

Possible feedbacks of aerosols on meteorology

Part 1: Physical processes

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MUSCATEN summer school

"Integrated Modelling of Meteorological and Chemical Transport Processes / Impact of Chemical Weather on Numerical Weather Prediction and Climate Modelling"

Odessa, Ukraine, 3-9 July 2011



Lectures Objective and Goal:

- Description of the main feedback mechanisms of the chemical weather (atmospheric green-house gases and aerosols) impact on NWP and climate processes, in order to understand how important it is to include feedbacks from gases, aerosols, clouds, etc. in NWP, CWF and climate models.
- The goal is to give an orientation/understanding of which feedback processes are the most important: impact of feedbacks from gases, aerosols (direct, semi-direct, indirect effects), clouds, etc. on short and long time-range meteorological models.
- This subject is the main focus of the school. First part (1st lecture) focuses on the physical processes behind these feedbacks. Second part (2nd lecture) focuses on model examples.



(after Jacobson, 2002)

2007 IPCC Estimate of Gas and Aerosol Radiative Effects

Radiative Forcing Components



Comparison of anthropogenic aerosol forcing with greenhouse forcing Ramanathan et al, Science, 2001





Atmosphere Interactions:

Gases, Aerosols, Chemistry, Transport, Radiation, Climate



After Y. Zhang, DMI, Copenhagen, 2007

Examples of Important Feedbacks

• Effects of Meteorology and Climate on Gases and Aerosols

- Meteorology is responsible for atmospheric transport and diffusion of pollutants
- Changes in temperature, humidity, and precipitation directly affect species conc.
- The cooling of the stratosphere due to the accumulation of GHGs affects lifetimes
- Changes in tropospheric vertical temperature structure affect transport of species
- Changes in vegetation alter dry deposition and emission rates of biogenic species
- Climate changes alter biological sources and sinks of radiatively active species
- Effects of Gases and Aerosols on Meteorology and Climate
 - Decrease net downward solar/thermal-IR radiation and photolysis (direct effect)
 - Affect PBL meteorology (decrease near-surface air temperature, wind speed, and cloud cover and increase RH and atmospheric stability) (semi-indirect effect)
 - Aerosols serve as CCN, reduce drop size and increase drop number, reflectivity, and optical depth of low level clouds (LLC) (the Twomey or first indirect effect)
 - Aerosols increase liquid water content, fractional cloudiness, and lifetime of LLC but suppress precipitation (the second indirect effect)



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Do Cities Affect the Diurnal Cycle of Rainfall?



Average **annual** diurnal rainfall distributions at gage 4311 (UA) for the urban (1984-1999) and pre-urban (1940-1958) time periods



Average warm season diurnal rainfall distribution at gage 4311 for the urban (1984-1999) and preurban (1940-1958) time periods

(NASA, Shepherd, 2004)

The peak fraction of daily rainfall is more pronounced for the 12-16 and 16-20 4hr time increments for the urban time period compared to the pre-urban time period; <u>The warm season experiences a greater diurnal modification</u>

Main feedback mechanisms of aerosol forcing

• Direct effect via radiation:

(i) warm the air by absorbing solar and thermal-IR radiation,(ii) cool the air by backscattering incident short wave radiation to space

- Semi-direct effect: via PBL meteorology, photochemistry, photolysis and aerosol emission/blowing changes
- **First indirect effect**: via reflectivity, optical depth, cloud albedo and other radiation characteristics due to growing CCN/IN
- Second indirect effect: via microphysics of clouds, interacting with aerosols, CCN/IN growing, washout and rainout => precipitation

They have to be prioritised and considered in on-line coupled modelling systems.

Sensitivity studies are needed to understand the relative importance of different feedback effects.

Feedbacks classification is not complete

- Aerosols affect the climate system by changing cloud characteristics in many ways (and different directions).
- They act as cloud condensation and ice nuclei, they may inhibit freezing and they could have an influence on the hydrological cycle.
- While the cloud albedo enhancement (Twomey effect) of warm clouds received most attention so far and traditionally is the only indirect aerosol forcing considered in transient climate simulations, the multitude of effects should be considered.

