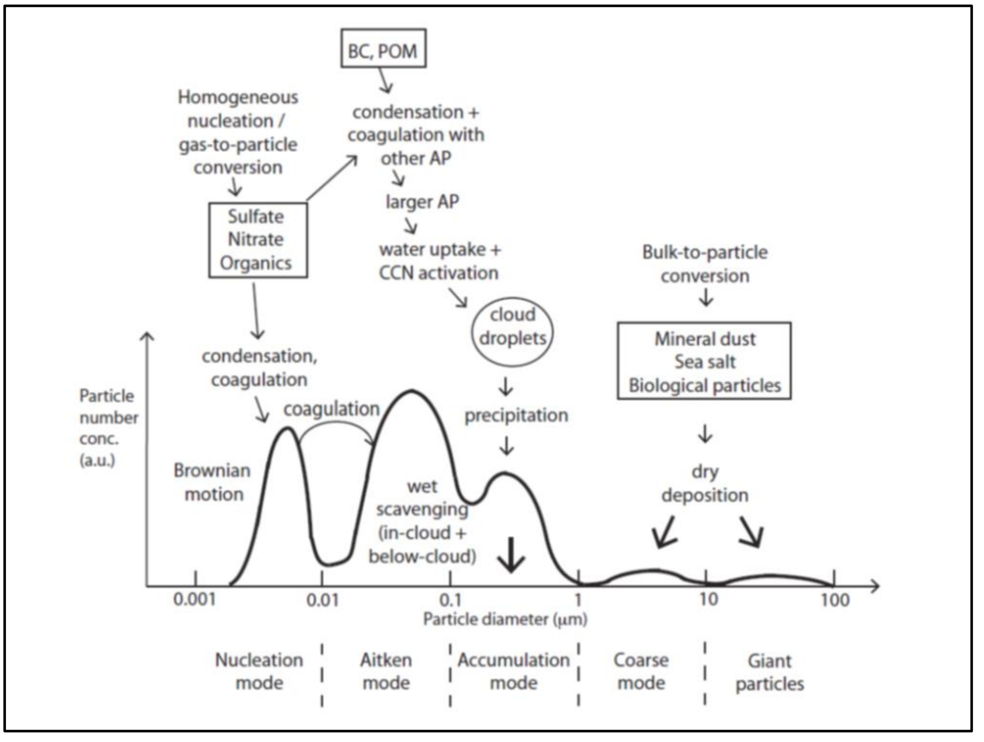


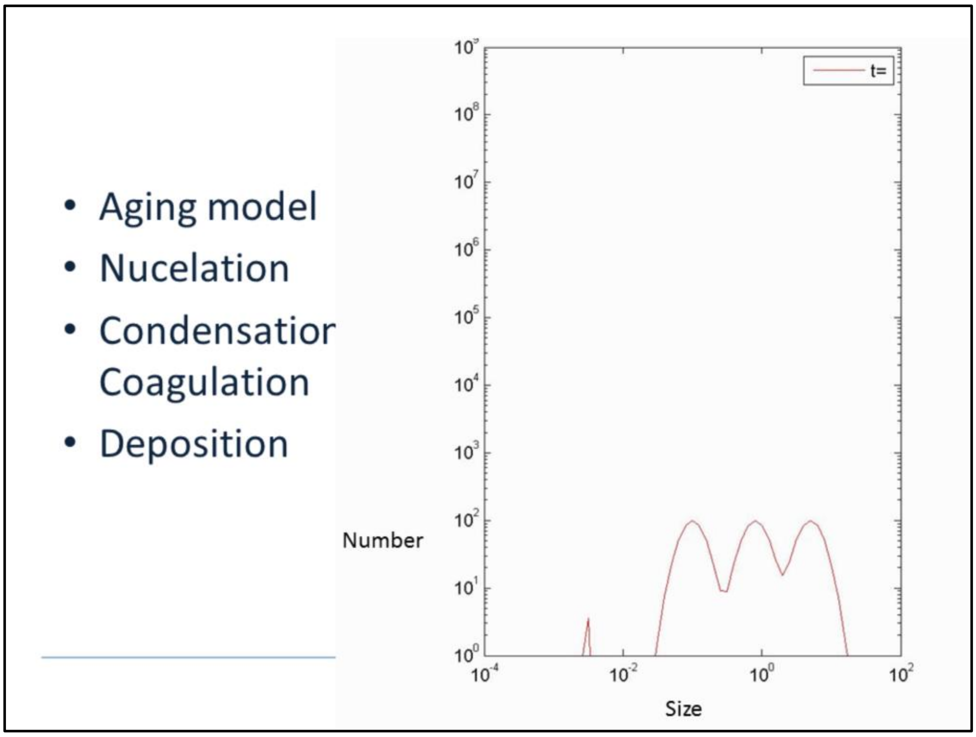

Diagram of the life cycle of aerosol in the atmosphere, showing emmission, deposition and transport processes and the action of aerosol while in the atmosphere

