

# Atmospheric particulate matter and its impacts on climate, public health and ecosystems

*Athanasis Nenes*

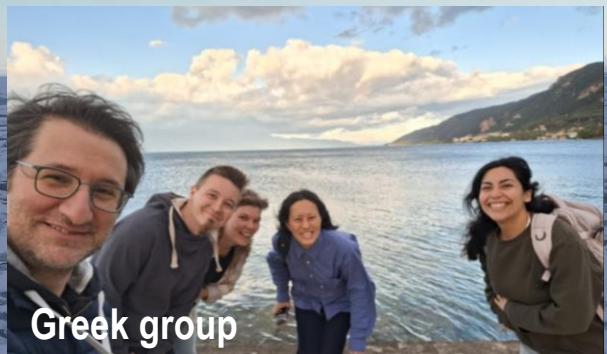
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Federale de Lausanne, Switzerland

Center for the Study of Air Quality and Climate Change  
Institute of Chemical Engineering & High Temperature Chemical  
Processes, Foundation for Research and Technology Hellas,  
Patras, Greece

*ENV 167 Presentation, December 2, 2024*

## LAPI – Athanasios (Thanos) Nenes

Laboratory of atmospheric processes and their impacts



<http://lapi.epfl.ch>

<http://cstacc.iceht.forth.gr>



Biogeochemical Cycles



Aerosol – Cloud – Climate Interactions



Air Quality and Health



Aerosol Chemistry and Impacts



ReCLEAN