Effects of aerosol particles on climate: Jacobson (2002) classification and Some examples

- Self-Feedback Effect
- Photochemistry Effect
- Smudge-Pot Effect
- Daytime Stability Effect
- Particle Effect Through Surface Albedo
- Particle Effect Through Large-Scale Meteorology
- Indirect Effect
- Semidirect Effect
- BC-Low-Cloud-Positive Feedback Loop



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Different aerosol effects on water clouds

Cloud albedo effect (pure forcing)

– for a constant cloud water content, more aerosols lead to more and smaller cloud droplets \Rightarrow larger cross sectional area \Rightarrow more reflection of solar radiation

Cloud lifetime effect (involves feedbacks)

– the more and smaller cloud droplets will not collide as efficiently \Rightarrow decrease drizzle formation \Rightarrow increase cloud lifetime \Rightarrow more reflection of solar radiation

Semi-direct effect (involves feedbacks)

- absorption of solar radiation by black carbon within a cloud increases the temperature \Rightarrow decreases relative humidity \Rightarrow evaporation of cloud droplets \Rightarrow more absorption of solar radiation (opposite sign)

=> Online integrated models are necessary to simulate correctly these effects involved 2nd feedbacks

Overview of the different aerosol indirect effects (acc. to Lohmann and Feichter, 2005)

ا الا الجي الم Table 1. Overview of the different aerosol indirect effects and range of the radiative budget perturbation at the top-of-the atmosphere (FTOA) $[Wm^{-2}]$, at the surface (F_{SFC}) and the likely sign of the change in global mean surface precipitation (P) as estimated from Fig. 2 and from the literature cited in the text.

Effect	Cloud type	Description	F_{TOA}	F_{SFC}	Р
Indirect aerosol effect for clouds with fixed water amounts (cloud albedo or Twomey effect)	All clouds	The more numerous smaller cloud particles reflect more solar radiation	-0.5 to -1.9	similar to F _{T O A}	n/a
Indirect aerosol effect with varying water amounts (cloud lifetime effect)	All clouds	Smaller cloud particles decrease the precipitation efficiency thereby prolonging cloud lifetime	-0.3 to -1.4	similar to F _{T O A}	decrease
Semi-direct effect	All clouds	Absorption of solar radiation by soot may cause evaporation of cloud particles	+0.1 to -0.5	larger than F _{T O A}	decrease
Thermodynamic effect	Mixed-phase clouds	Smaller cloud droplets delay the onset of freezing	?	?	increase or decrease
Glaciation indirect effect	Mixed-phase clouds	More ice nuclei increase the precipitation efficiency	?	?	increase
Riming indirect effect	Mixed-phase clouds	Smaller cloud droplets decrease the riming efficiency	?	?	decrease
Surface energy budget effect	All clouds	Increased aerosol and cloud optical thickness decrease the net surface solar radiation	n/a	-1.8 to -4	decrease



Main sources and components of nucleation, accumulation and coarse mode particles

Nucleation Mode	Accumulation Mode	Coarse Mode
Nucleation	Fossil-fuel emissions	Sea-spray emissions
H ₂ O(aq), SO ₄ ²⁻ , NH ₄ ⁺	BC, OM, SO ₄ ²⁻ , Fe, Zn	H ₂ O, Na ⁺ , Ca ²⁺ , Mg ²⁺ , K ⁺ , Cl ⁻ , SO ₄ ²⁻ , Br ⁻ , OM
Fossil-fuel emissions	Biomass-burning emissions	Soil-dust emissions
BC, OM, SO ₄ ²⁻ , Fe, Zn	BC, OM, K ⁺ , Na ⁺ , Ca ²⁺ , Mg ²⁺ ,	Si, Al, Fe, Ti, P, Mn, Co, Ni, Cr,
	SO ₄ ²⁻ , NO ₃ ⁻ , CI ⁻ , Fe, Mn, Zn,	Na ⁺ , Ca ²⁺ , Mg ²⁺ , K ⁺ , SO ₄ ²⁻ ,
	Pb, V, Cd, Cu, Co, Sb, As, Ni,	CI ⁻ , CO ₃ ²⁻ , OM
	Cr	
Biomass-burning emissions	Industrial emission	Biomass burning ash, industrial
BC, OM, K ⁺ , Na ⁺ , Ca ²⁺ , Mg ²⁺ ,	BC, OM, Fe, Al, S, P, Mn, Zn, Pb,	fly-ash, tire-particle emissions
SO ₄ ²⁻ , NO ₃ ⁻ , CI ⁻ , Fe, Mn,	Ba, Sr, V, Cd, Cu, Co, Hg, Sb,	
Zn, Pb, V, Cd, Cu, Co, Sb,	As, Sn, Ni, Cr, H ₂ O, NH ₄ ⁺ , Na ⁺ ,	
As, Ni, Cr	Ca ²⁺ , K ⁺ , SO ₄ ²⁻ , NO ₃ , Cl , CO ₂ ²⁻	
Condensation/dissolution	Condensation/dissolution	Condensation/dissolution
H ₂ O(aq), SO ₄ ²⁻ , NH ₄ ⁺ , OM	H ₂ O(ag), SO ₄ ²⁻ , NH ₄ ⁺ , OM	H ₂ O(ag), NO ₂ ⁻
	Coagulation of all components from	Coagulation of all components
	nucleation mode	from smaller modes