Contact: Jocelyn Duffy, jhduffy@andrew.cmu.edu, 412-268-9982

# Evolution of size

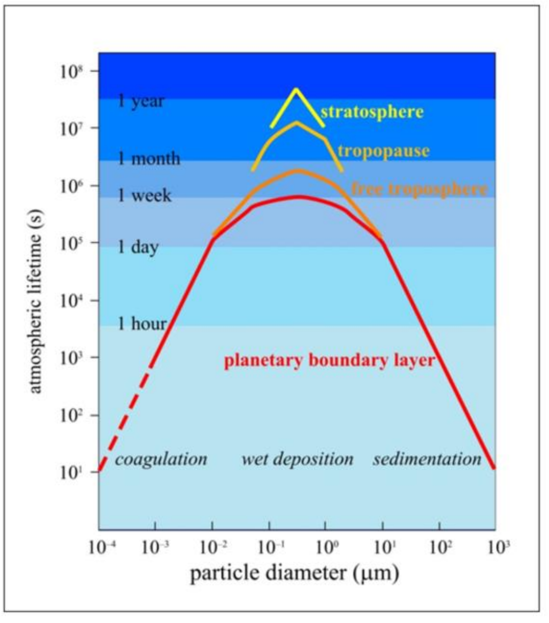



BC, POM
Homogeneous nucleation / gas-to-particle conversion
condensation + coagulation with other AP
larger AP
water uptake + CCN activation
Sulfate Nitrate Organics
cloud droplets
Bulk-to-particle conversion
Mineral dust Sea salt Biological particles
condensation, coagulation
Particle number conc. (a.u.)
coagulation
precipitation
Brownian motion
wet scavenging (in-cloud + below-cloud)
dry deposition
0.001
0.01
0.1
1
10
100
Particle diameter (μm)
Nucleation mode
Aitken mode
Accumulation mode
Coarse mode
Giant particles




Simple model of aerosol evolving towards accumulation mode size due to physical processes of condensation, coagulation and deposition

# Lifetime




Course dust 10 – 100 μm:
1d to 15 min

Fine particles – accumulation mostly removed in rain & clouds

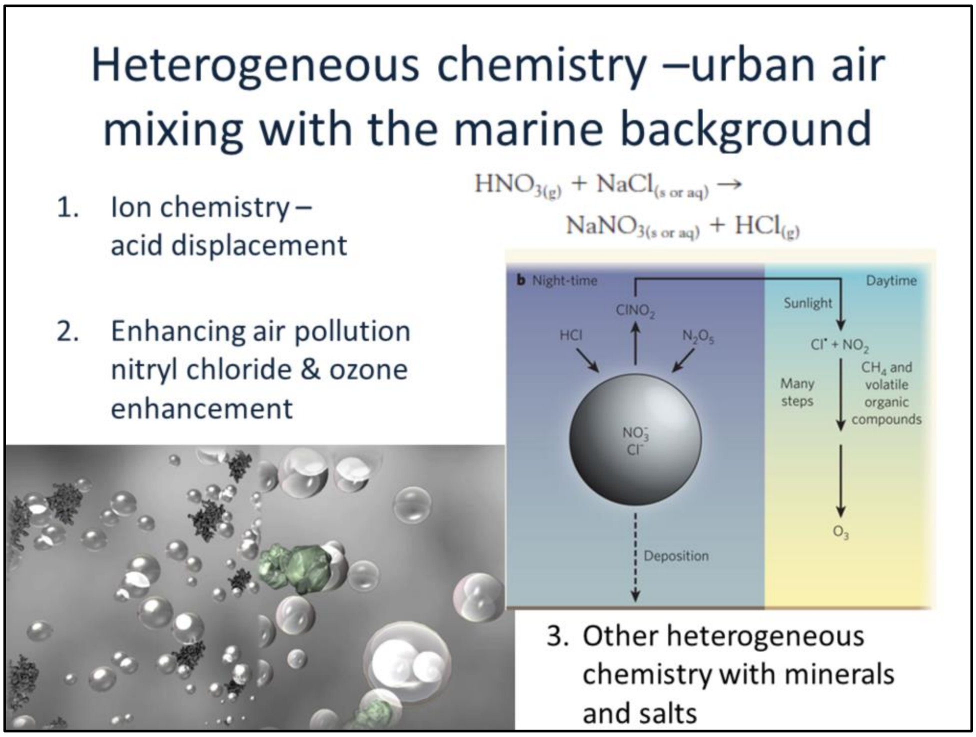
Above the PBL surface layer:
3 to 5 days

When aerosol lofted to 10 km +:
~ a year or more

Jaenicke, 1980

It is not just size but height in the atmosphere that determines aerosol lifetime

Effects of aging. Organic aerosols from different sources undergo chemical processing and mix with each other as well as with inorganic aerosols. This leads to a convergence in their chemical and optical properties and in their ability to nucleate cloud droplets.