## LAPI – Athanasios (Thanos) Nenes

### Laboratory of atmospheric processes and their impacts



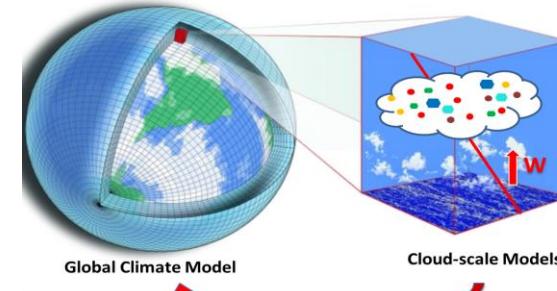
<http://lapi.epfl.ch>

<http://cstacc.iceht.forth.gr>

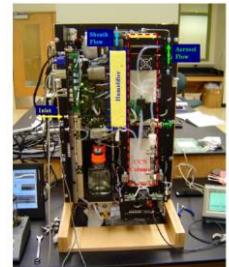
### Field and Laboratory Observations



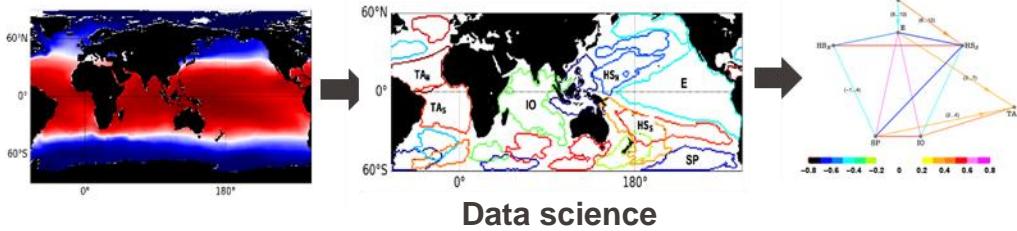
### Modeling



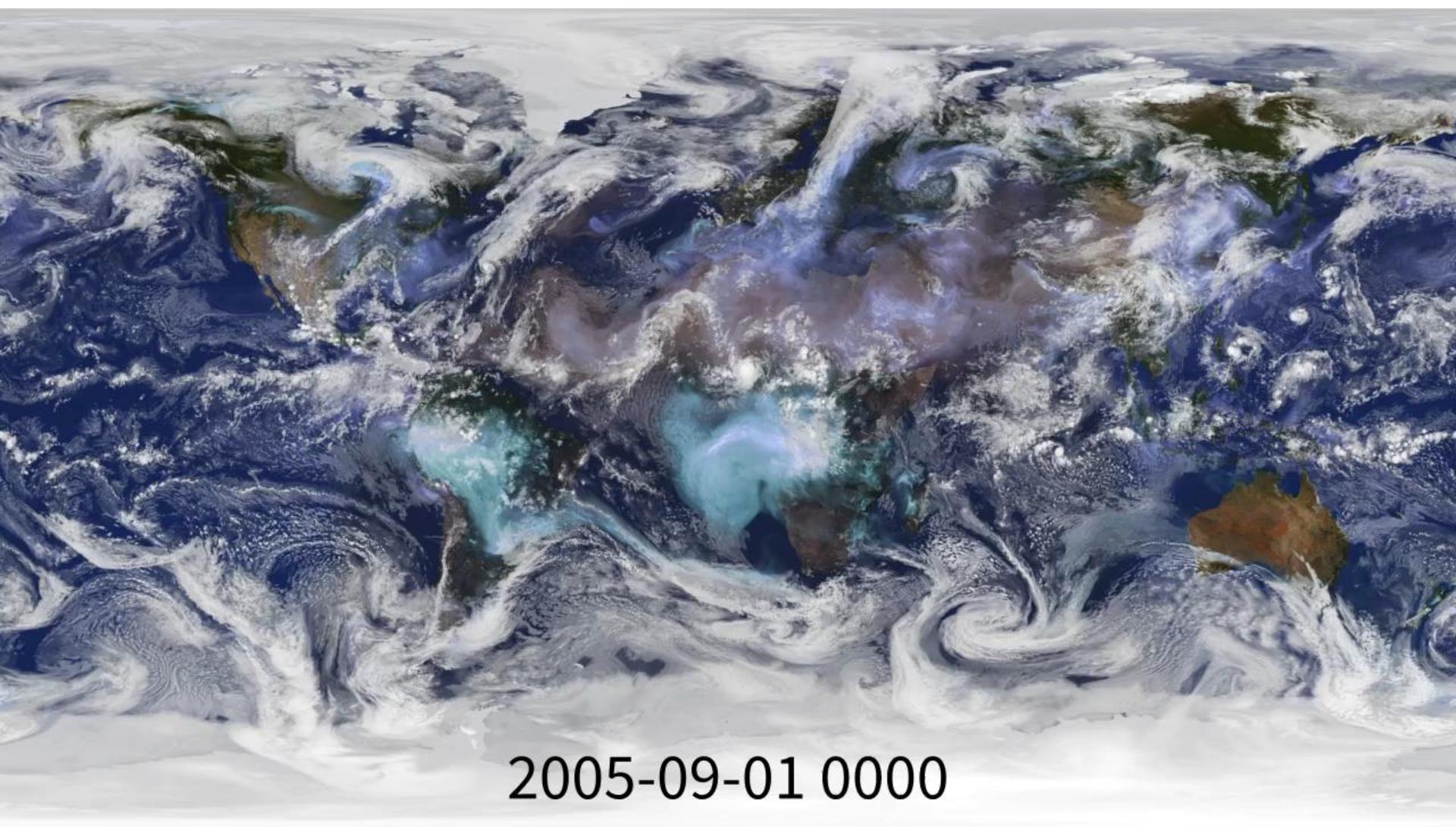
### Instrumentation



Cloud Condensation Nuclei Counter, US Patent 7,656,510

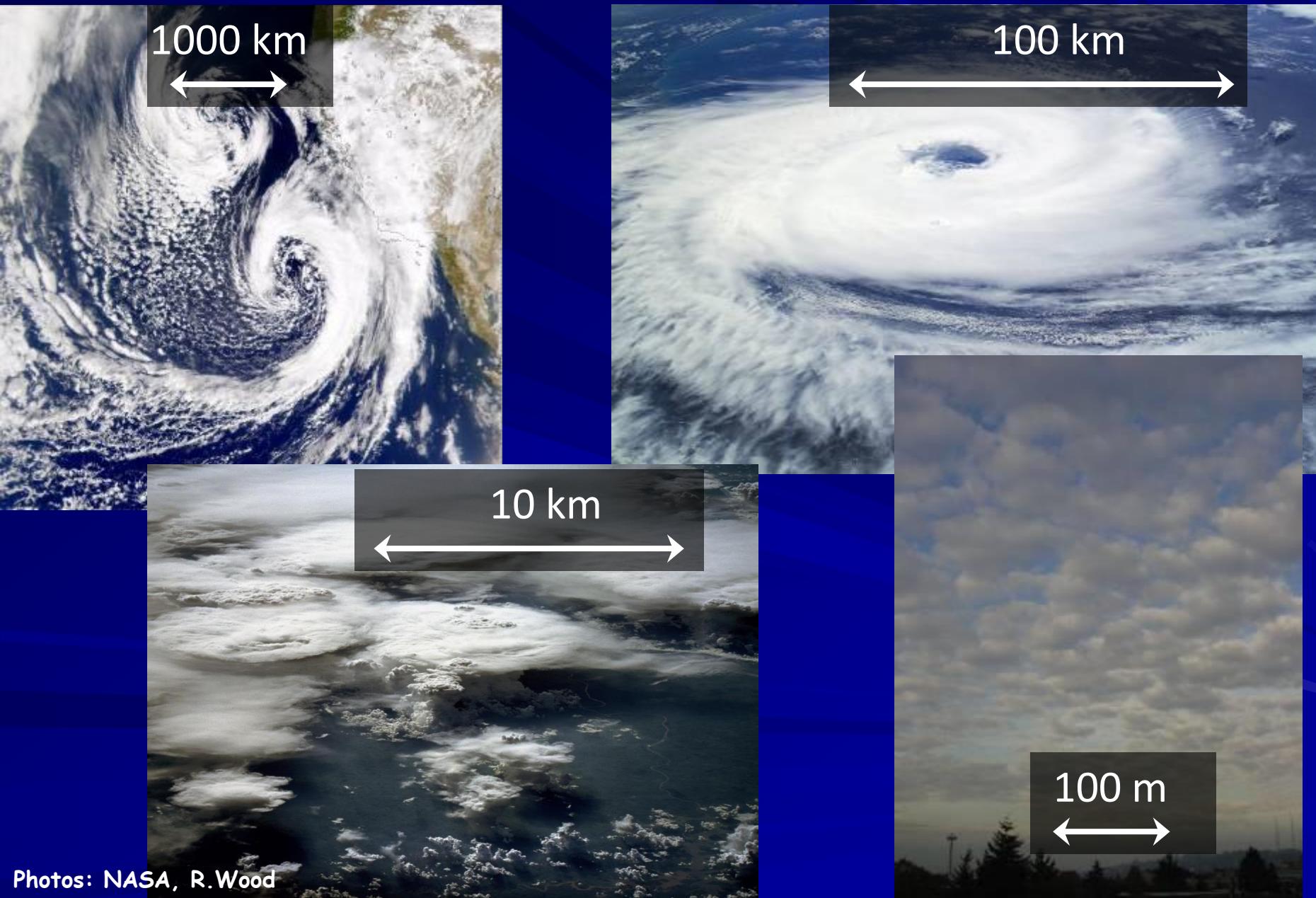


**Clouds are everywhere and at all scales...**



2005-09-01 0000

# Clouds are everywhere and at all scales...



Clouds have an important **radiative** impact.

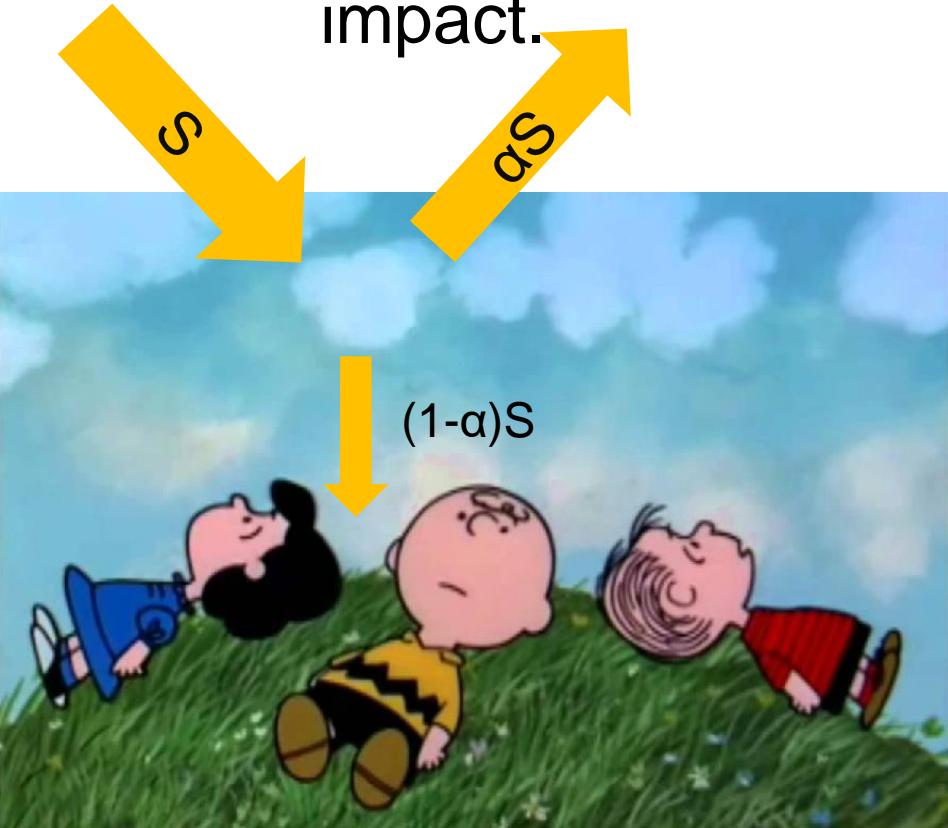
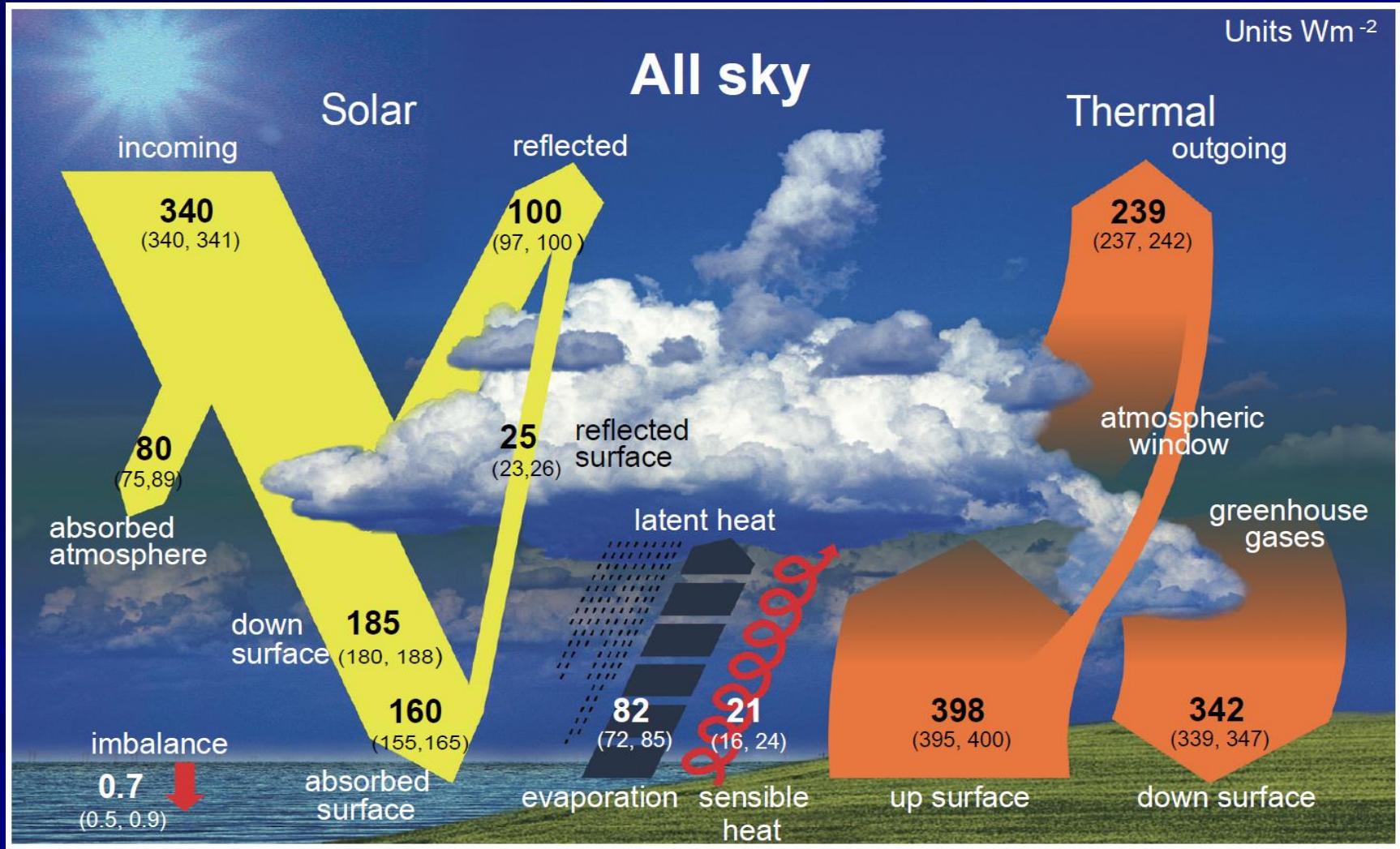


Photo from Wynn Bullock

Clouds also have an important **hydrological** impact.

