Relative humidity RH in aerosol modeling and the associated uncertainties

Eric DeMeij, JRC

Why important in modelling?

- Plays a key role in aerosol calculation
- Water uptake changes the size of the aerosol (sea salt, black carbon, sulfate)
- Changes optical and chemical properties

Some points where RH plays a key role in aerosol water calculation (1)

- The activities of aerosols in equilibrium with the environment are governed by the relative humidity (RH).

  This means that the water activity (aw) is fixed to the RH:

  $$aw = f(RH)$$

The activity coefficients are needed to calculate the equilibrium concentrations (aqueous phase).

$\Rightarrow$ RH ↑ ⇨ aerosol water mass ↑ RH ↓ ⇨ aerosol water mass ↓

This can change the aerosol composition due to condensation (evaporation / crystallization), because the vapor pressure above the aerosol reduces (increases)

Some points where RH plays a key role in aerosol water calculation (2)

Gas – aerosol equilibria are calculated between:

1. Gas phase and a aerosol pure solid.
2. Gas phase and ions in an aqueous solution.
3. Gas phase and aerosol with both aqueous and solid phases.

The behavior of the aerosol on the deliquescence relative humidity needs to be considered.

e.g.

- $(NH4)_2SO4$ and NH4NO3 deliquesce at a certain RH, below that RH, it may be crystalline.
- solutions containing sulfuric acid do not deliquesce.

The liquid / solid phase is determined according to the aerosol composition and the corresponding deliquescence RH (DRH) values. The salts are treated as solid or as liquid, depending on the RH. The DRH is a function of temperature.

BOXMODEL comparisons

Changing the RH, what is the impact of these changes on the aerosol water calculation?

Boxmodel of Pandis[1*] is used, where ISORROPIA is replaced with EQSAM[2*].

At different time steps:
- temperature and RH is changed,
- HNO3, NH3, H2SO4 is added,
- pressure is constant.

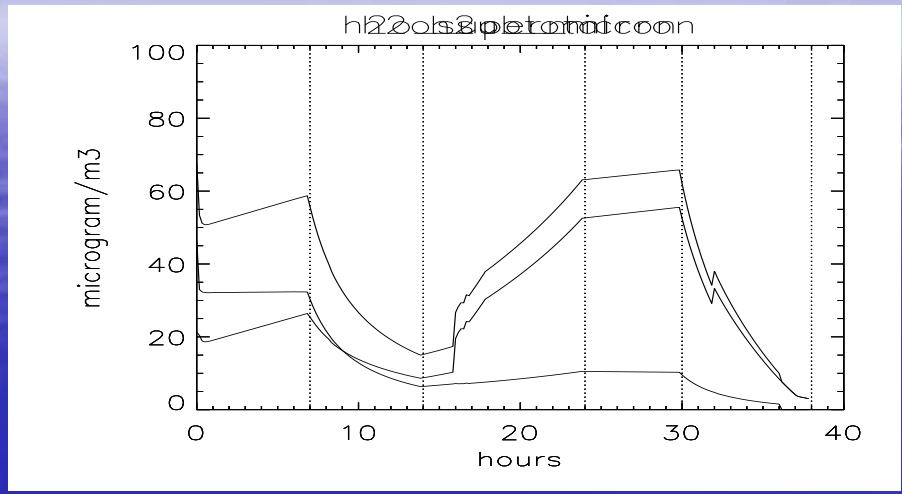
Initial conditions of aerosol species, temp, RH, pressure are set.
Boxmodel simulates 38hrs.

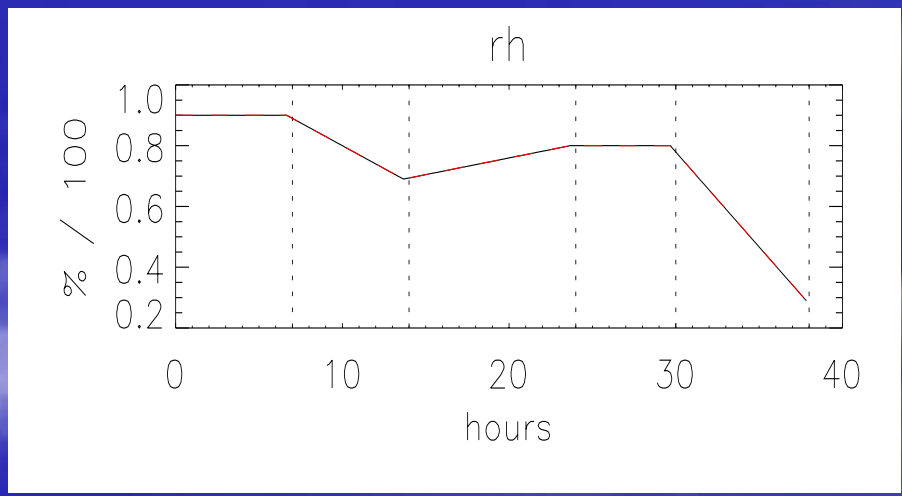
1*J. Geophysical Research, 107, 10.1029/2001JD001102, 2002.
2* Swen Metzger, et al. , 2000