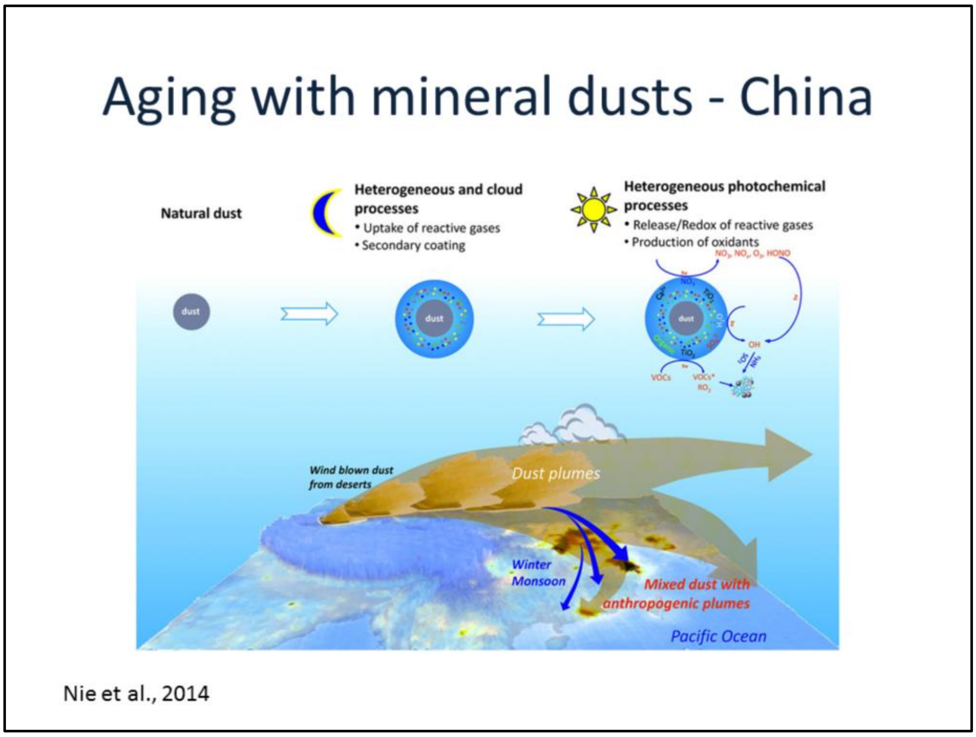

Conventional picture of aerosol uptake of hydrochloric acid (HCl) and dinitrogen pentoxide ($N_2O_5$), leading to chloride ($Cl^-$) and nitrate ($NO^-{}_3$) ions, respectively. In the absence of further reactions both are eventually deposited on the ground with the aerosol particle. **b**, Mechanism supported by the measurements of Thornton *et al*.[1]: nitryl chloride ($ClNO_2$) is formed at night in the reaction of $N_2O_5$ on particles containing chloride. The next day, sunlight breaks $ClNO_2$ into chlorine radicals ($Cl^\bullet$) and nitrogen dioxide ($NO_2$). The chlorine radicals can then attack the greenhouse gas methane ($CH_4$) and volatile organic compounds, and the resulting peroxy radicals and $NO_2$ lead to the formation of ozone ($O_3$).

What happens when air pollution meets sea-salt

For nitryl chloride hypothesis see von Glasow, R. (2008) Atmospheric chemistry: Pollution meets sea salt. *Nature Geosci*, 1(5): 292-293. http://dx.doi.org/10.1038/ngeo192

And subsequent publications

eg: Thornton, J.A., Kercher, J.P., Riedel, T.P., Wagner, N.L., Cozic, J., Holloway, J.S., Dubé, W.P., Wolfe, G.M., Quinn, P.K., Middlebrook, A.M., Alexander, B., Brown, S.S. (2010) A large atomic chlorine source inferred from mid-continental reactive nitrogen chemistry. *Nature*, 464(7286): 271-274. http://www.nature.com/nature/journal/v464/n7286/suppinfo/nature08905_S1.html