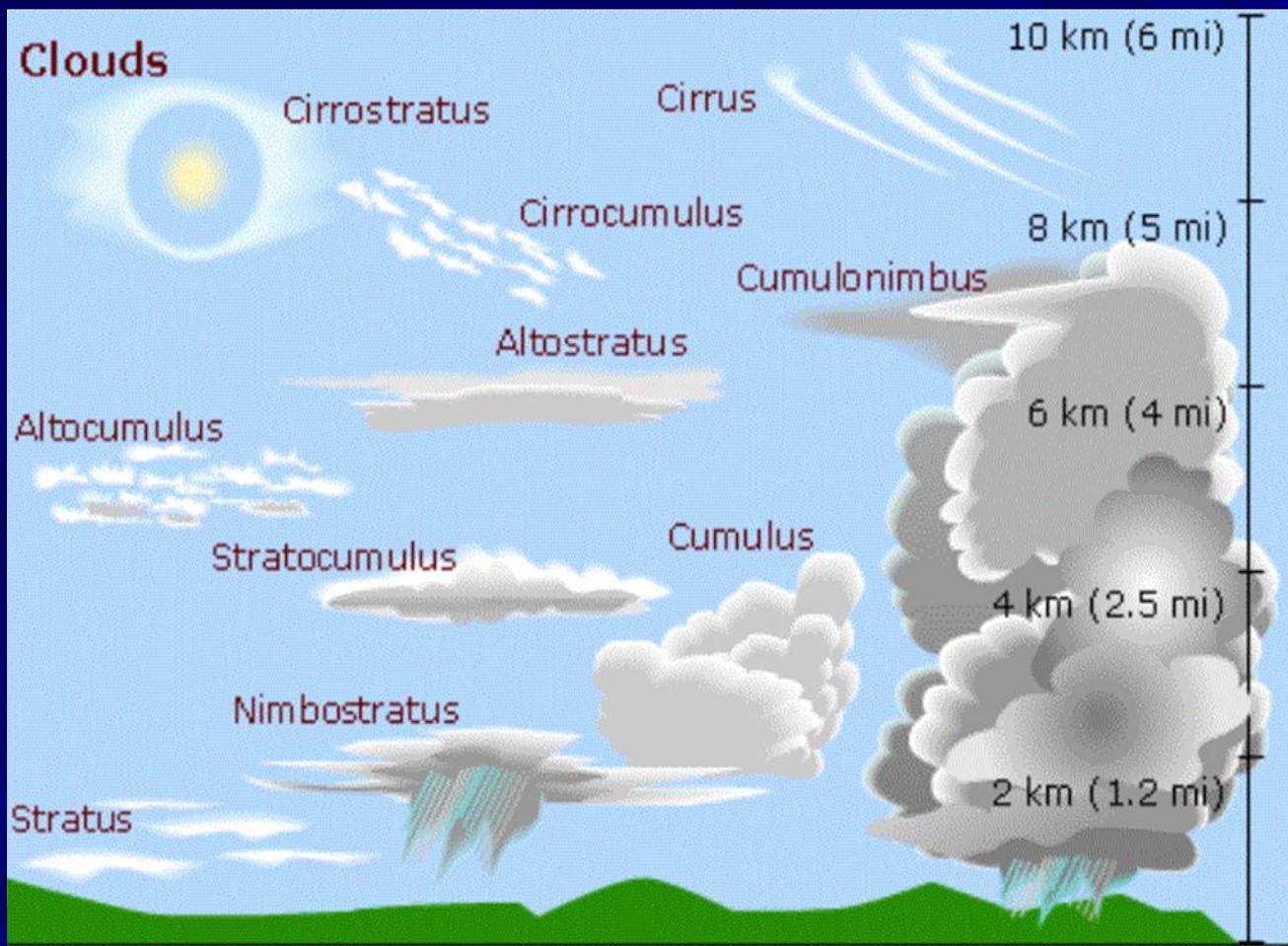
Both critically important for regional and global climate

# Clouds play a central role in the climate system



Based on J.T. Houghton: "The science of climate change"