Main emission components:

DMS:	(Bates et al., 1987)
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- SO2: (Spiro et al., 1992) (Hao et al., 1991)
- BC: (Cooke and Wilson, 1996)

 OC: biomass burning: proportional to BC 7:1 fossil fuel burning: proportional to SO4 1:1
 Sea Salt: burden proportional to wind speed (Blanchard & Woodcock, 1980)

Carbonaceous Aerosols

•Carbonaceous aerosols are divided into two categories: black carbon (BC) and organic carbon (OC). BC is a strong absorber of visible and near-IR light; OC mostly scatters radiation.

•OC is further divided into primary organic aerosol (POA) and secondary organic aerosol (SOA).

•The dominant emissions of BC and POA are fossil fuel and biomass burning.

•SOAs are formed when volatile organic compounds (VOCs) are oxidized to form semi-volatile products.

•Biogenic VOCs, especially monoterpenes ($C_{10}H_{16}$), are the most important VOCs for SOA formation.



Sulphate aerosol radiative forcing

CDNC is the cloud droplet number concentration and

CCN is the cloud condensation nuclei.





Direct Aerosol Forcing

(i) <u>warm the air by</u> <u>absorbing solar and</u> <u>thermal-IR radiation</u> (black carbon, iron, aluminium, polycyclic and nitrated aromatic compounds),
(ii) <u>cool the air by</u> <u>backscattering</u> incident short wave

<u>radiation to space</u> (water, sulphate, nitrate, most of organic compounds)





Single-particle absorption (Q_a), total scattering (Q_s), and forward scattering (Q_f) efficiencies



 \uparrow (a) BC particles

(b) liquid water drops \Rightarrow

different sizes at λ =0.5 µm

(after Jacobson, 2002)



Particle Scattering and Absorption Extinction Coefficients

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Despite big similarities with gases (*will be considered in the next lecture*) the particle scattering absorption is more complex due to variety of size and composition aerosols.

Aerosol particle absorption and scattering extinction coefficients (in cm⁻¹) at a given wavelength can be estimated as:

$$\sigma_{a,p} = \sum_{i=1}^{N_b} n_i \pi r_i^2 Q_{a,i} \qquad \sigma_{s,p} = \sum_{i=1}^{N_b} n_i \pi r_i^2 Q_{s,i}$$

Where the summations are over N_b particle sizes, ni is the number concentration (part. per cm3 of air) of particles of raius ri (cm), πr_i^2 is the actual cross-section of a particle (cm² per part.), Q_{ai} and Q_{si} are single-particle absorption and scattering efficiencies (dimensionless).

Direct aerosol effect in models

- Realisation depends on the radiation scheme used in the model.
- The first simple version of implementation into the Enviro-HIRLAM model with the radiation scheme of Savijärvi (1990) is realised based on Li et al (2001) parameterisation.
- Following (Seinfeld and Pandis, 1998) it is possible to estimate the effect of a layer of scattering aerosol accounting for surface reflections, by modifying the surface albedo accordingly.
- Another approach would be to use look-up tables for the complex index of refraction for various aerosol compositions, assuming that the aerosol is in the Rayleigh scattering regime.



UNCERTAINTIES IN RADIATIVE FORCING OF BLACK CARBON



(Seinfeld, 2003)

Estimations of the aerosol direct forcing (after Seinfeld, 2003)

•Global BC and OC burdens are estimated to have increased by an order of magnitude since the preindustrial period.

•SOA contribution to the total OC is predicted to be the greatest in the upper troposphere where lower temperatures allow more semi-volatile products to condense to the aerosol phase.

 Predicted regional BC and OC concentrations are consistently low, suggesting that emissions need to be revised.

•Globally averaged, anthropogenic BC, OC, and sulfate are predicted to exert a radiative forcing of -0.39 to -0.78 W m⁻², depending on the assumptions of aerosol mixing and water uptake by OC.

Direct effect in Enviro-HIRLAM

- Improved direct aerosol effects in the Savijarvi (1990) radiation scheme.
- 1 shortwave spectral band (and 1 LW band);
- Heating rates at model levels;
- Transmittances as horizontal elds.
- Assumptions:
- Spectrally weighted optical properties used;
- 2-stream raditiative transfer theory used.