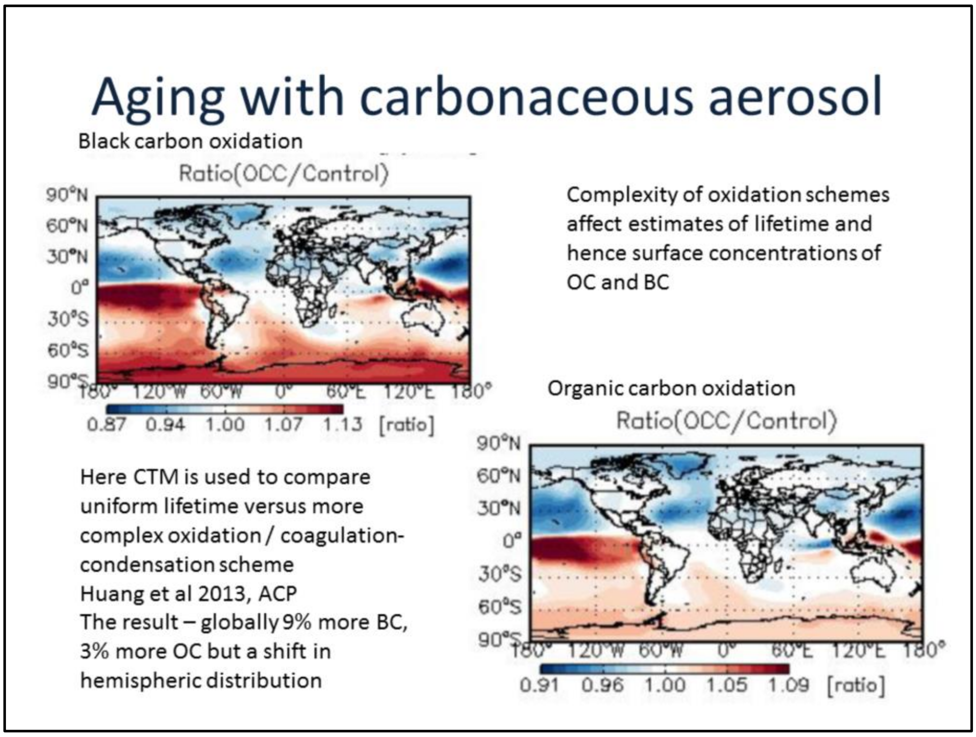



Nie, W., Ding, A., Wang, T., Kerminen, V.-M., George, C., Xue, L., Wang, W., Zhang, Q., Petäjä, T., Qi, X., Gao, X., Wang, X., Yang, X., Fu, C., Kulmala, M. (2014) Polluted dust promotes new particle formation and growth. *Sci. Rep.*, 4. 10.1038/srep06634
http://www.nature.com/srep/2014/141016/srep06634/abs/srep06634.html#supplementary-information

Stage I-Mineral dusts are injected into the atmosphere in the remote area.

Stage II - dust particles uptake anthropogenic reactive gases and form secondary coatings after them transported over area with high anthropogenic emissions. Heterogeneous photochemical processes should play a role in this stage, but easily be covered up by strong gas phase photochemistry.

Stage III - aged dusts transport to remote Asia-Pacific region, where the plumes experience heterogeneous photochemical reactions favoring the new particle formation and growth. The map in the figure was drawn by Global Mapper.




doi:10.5194/acp-13-6329-2013

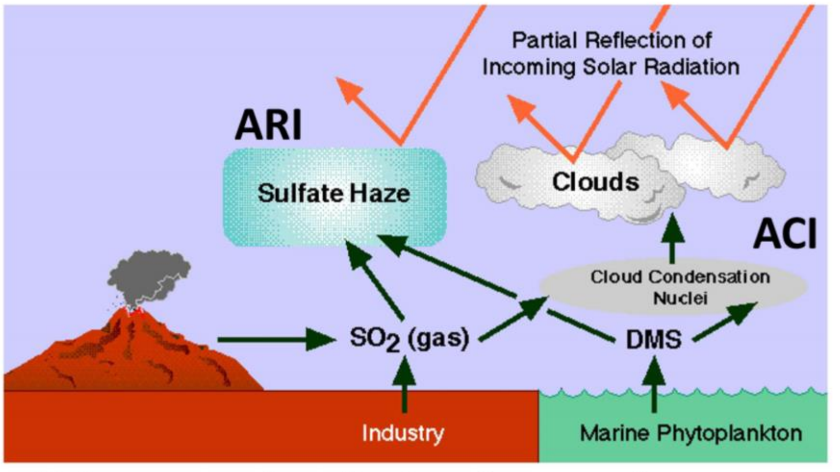
Huang, Y., Wu, S., Dubey, M.K., French, N.H.F. (2013) Impact of aging mechanism on model simulated carbonaceous aerosols. *Atmos. Chem. Phys.*, 13(13): 6329-6343. 10.5194/acp-13-6329-2013