# Cloud impacts vary a lot

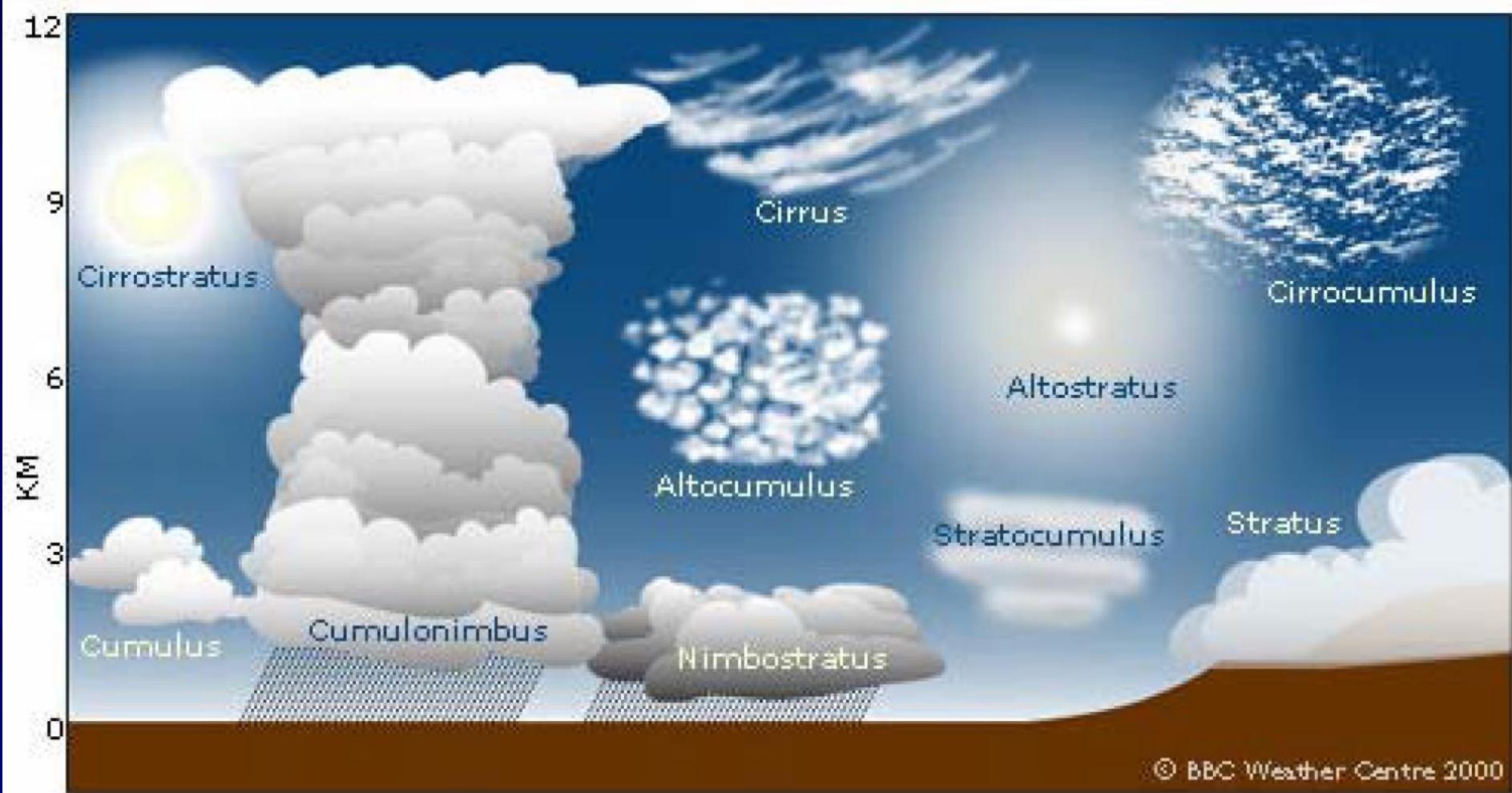


High clouds  
(ice crystals):  
*warm climate*

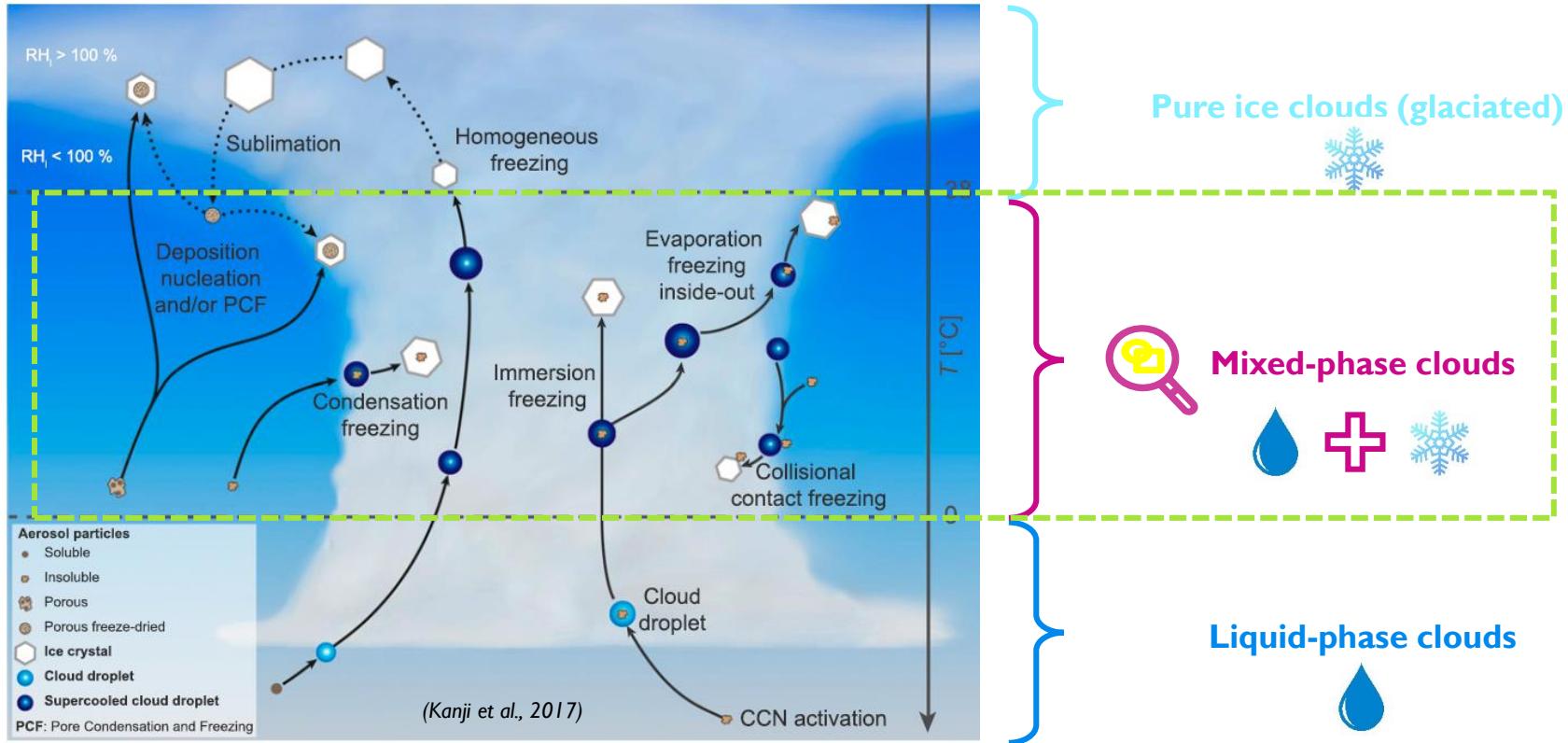
Mid-level:  
**Warm/cool**

Low clouds  
(liquid drops):  
*cool climate*

## Common Cloud Names, Shapes, and Altitudes:

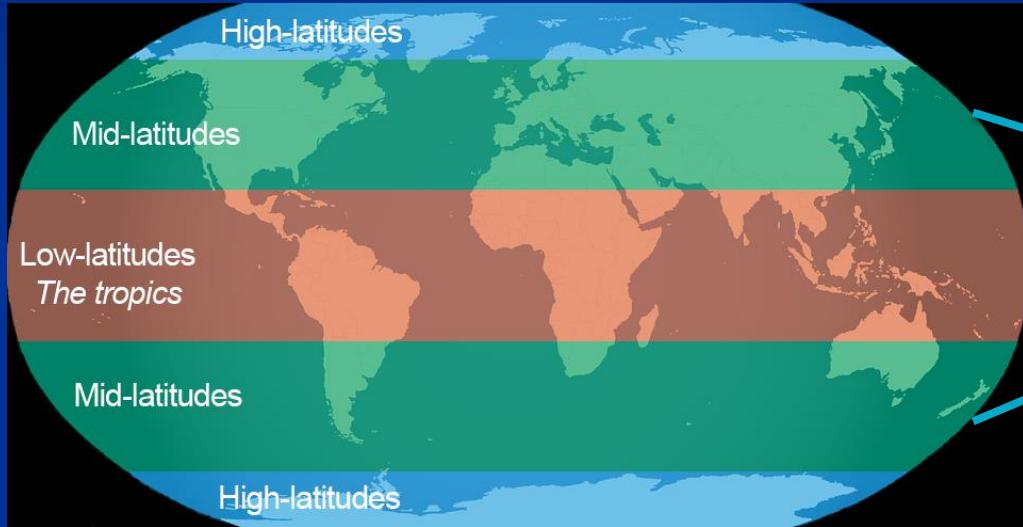


# Clouds types in the atmosphere



Atmospheric Particles (“aerosol”) are the seeds for cloud formation  
Aerosol/Cloud/Climate interactions are a major source of uncertainty in climate projections

# Liquid+ice ("mixed-phase") clouds Are very important for climate



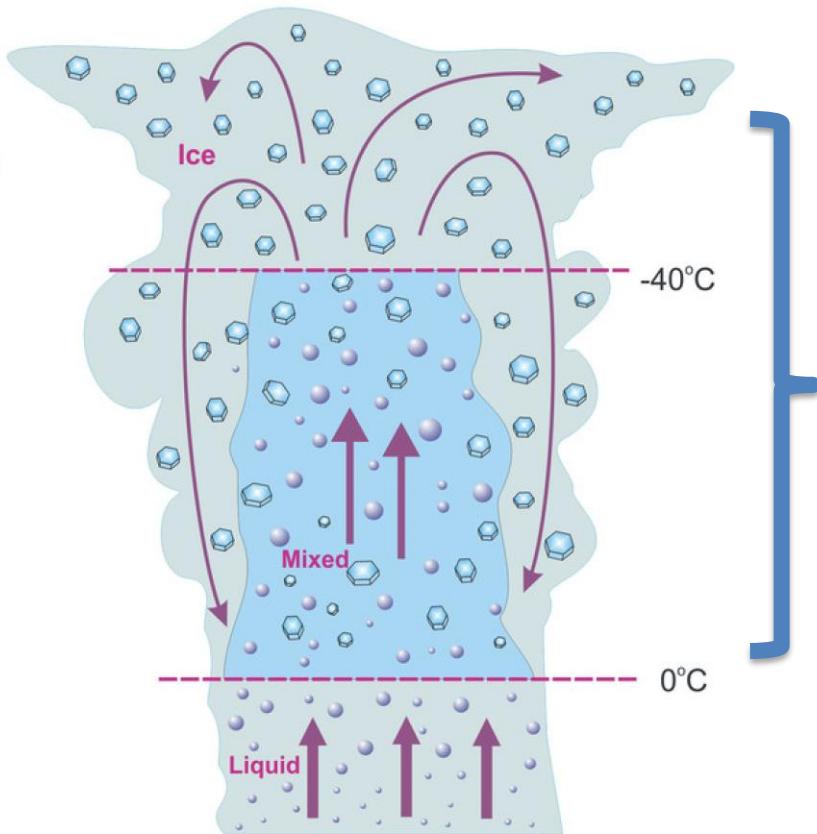
30-50% of precipitation  
occurs from the ice  
phase



*Field and Heymsfield, 2015*  
*Müllmenstädt et al. 2015*

*“...much of what is rain, when it arrives at the surface of the Earth, might have been snow, when it began its descent . . .”*

# Mixed-Phase clouds are important for extremes and control precipitation on a regional and global scale

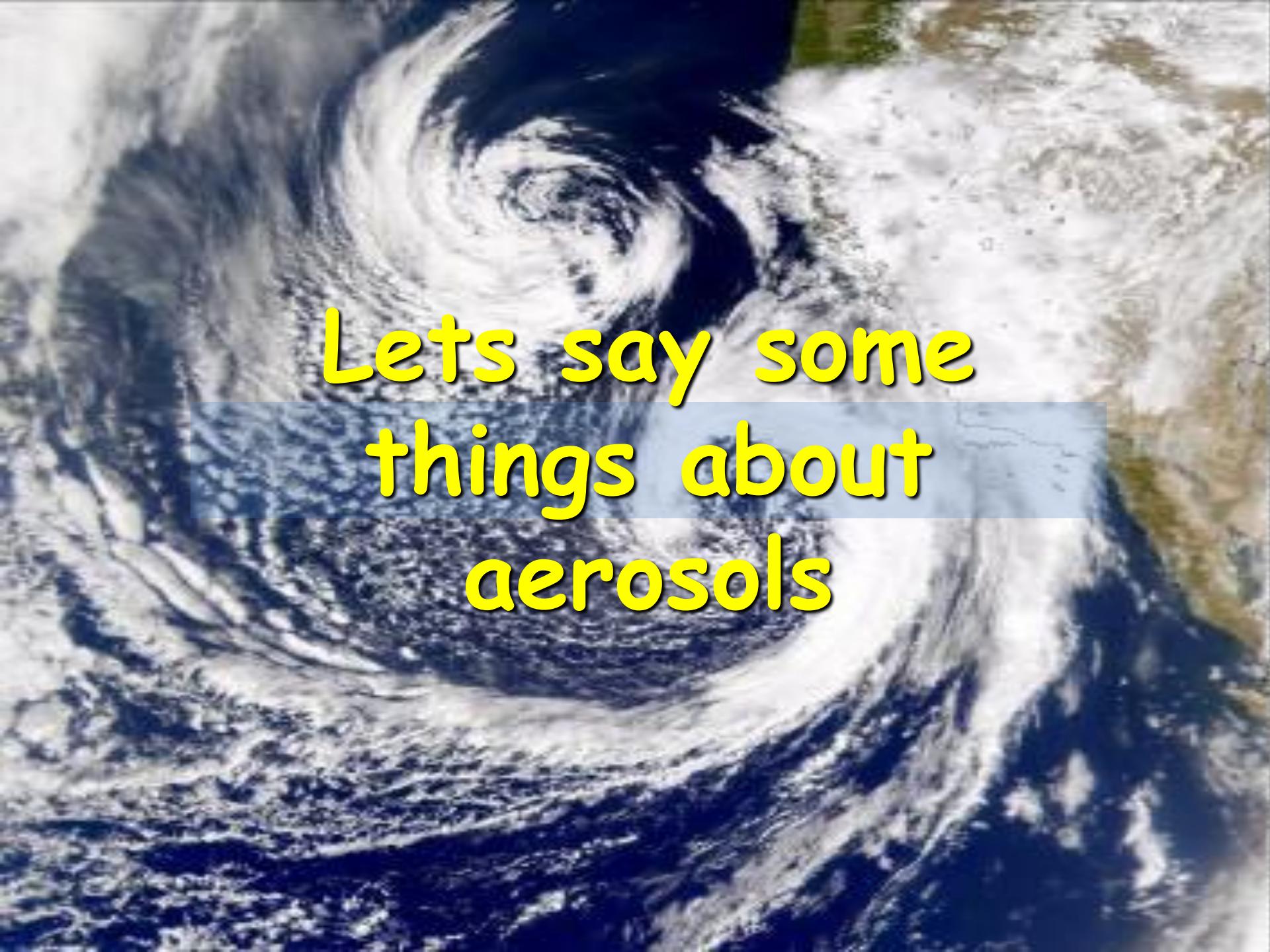


Precipitation at mid- and high-latitudes mostly generated from the mixed- and ice- cloud phase