Slide from Kristian P. Nielsen

Direct effect modules in Enviro-HIRLAM

- 1. Calculation of spectrally weighted aerosol inherent optical properties => Look-up table;
- 2. Subroutine for calculating transmittances and absorption optical depths;
- 3. Implementation of these in the Savijarvi scheme:
 (a) Aerosol SW transmittances for cloud free, in clouds, above clouds and below clouds included.
 (b) Aerosol SW absorptance the semi direct effect included.
- (c) Aerosol LW effects included as the SW effects.

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Module test and validation

- Test of subroutine against highly accurate DISORT calculations
- { Good results, for mineral aerosols, sea salt and sulphate aerosols;
- { For soot aerosols, results are not valid for very high concentrations.
- Test of the subroutine itself in 0-D by Katya Khoreva (see her poster).



Direct aerosol effects on SW radiation was run in detail for the aerosol species in GADS (1997) by the DISORT model, considering the full spectral radiance field. The species include BC (soot), Minerals (nucleus mode, acc. mode, coarse mode and transported mode), Sulphuric acid, Sea salt (acc. mode and coarse mode), "water soluble", and "water insoluble"

Slide from Kristian P. Nielsen

First Indirect Aerosol Effect

Polluted airmass has more aerosols => hence more cloud droplets

Unpolluted	Polluted

• Cloud albedo depends on droplet surface area, so second cloud is brighter (so-called 'Twomey effect' or 'first indirect effect')

- For a constant cloud water content, more aerosols lead to more and smaller cloud droplets \Rightarrow larger cross sectional area \Rightarrow more reflection of solar radiation
- Lots of evidence to support this effect.
- Quantified by effective radius Reff.

Observational evidence: Ship tracks

- Classical example of indirect aerosol effect
- Ship tracks off the coast of Washington

Durkee et al., 2000





First indirect aerosol effect in Enviro-HIRLAM (Korsholm et al., 2008)

As anthropogenic aerosols enter cloud environments the number concentration of cloud condensation nuclei (CCN) is modified, generally, resulting in more numerous and smaller CCN (decreased mean diameter). The cloud radiation characteristics depend on bulk cloud properties and a decrease in droplet mean diameter results in a modification (whitening) of the cloud albedo. The HIRLAM radiation scheme is based on (Savijärvi, 1990) and all water cloud radiation is parameterized via the cloud droplet effective radius, r_e , which may be written as:

$$r_e = \left(\frac{3L_c}{4\pi\rho_w kN}\right)^{1/2}$$

where L_c is the cloud condensate content, ρ_w is the density of water, k is a fitting parameter which distinguishes between land and water surfaces and N is the cloud droplet number concentration (Wyser, et al., 1999). N may be decomposed into a natural background and an anthropogenic contribution: $N = N_{back} + N_{anthr}$, where N_{back} is a constant for clean air supplied in HIRLAM depending only on the surface type (land or water), while N_{anthr} is calculated in the aerosol module assuming that all accumulation mode aerosols may act as CCN.

The new ALADIN/HARMONIE cloud scheme (*Pinty and Jabouille, 1998; Caniaux et al., 1994*) is more suitable for implementation of more comprehensive aerosol dynamics and indirect effects of aerosols (CCN/IN) models, but will be more expensive computationally.

Second Indirect Aerosol Effect

Suppression of precipitation in polluted areas

- 'Cloud lifetime' or 'second indirect effect'
- Much less evidence to support this effect.

Stevens & Feingold, 2009

The local inhibition of precipitation helps precondition the environment for deeper convection, which then rains more (Stevens & Feingold, 2009)

Scheme of Aerosol-CCN/IN dynamics modelling

Raes et al., AE, 2000

Mechanisms of the indirect aerosol effects

CDNC denotes the cloud droplet number concentration and

IP the number concentration of ice particles.