A mix of oxidation and coagulation/condensation aging is considered

# Climatic impacts

# THE "WHITEHOUSE EFFECT"

RADIATIVE FORCING OF CLIMATE CHANGE BY AEROSOLS

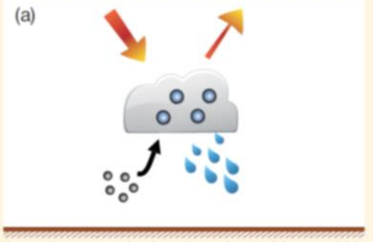



**ARI - aerosol radiation interaction**

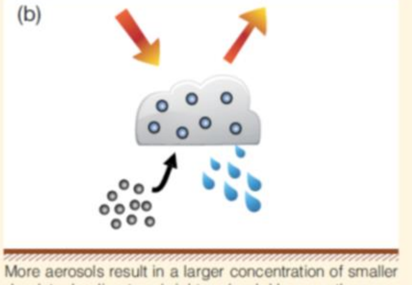
**ACI - aerosol cloud interaction**

# What is "aerosol – cloud interaction"

**Aerosol-cloud interactions**

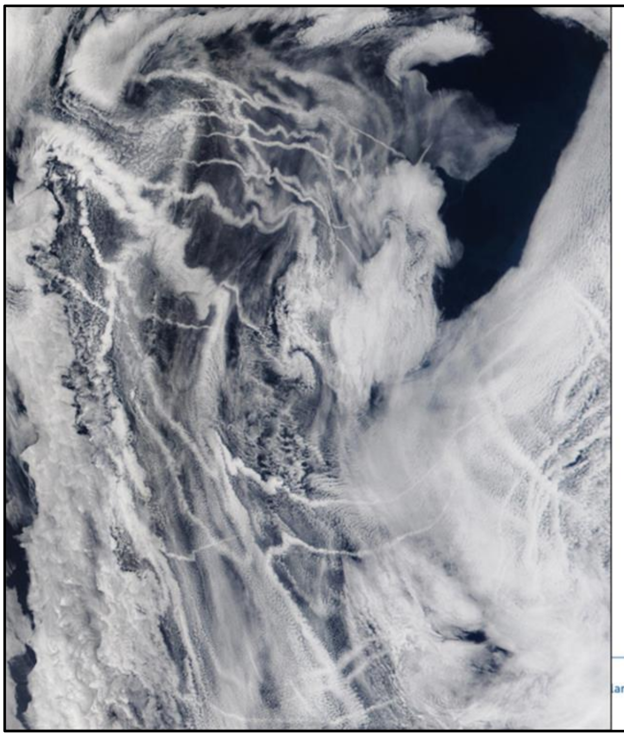



Aerosols serve as cloud condensation nuclei upon which liquid droplets can form.