*Mulmenstadt et al.*  
2015

Precipitation extremes have huge impacts on economy and society at large.





Lets say some  
things about  
aerosols

# Aerosol sizes and “names”

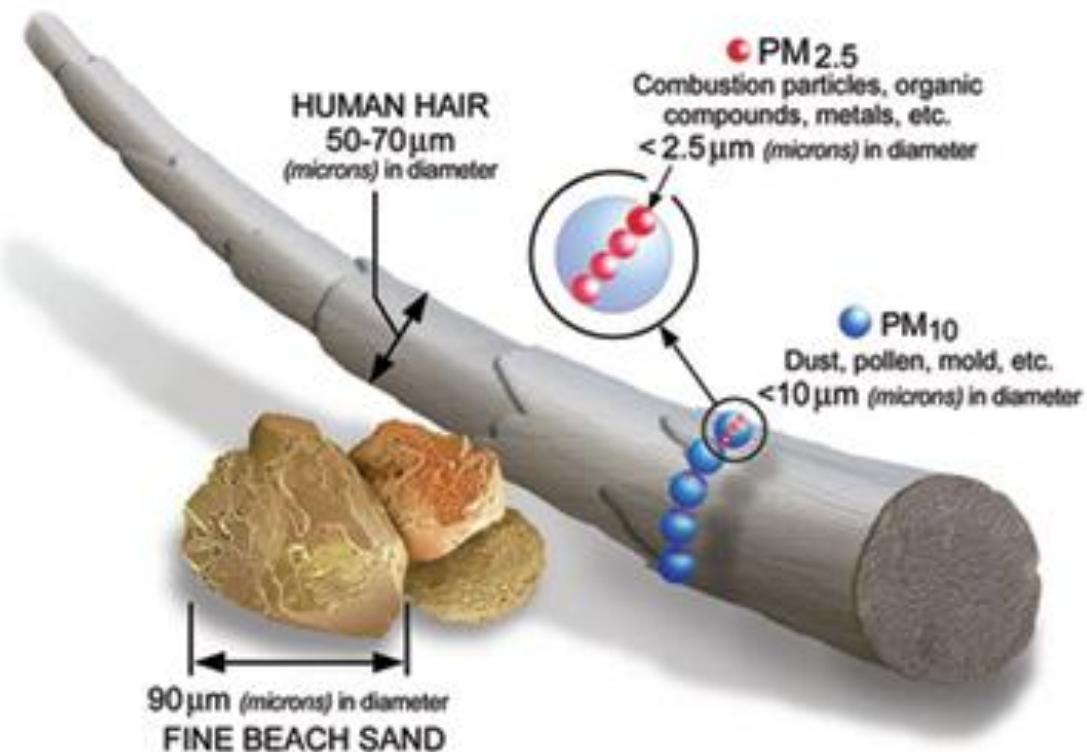


Image courtesy of the U.S. EPA

# Origins of Aerosol



## Primary emissions

Automobiles, industry, domestic, vegetation, forest fires, seasalt, ...

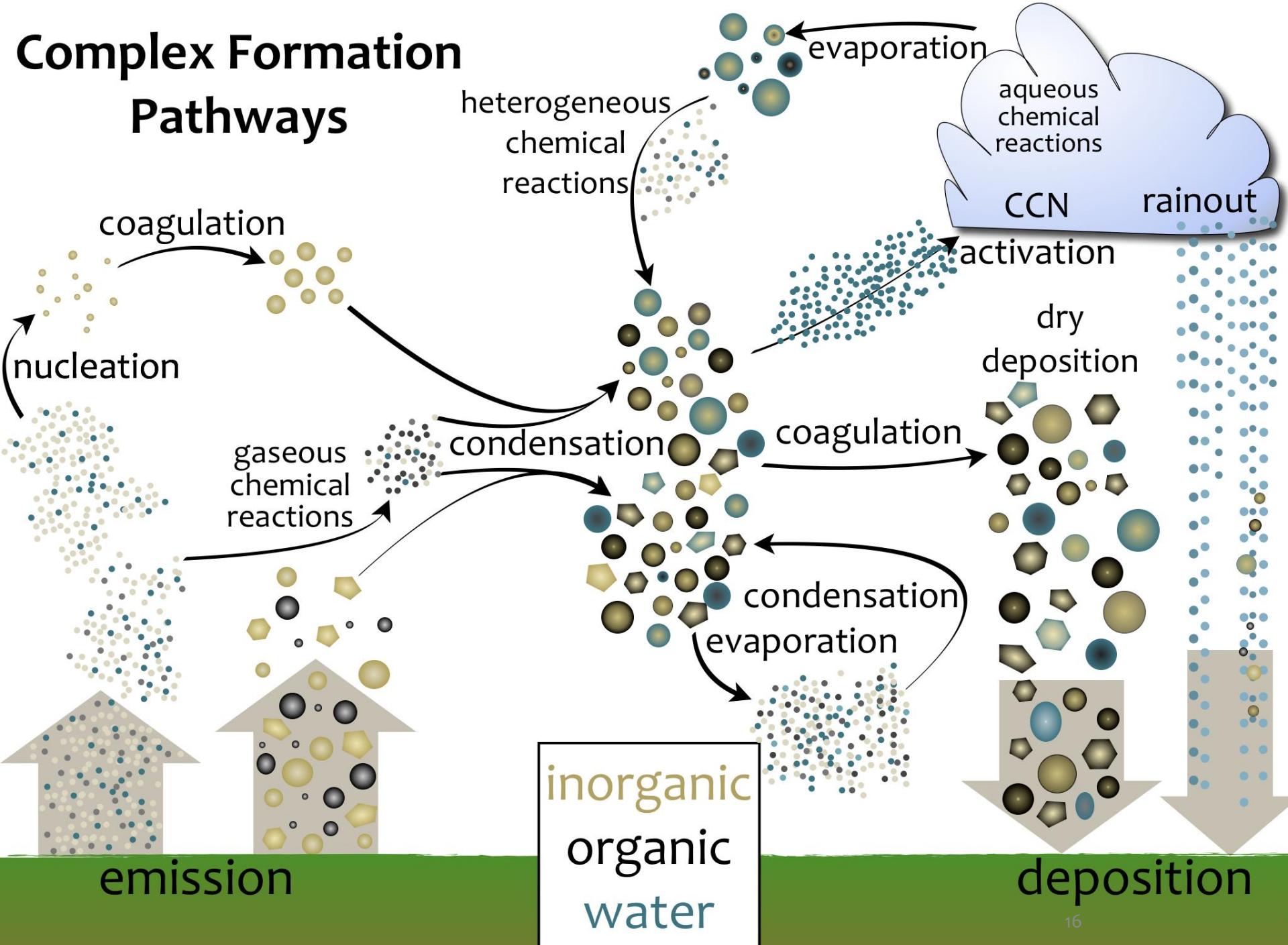
## Secondary compounds

Oxidation of precursors (by  $O_3$ ,  $H_2O_2$ ,  $OH$ ,  $NO_3$ , etc.) generates organic compounds.

Reaction of volatile bases ( $NH_3$ ) with acids, dust and seasalt form  $(NH_4)_2SO_4$ ,  $NH_4NO_3$  and many other salts.



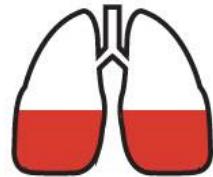
# Complex Formation Pathways



# Why do we care about aerosols? They can kill you

## THE **INVISIBLE KILLER**

Air pollution may not always be visible, but it can be deadly.