Initialization RH = 90%, T = 296K, pressure is constant.

At 7h, 14h, 24h, 30h and RH changes

NH3, HNO3, H2SO4 are added at several time steps





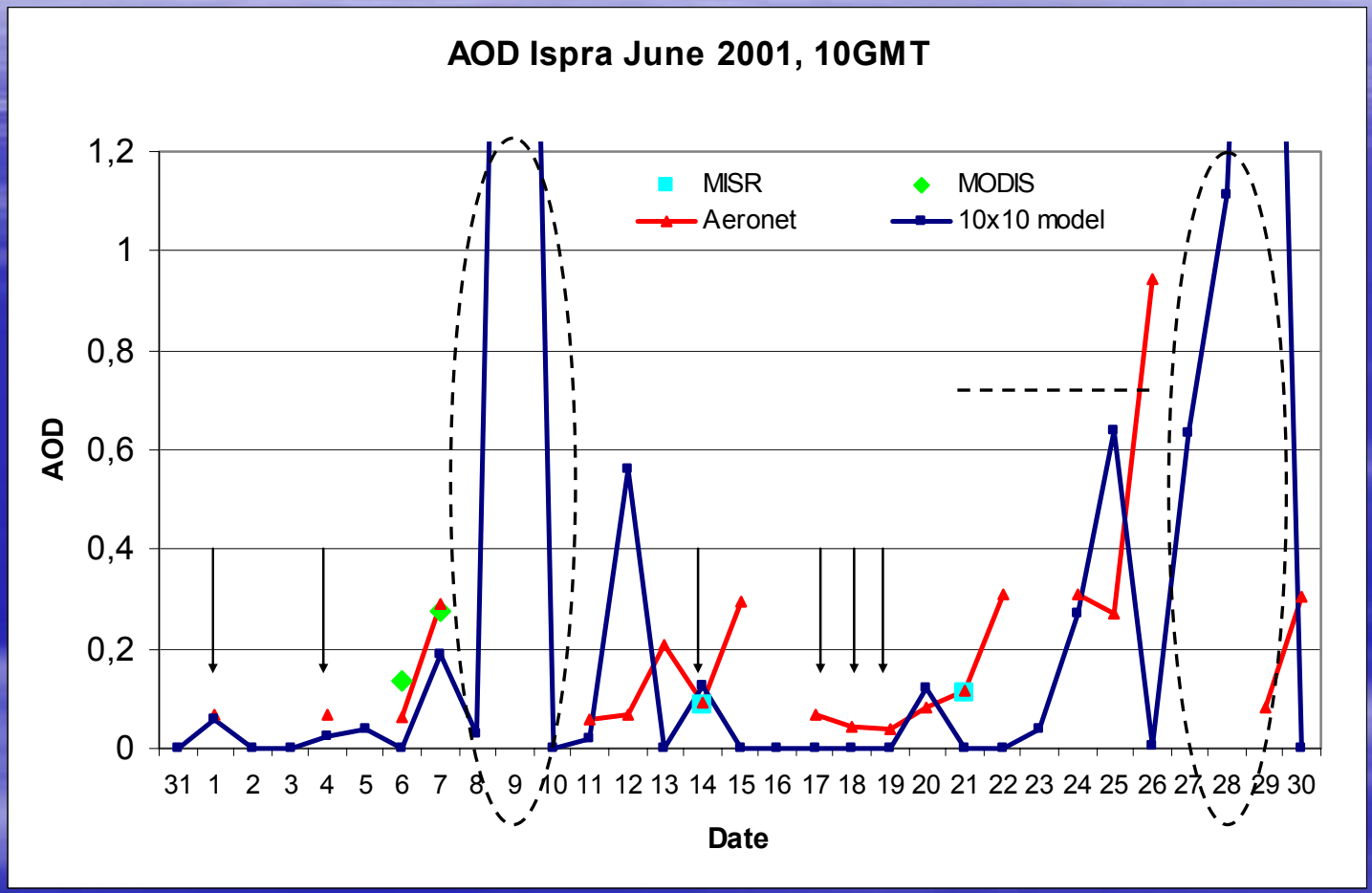
Tapom*

Transport Atmospheric Air Pollutant Model

- Off-line Mesoscale model
- Gasphase chemistry: RACM (Regional Atmospheric Chemistry Modelling)
- Aerosols: EQSAM (Equilibrium Simplified Aerosol Model)
- 300 x 300 km, Milan a centre domain, different resolutions
- 21 levels up to 6000m
- June 2001