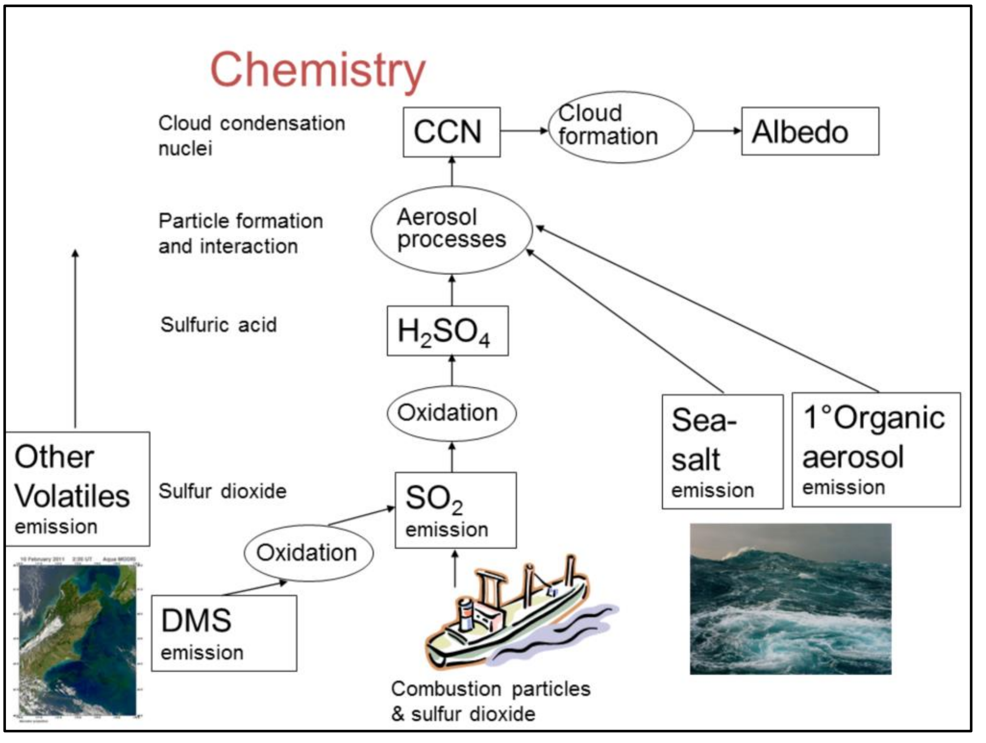


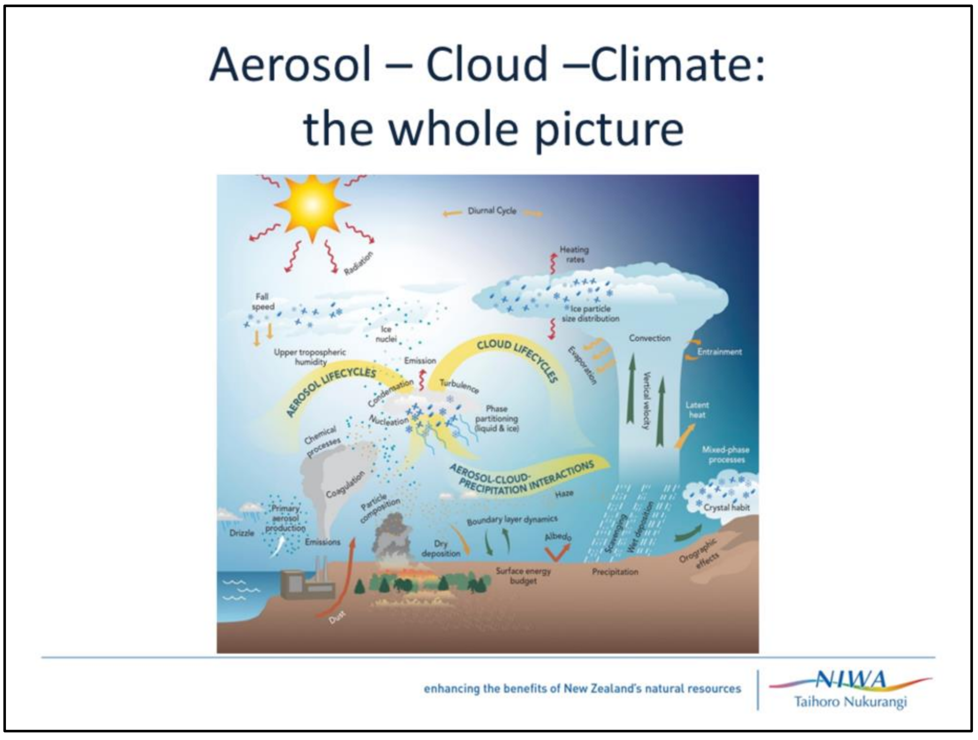

More aerosols result in a larger concentration of smaller droplets, leading to a brighter cloud. However there are many other possible aerosol–cloud–precipitation processes which may amplify or dampen this effect.

AR5, Twomey effect, Indirect aerosol effect,
a mechanism in the "CLAW" hypothesis



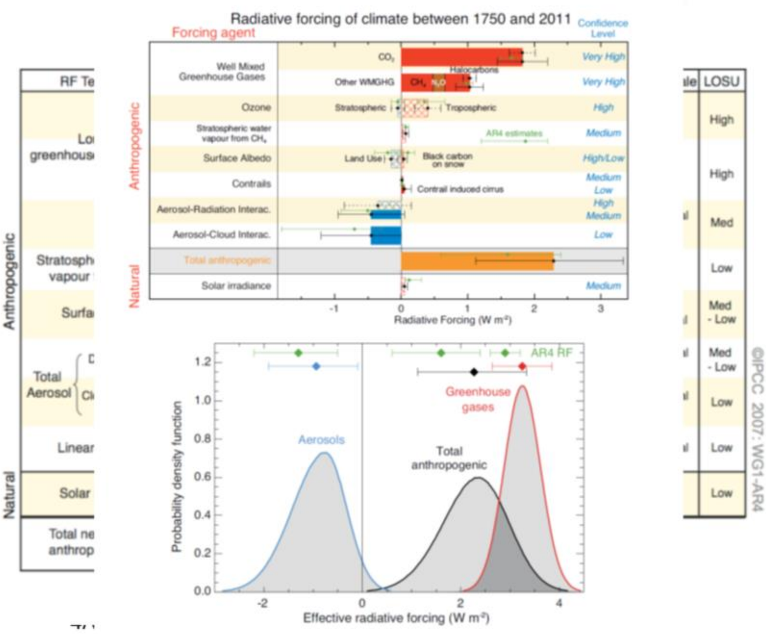
Shiptracks, seen from space, are an example of aerosol-cloud interaction. Ships sailing beneath these clouds have released particles which have seeded them with more CCN, creating lines of enhanced reflectivity.


Chemistry
Cloud condensation nuclei
CCN
Cloud formation
Albedo
Particle formation and interaction
Aerosol processes
Sulfuric acid
H2SO4
Oxidation
Sea-salt emission
1°Organic aerosol emission
Other Volatiles emission
Sulfur dioxide
SO2 emission
Oxidation
DMS emission
Combustion particles & sulfur dioxide




The complex suite of physical, chemical and dynamical processes governing aerosol-cloud-climate processes

# IPCC assessments




**5AR: Figure TS.6 |** Radiative forcing (RF) and Effective radiative forcing (ERF) of climate change during the Industrial Era. (Top) Forcing by concentration change between 1750 and 2011 with associated uncertainty range (solid bars are ERF, hatched bars are RF, green diamonds and associated uncertainties are for RF assessed in AR4). (Bottom) Probability density functions (PDFs) for the ERF, for the aerosol, greenhouse gas (GHG) and total. The green lines show the AR4 RF 90% confidence intervals and can be compared with the red, blue and black lines which show the AR5 ERF 90% confidence intervals (although RF and ERF differ, especially for aerosols). The ERF from surface albedo changes and combined contrails and contrail-induced cirrus is included in the total anthropogenic forcing, but not shown as a separate PDF. For some forcing mechanisms (ozone, land use, solar) the RF is assumed to be representative of the ERF but an additional uncertainty of 17% is added in quadrature to the RF uncertainty. {Figures 8.15, 8.16} The cooling estimates are slightly less than AR4 given improved model