**29%**

OF DEATHS FROM  
**LUNG CANCER**



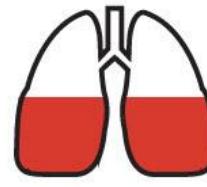
**24%**

OF DEATHS FROM  
**STROKE**



**25%**

OF DEATHS FROM  
**HEART DISEASE**



**43%**

OF DEATHS FROM  
**LUNG DISEASE**

- The WHO estimates that 4.2 million people die prematurely every year due to ambient (outdoor) air pollution.
- Pollutants with the strongest evidence for public health concern, include particulate matter (PM), ozone ( $O_3$ ), nitrogen dioxide ( $NO_2$ ) and sulphur dioxide ( $SO_2$ ).

**BREATHE LIFE.**  
Clean Air. Healthy Future.



World Health  
Organization

UN  
environment



CLIMATE &  
CLEAN AIR  
COALITION  
TO REDUCE SHORT-LIVED  
CLIMATE POLLUTANTS

# Why do we care about aerosols? They reflect/absorb sunlight & affect climate



Source: NASA

Dust and smoke over  
East Mediterranean  
(cooling + heating)

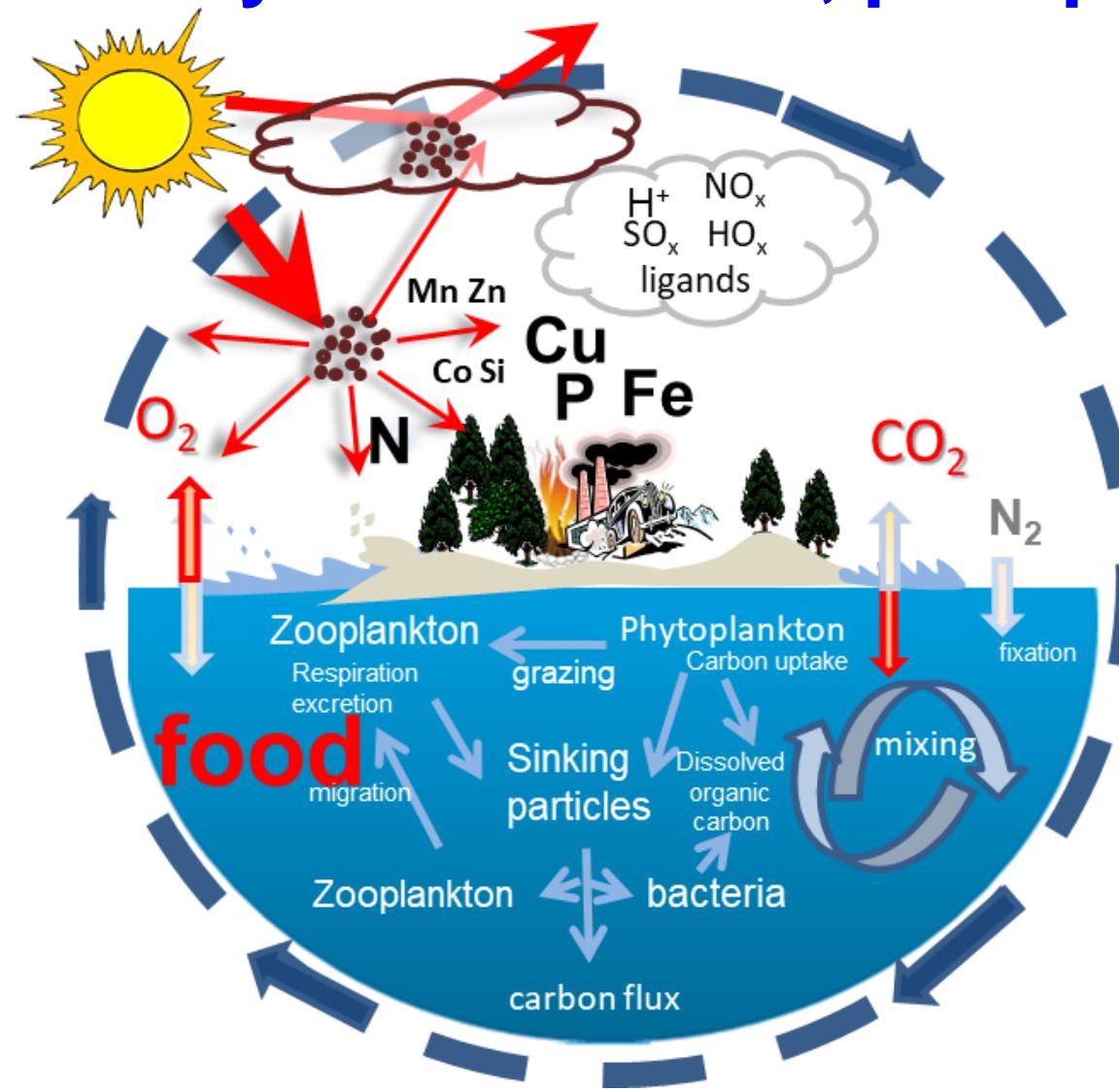


Source: Jay Apt (via P.Adams)

Soot from Kuwaiti  
oil fires  
(heating effect)

# Why do we care about aerosols?

## They affect clouds, precipitation & climate



Atmosphere is a major corridor for transporting nutrients between land and ocean.

**Anthropogenic emissions perturb nutrient fluxes considerably.**

**Large but very uncertain impacts on ecosystems, food, climate**

A satellite image of Earth's atmosphere, showing various cloud formations and aerosol plumes against the blue of the oceans and the green and brown of the continents.

Now let's switch  
to clouds and  
aerosols again!

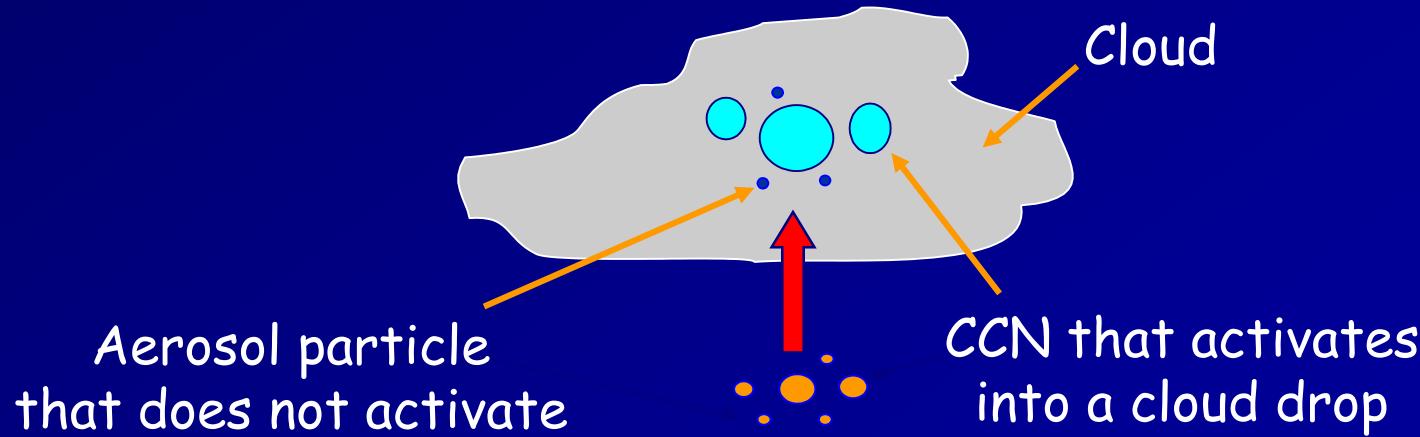
# How do (liquid water) clouds form?

Clouds form in regions of the atmosphere where there is too much water vapor (it is "supersaturated").

This happens when air is cooled (primarily through expansion in updraft regions and radiative cooling).

Cloud droplets nucleate on pre-existing particles found in the atmosphere (aerosols) with  $\sim 0.1\mu\text{m}$  diameter.

Aerosols that can become droplets are called cloud condensation nuclei (CCN).



# Ice formation mechanisms

Multiple mechanisms for ice formation can be active.

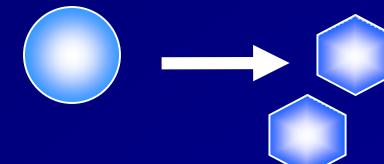
Wet aerosol  
particles



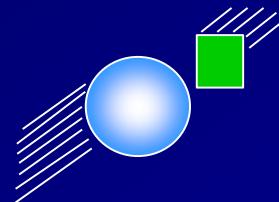
Soluble and  
Insoluble Aerosol  
Ice Nuclei (IN)  
Mostly dust, soot,  
and biological  
material



Homogeneous  
Freezing



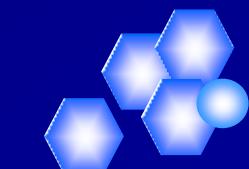
Immersion  
Freezing of cloud  
droplets ( $a_w \approx 1.0$ )



Contact  
(mostly at  
high T)

250 K

Deposition  
(low RH, low T)



Deliquesced -  
Heterogeneous  
Freezing (DHF)  
( $a_w < 1.0$ ; immersion in  
solution, condensation)

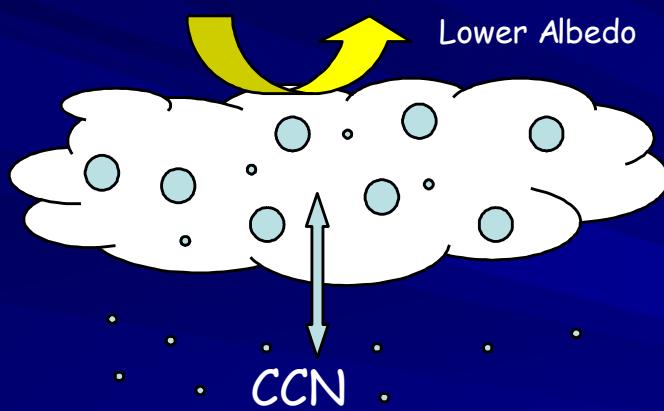
235 K

Increasing  $RH_i$ , decreasing T

273 K

# Increases in aerosol affects warm clouds

You make clouds that are "whiter", precipitate less (persist longer) and potentially cover larger areas of the globe. This is thought to yield a net cooling on climate and is termed as the **"indirect climatic effect of aerosols"**.