* EPFL Lausanne
JRC IES, Ispra

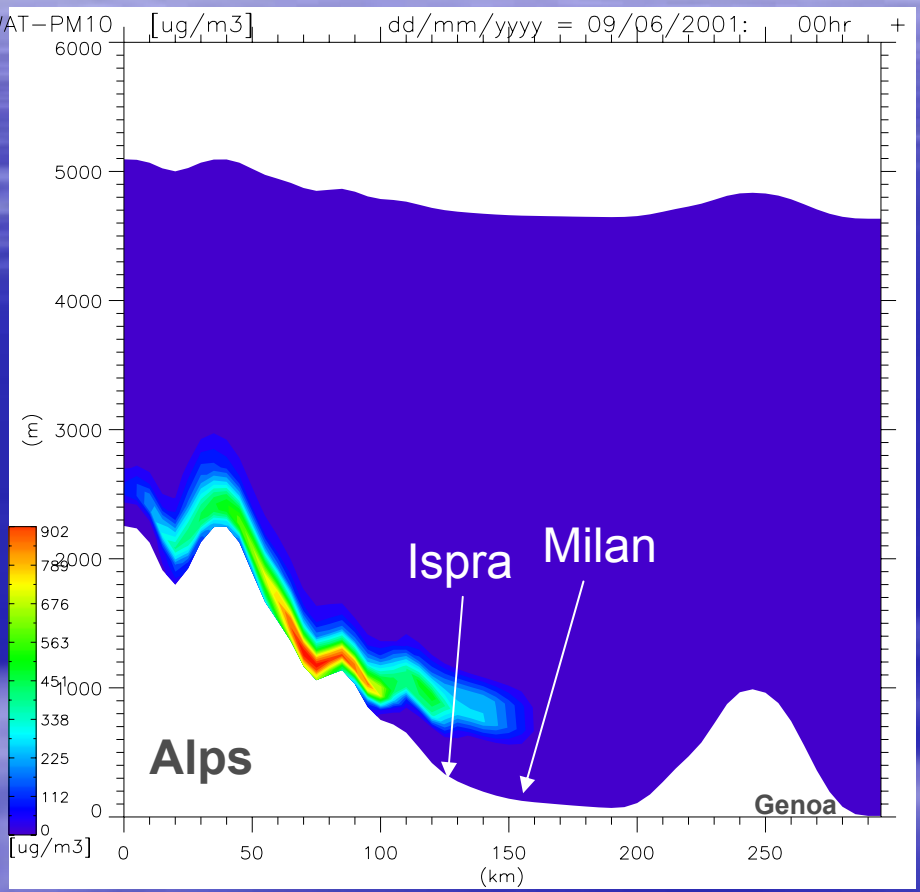
AOD (550nm) Ispra June 2001

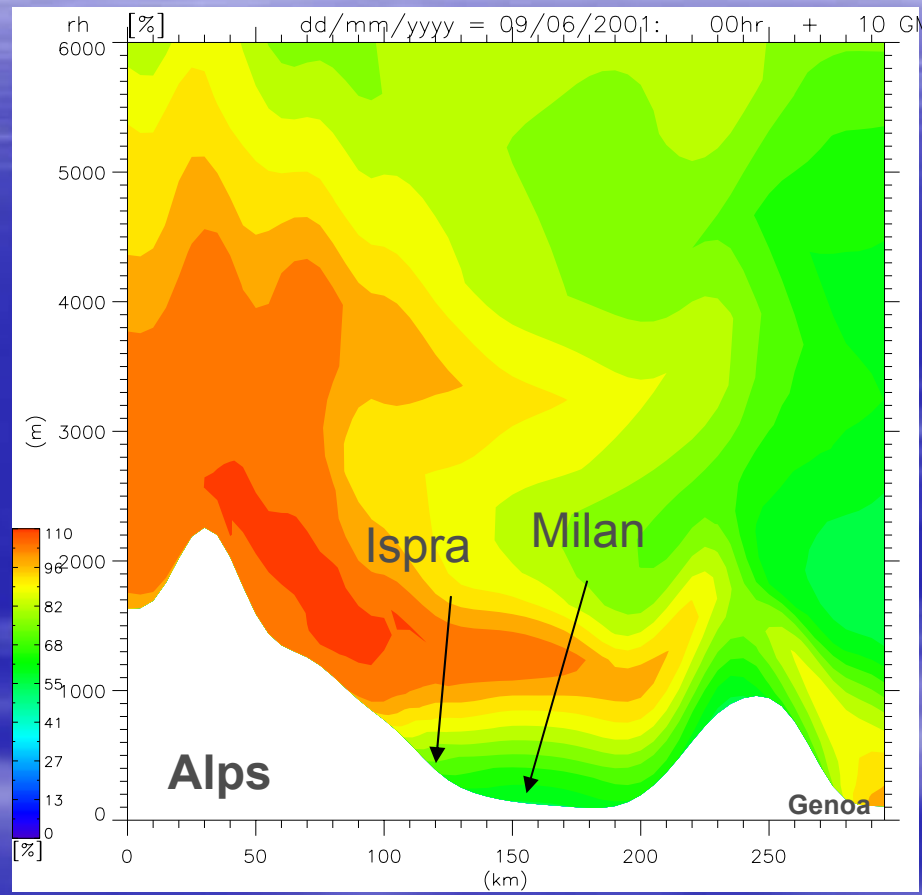


- Dry, clean days good agreement ↓
- Days high RH up to 90%, more discrepancy
- Cirrus clouds --