# Geoengineering proposals


Halt runaway climate change


Buy time ahead of long-term solution

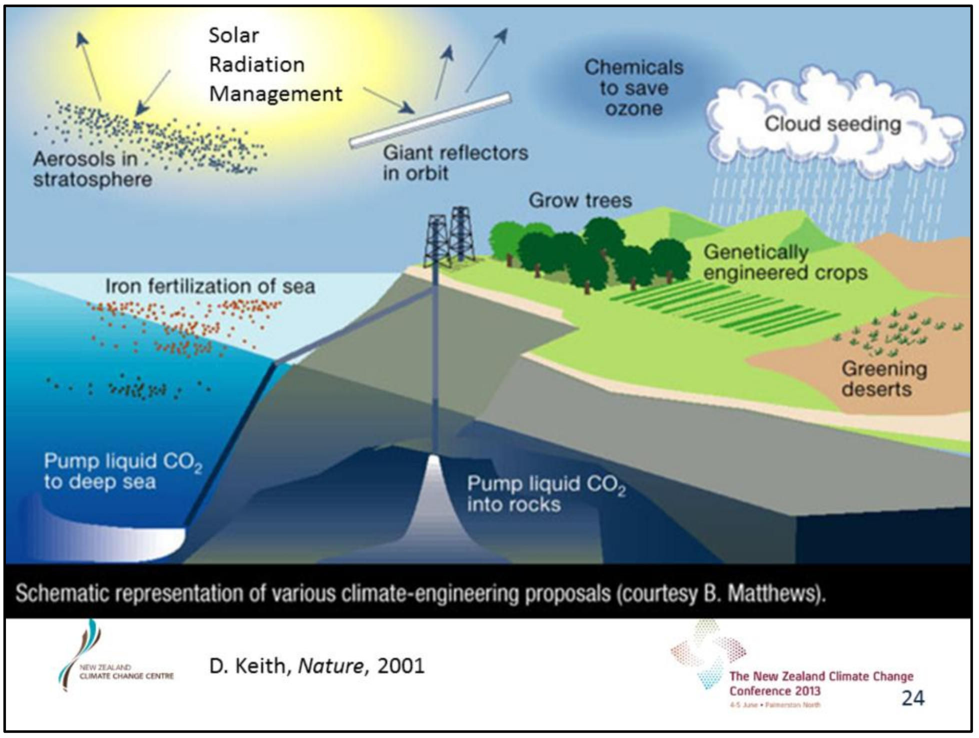
Approach this with extreme caution due to possibility of unintended consequences

# Climate – (geo) engineering

- "deliberate large-scale manipulation of the planetary environment to counteract anthropogenic climate change" *- Royal Society*

- "deliberately manipulating physical, chemical, or biological aspects of the Earth system… to counteract the consequences of increasing greenhouse gas emissions" *- American Meteorological Society*

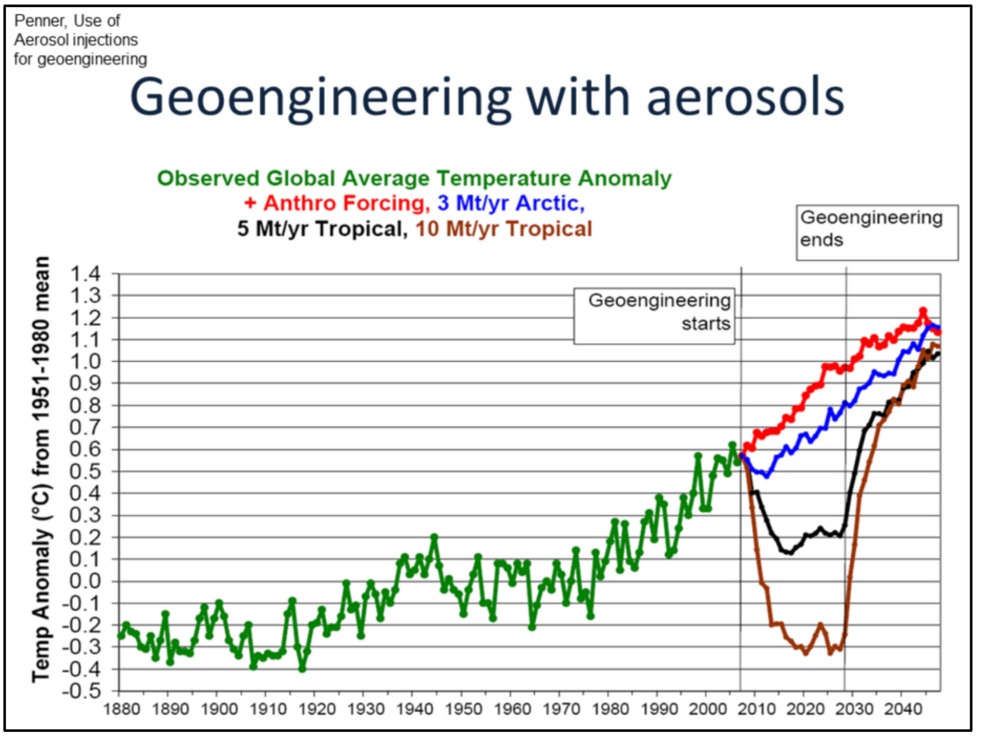
- "deliberate, large-scale manipulations of Earth's environment designed to offset some of the harmful consequences of GHG-induced climate change. Geoengineering encompasses two very different classes of approaches: CO2 removal and solar radiation management." *- America's Climate Choices/NAS*