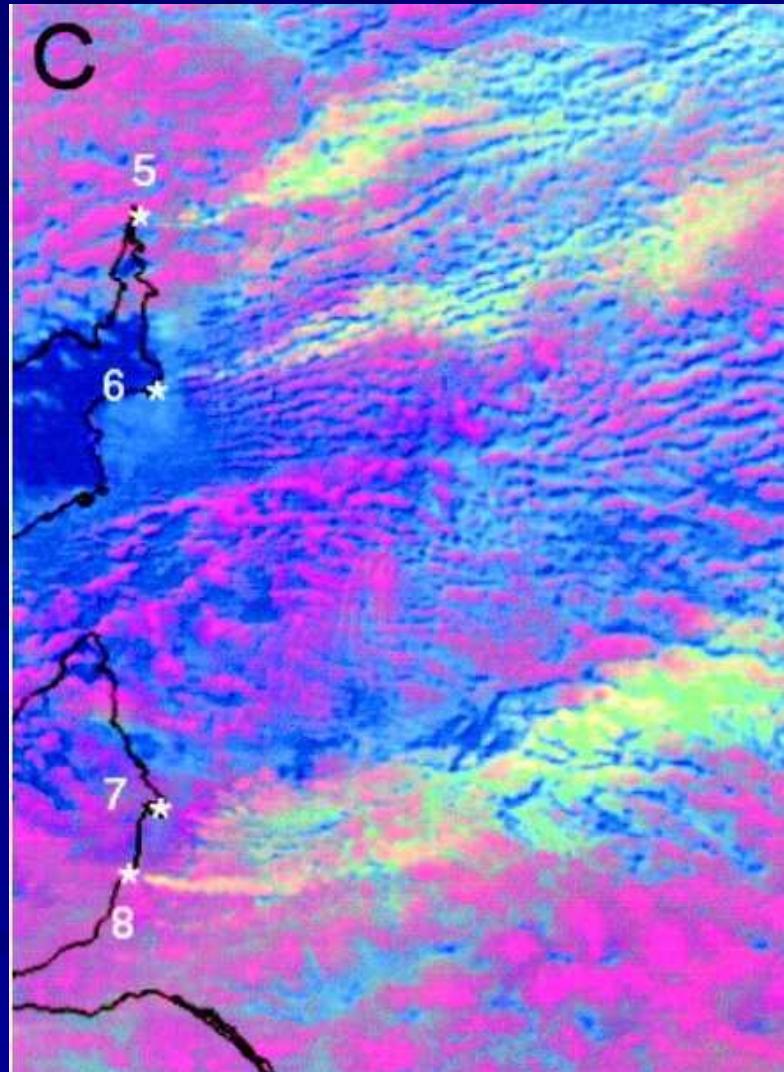


Increasing particles tends to cool climate (potentially a lot).  
Quantitative assessments done with climate models.

# Observational evidence of indirect effect

Satellite observations of clouds off W. Australia.

**Red:** Clouds with low reflectivity.  
**White:** Clouds that reflect a lot.  
**Blue:** Clear sky.



# Observational evidence of indirect effect

## Air pollution can affect cloud properties

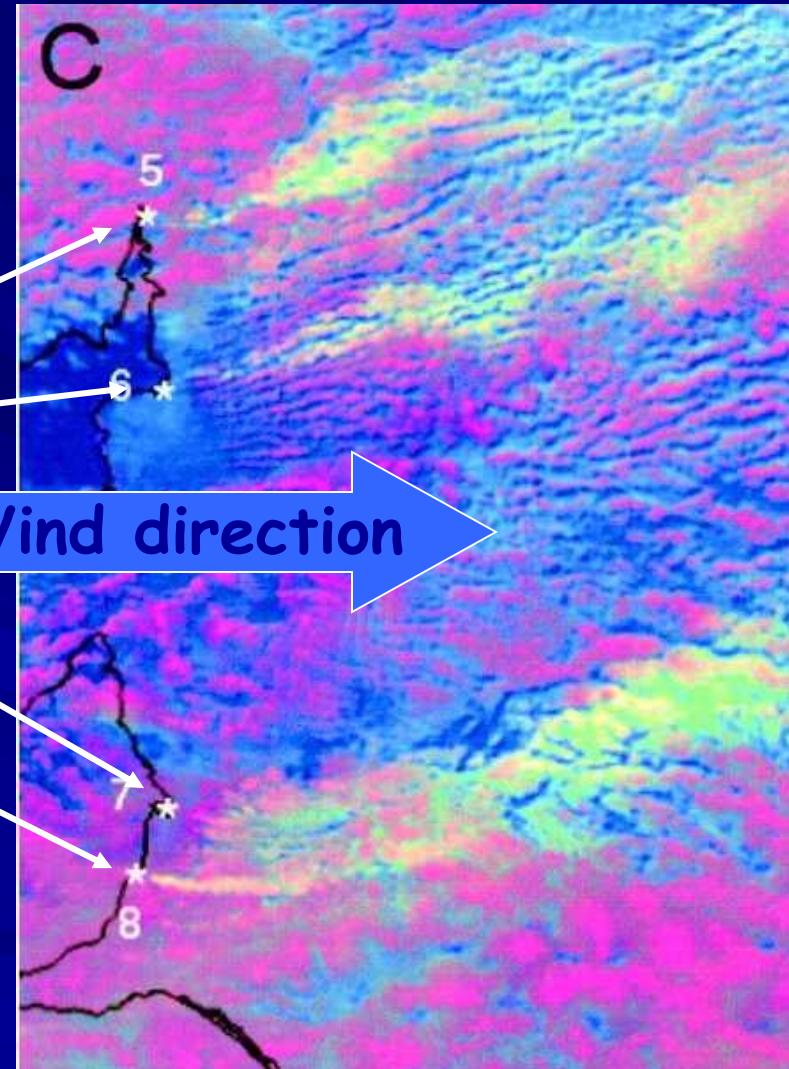
Satellite observations of clouds off W. Australia.

Power plant

Lead smelter

Port

Oil refineries



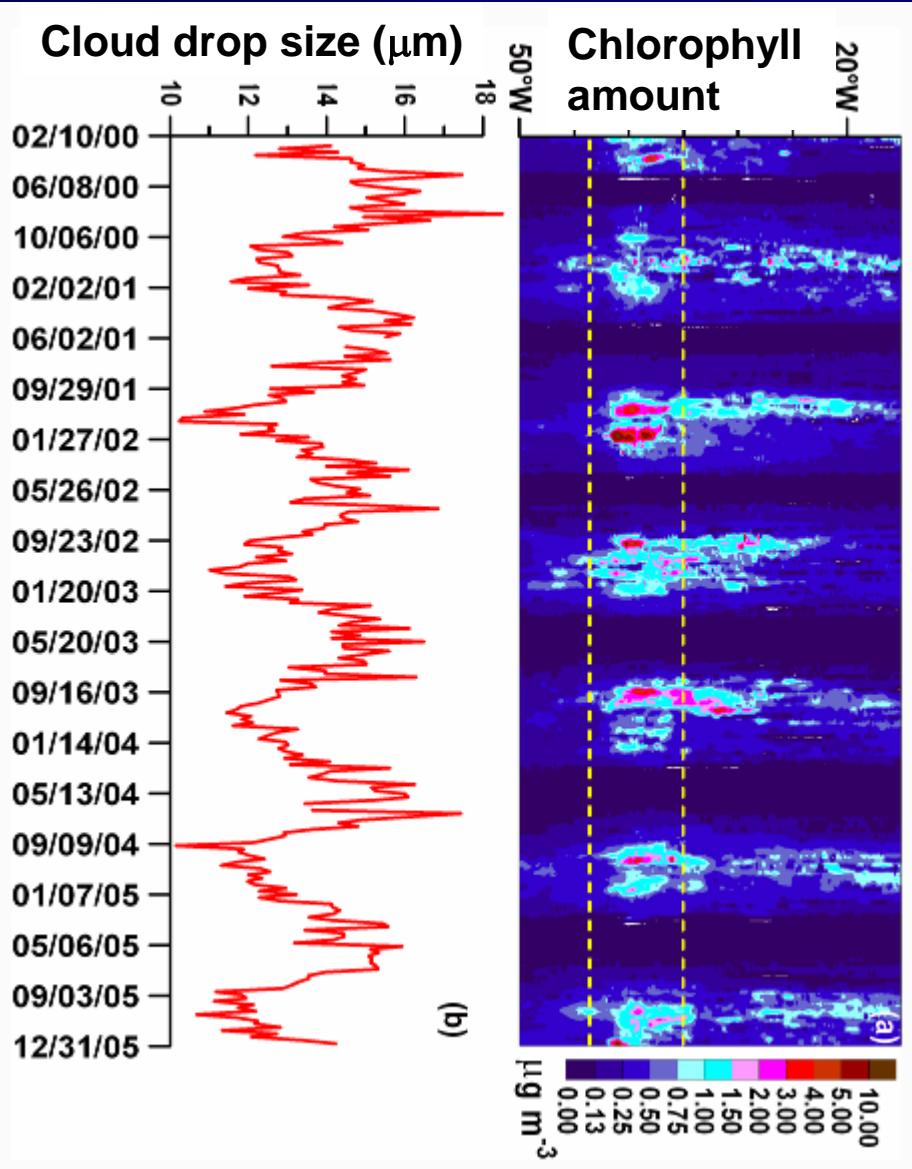
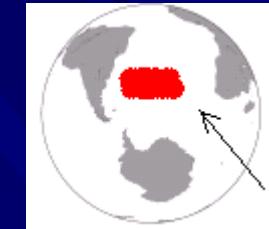
**Red:** Clouds with low reflectivity.