Aerosol water and RH profile, 9th June 2001

high RH measured at EMEP station Ispra



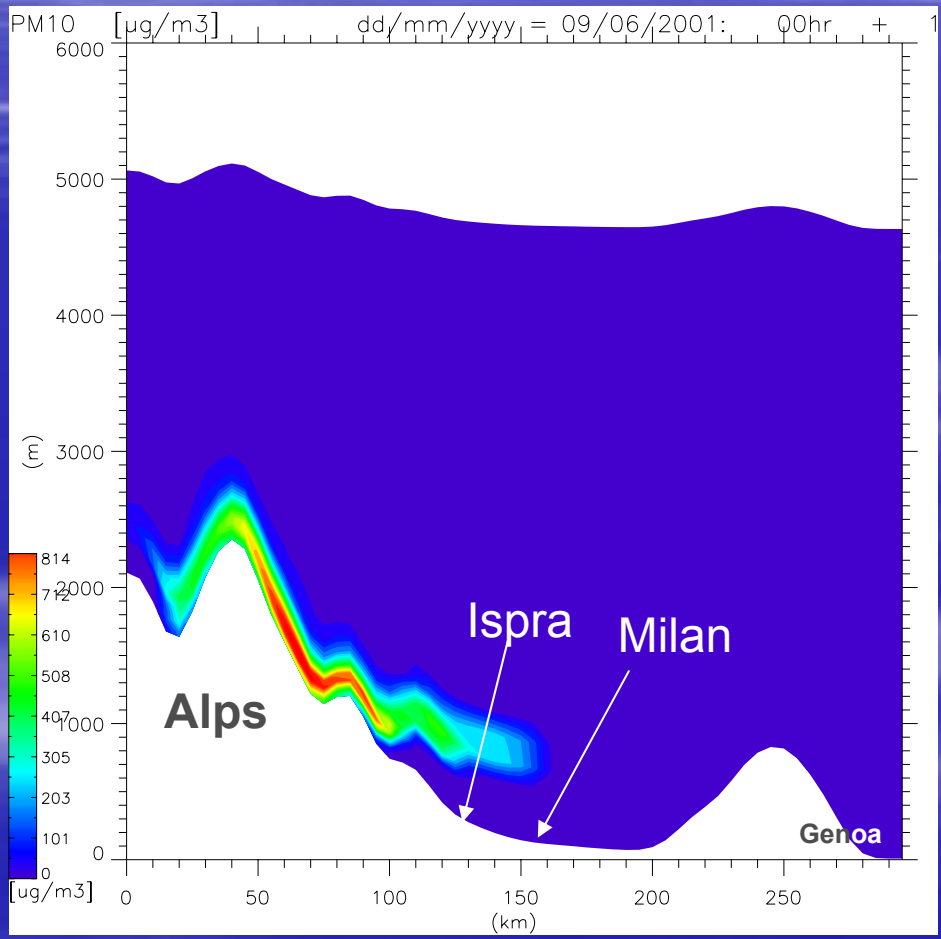


High aerosol water values
up to 9000 ug/m3

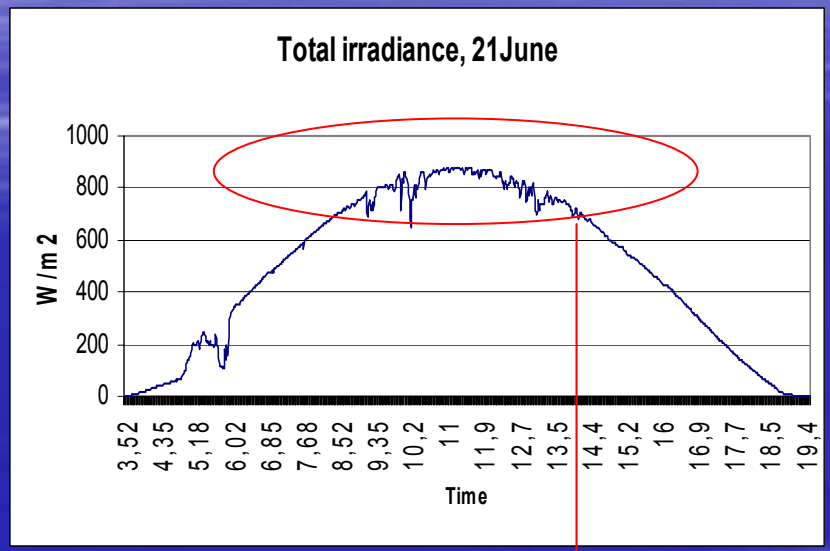
(Cloud formation?)

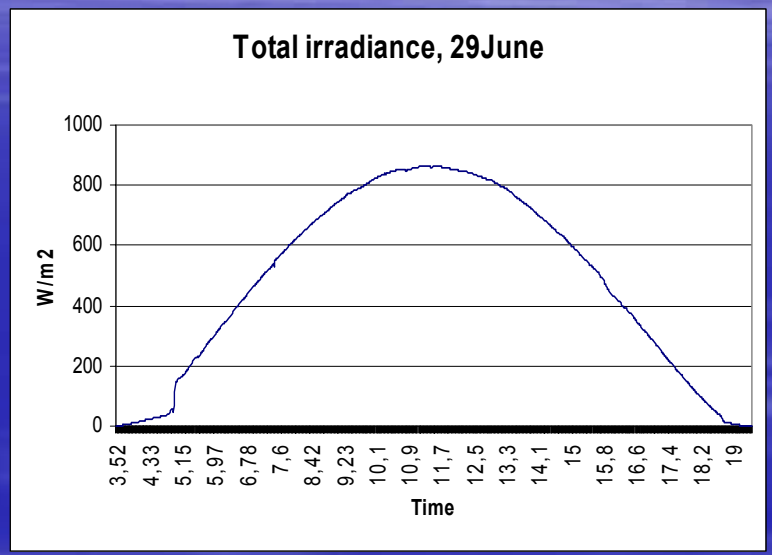
PM10, 9th June 2001

high RH measured at EMEP station Ispra



Haze / cirrus clouds





Indication of cirrus clouds

Dry clean day

J. Groebner, JRC,Ispra

Summary

- RH plays important role in aerosol modelling.
- Synoptic situation leads to high RH in the alpine foothills and cloud formation in the mountains.
- High RH in combination with high aerosol concentrations lead to very high amounts of aerosol water.
- It is difficult to distinguish this aerosol water from clouds.
- In addition measurements maybe perturbed by the presence of high cirrus clouds and / or haze.
- Does this happen also at other locations (or is it specific to Po Valley and Alps?). How do measurements take these high RH cases into account?