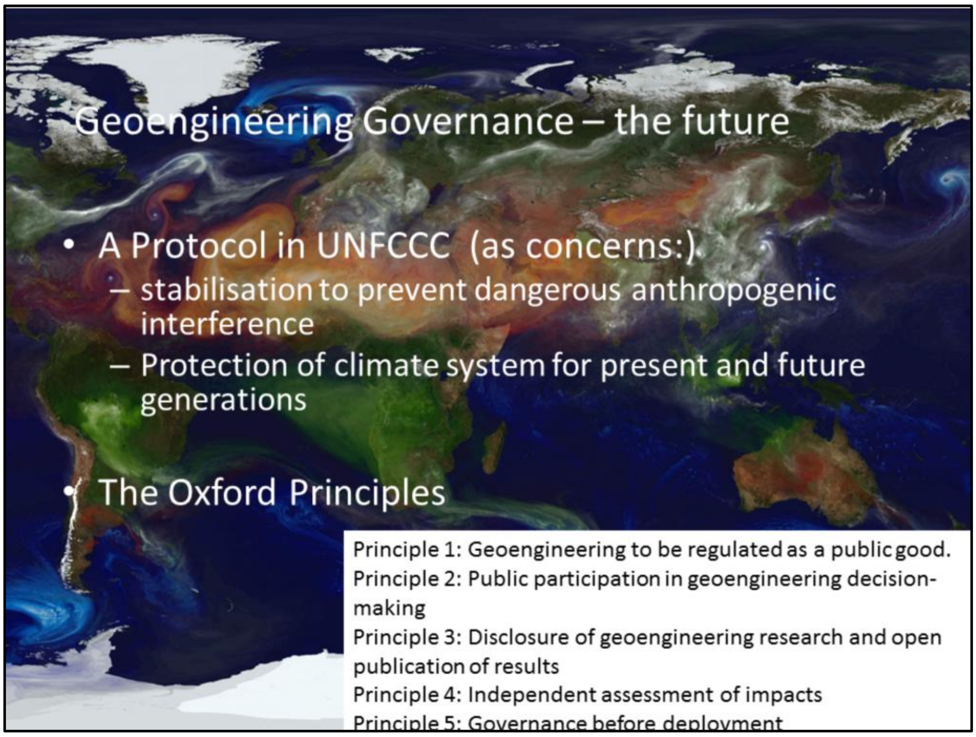

Atmospheric projects

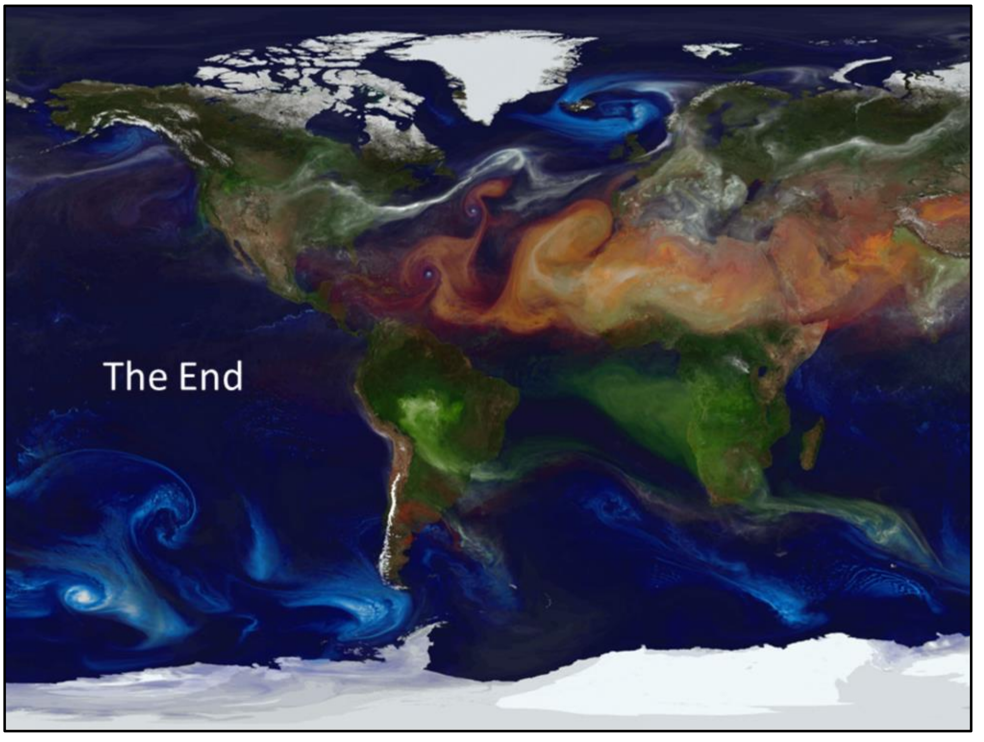
Stratospheric sulfur aerosols
Reflective aerosols or dust
Cloud whitening / marine cloud brightening / cloud reflectivity enhancement (CCN)
Cloud seeding
Ocean sulfur cycle enhancement (with OIF)
Reflective balloons
Low stratospheric soot

http://en.wikipedia.org/wiki/Solar_radiation_management




Penner et al, example of geoengineering model simulations


Geoengineering Governance – the future
• A Protocol in UNFCCC (as concerns:)
– stabilisation to prevent dangerous anthropogenic interference
– Protection of climate system for present and future generations
• The Oxford Principles
Principle 1: Geoengineering to be regulated as a public good.
Principle 2: Public participation in geoengineering decision-making
Principle 3: Disclosure of geoengineering research and open publication of results
Principle 4: Independent assessment of impacts
Principle 5: Governance before deployment


The End