Penner et al., IPCC, 2001

Second indirect aerosol effect in Enviro-HIRLAM *(Korsholm et al., 2008)*

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The description of cloud microphysics in the STRACO scheme is based on the Sundqvist parameterization (Sundqvist, 1988, Sundqvist, et al., 1989, Sundqvist, 1993). STRACO has been extended to include the effects of cloud drop number concentration and characteristic droplet radius r, by combining the autoconversion term for cloud water from (Rasch and Kristjansson, 1998) with the existing formulation in the STRACO scheme (Sass, 2002). In STRACO precipitation release is written $G_p = \Phi q_c (1 - \exp(-X^2))$ where q_c is the cloud condensate, $X = \hat{q}_c/\mu$ where $\hat{q}_c = q_c/f$ is the in-cloud specific cloud condensate and f is the grid box fractional cloud cover. The Φ term is defined as: $\Phi = \Phi_1 \Phi_2 \Phi_3 \Phi_4$ where Φ_2 describes the effect of collision/coalescense and the Bergeron-Findeisen mechanism, Φ_3 expresses a temperature dependency at cold temperatures, Φ_4 is height dependent and describes an enhanced sedimentation of cloud droplets from fog (clouds at very low levels) and Φ_1 is the autoconversion term which is now defined as:

$$\Phi_1 = C_{l,out} \widehat{q}_c \frac{\rho_a}{\rho_w} \left(\widehat{q}_c \frac{\rho_a}{\rho_w} N \right)^{\frac{1}{3}} H(r - r_0)$$

Here ρ_a represents air density, H is the Heavy-side step function, $C_{l,aut}$ is a constant (Rasch and Kristjansson, 1998), $r = [(3\rho_a q_c)/(4\pi N \rho_w)]^{1/3}$ is the mean cloud droplet radius and r_0 is a constant threshold drop radius (5µm).

As before $N = N_{back} + N_{anthr}$ where N_{back} depends only on surface type and N_{anthr} is calculated in the aerosol module. The modifications made to the STRACO scheme is currently being tested, but preliminary runs show that it gives results similar to the latest STRACO version.

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Surprising conclusion one of the authors ...

Emissions of sulfur dioxide have decreased considerably in North America and Europe after a peak in the late 1970s and early 1980s. This results from an interplay of political decisions to cut emissions, the replacement of 'dirty' fuels, and new technologies for removing sulfur from fossil fuel and for cleaning flue gases in power plants. Nonetheless, power generation and smelting remain major sources.

Less sulfates more warming?

Andreae et al., Nature 2005

Schematic diagram of the warm indirect aerosol effect (solid arrows) and glaciation indirect aerosol effect (dotted arrows)

(adapted from Lohmann, 2002)

How are aerosol effects on clouds simulated in meteorology/climate models?

- Predict aerosol mass concentrations:
- *sources* (aerosol emissions of the major aerosol species: sulfate, black carbon, organic carbon, sea salt, dust)
- *transformation* (aerosol formation and dynamics, dry and wet deposition, chemical transformation and transport)
- Need a good description of cloud properties:
- *precipitation formation* (collision/coalescence of cloud droplets and ice crystals, riming of snow flakes)
- Need to parameterize aerosol-cloud interactions:
- *cloud droplet nucleation* (activation of hygroscopic aerosol particles)
- *ice crystal formation* (contact and immersion freezing, homogeneous freezing in cirrus clouds)

Schematic aerosol-cloud interaction for marine air

(after Hobbs, 1993; see also Gross and Baklanov, 2004)

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Schematic aerosol-cloud interaction for continental air Sedimentation Detrain to from free troposphere free troposphere SO₄, NO₃ Production Cu Large Activation clouds CCN St - Sc Activation clouds

(after Hobbs, 1993; see also Enviro-HIRLAM realisation in Korsholm et al., 2008)

Implementation of the feedback mechanisms into integrated models:

One-way integration (off-line):

- 1. Simplest way (no aerosol forcing): NWP meteofields as a driver for ACTM (this classical way is used already by most of air pollution modellers);
- 2. ACTM chemical composition fields as a driver for Regional/Global Climate Models (including the aerosol forcing on meteo-processes, it could also be realized for NWP or MetMs).

Two-way integration:

- 1. Driver + partly aerosol feedbacks, for ACTP or for NWP (data exchange with a limited time period coupling: off-line or on-line access coupling, with or without second iteration with corrected fields);
- 2. Full feedbacks included on each time step (online coupling/integration).

Scientific hypotheses/questions to be tested/addressed

• Hypothesis

• Feedback mechanisms are important in accurate modeling of NWP/MM-ACT and quantifying direct and indirect effects of aerosols.

• Key questions

- What are the effects of climate/meteorology on the abundance and properties (chemical, microphysical, and radiative) of aerosols on urban/regional scales?
- What are the effects of aerosols on urban/regional climate/meteorology and their relative importance (e.g., anthropogenic vs. natural)?
- How important the two-way/chain feedbacks among meteorology, climate, and air quality are in the estimated effects?
- What is the relative importance of aerosol direct and indirect effects in the estimates?
- What are the key uncertainties associated with model predictions of those effects?
- How can simulated feedbacks be verified with available datasets?

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