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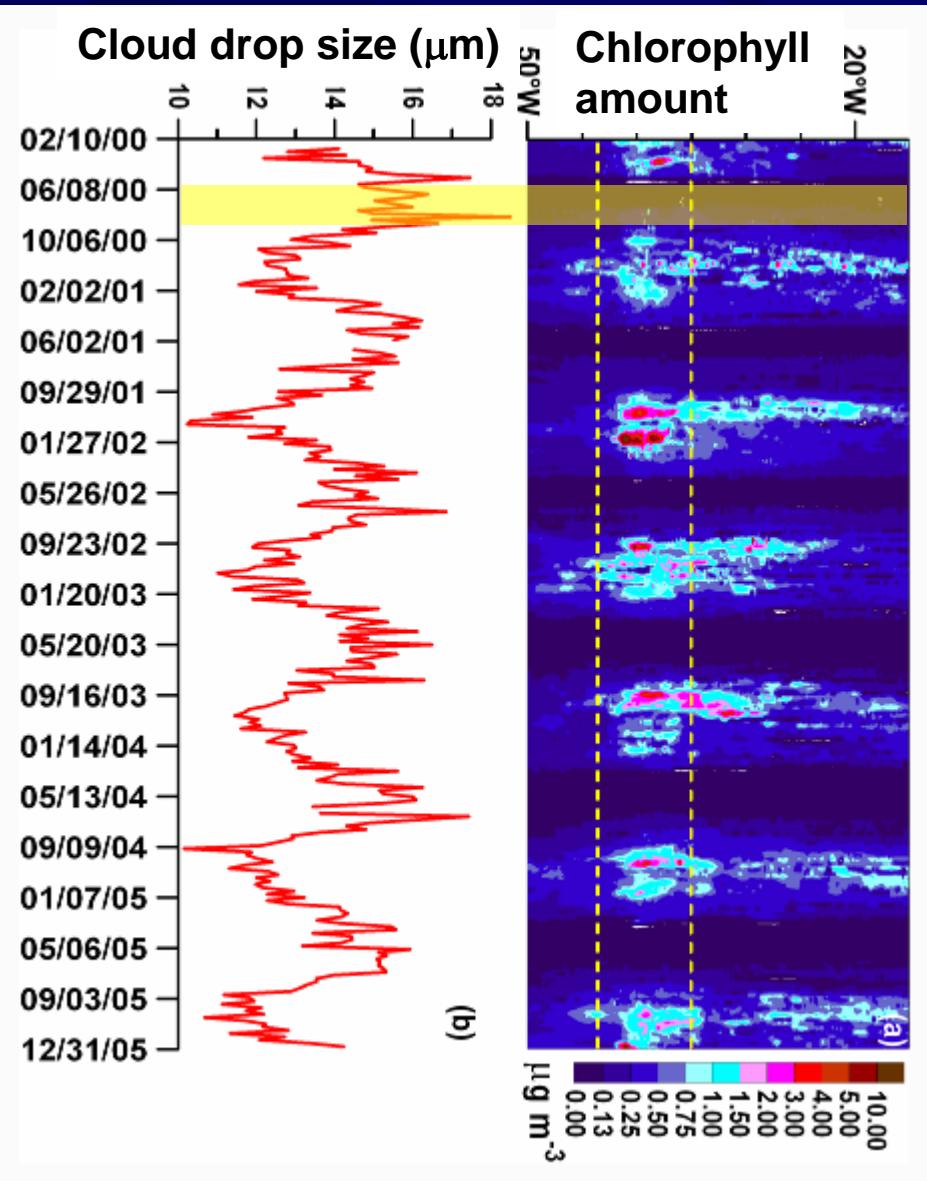
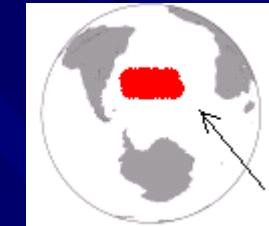
# Phytoplankton affect clouds too...

Location: East of Patagonia (South America)



# Phytoplankton affect clouds too...

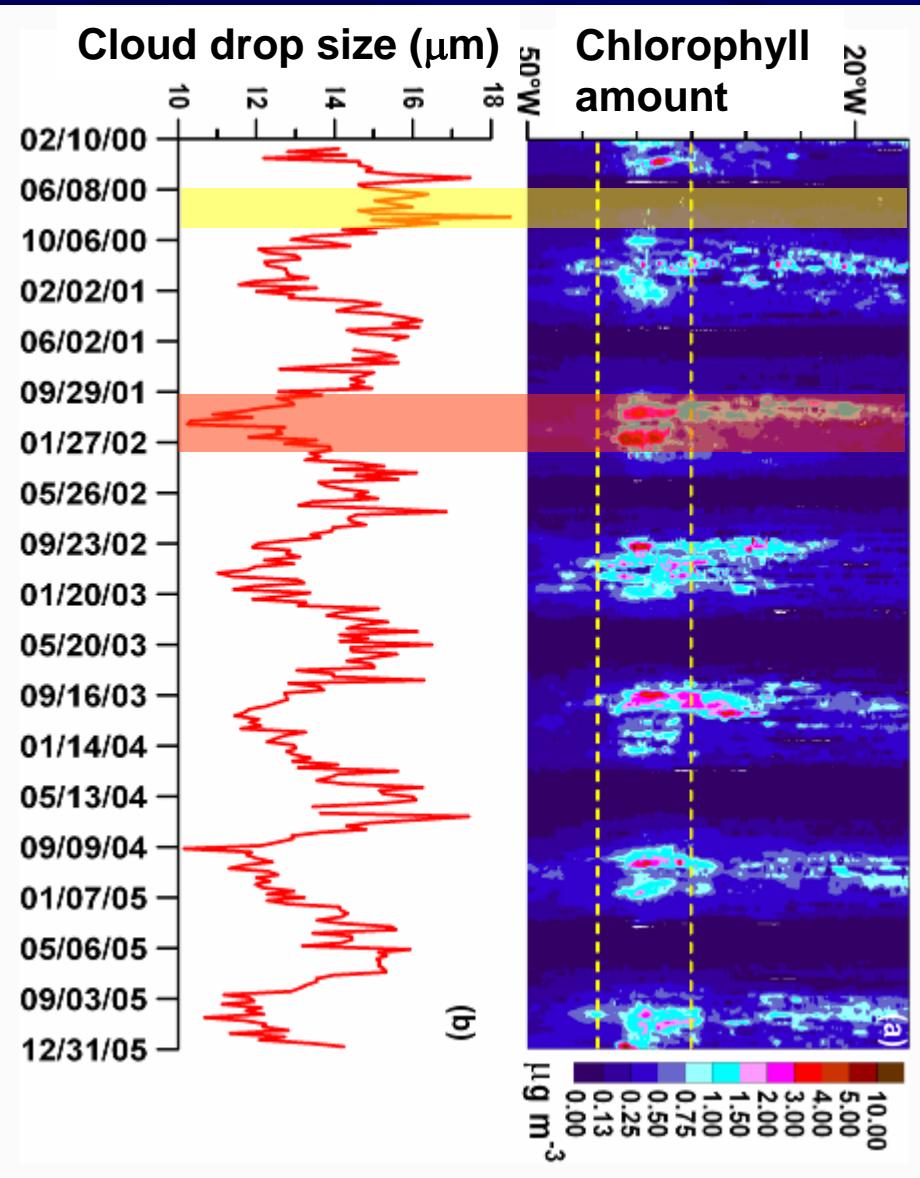
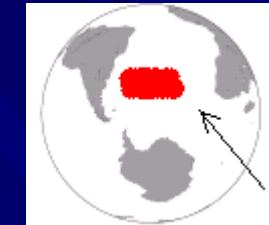
Location: East of Patagonia (South America)



Low chlorophyll period,  
clouds have large drops  
(not very reflective)

# Phytoplankton affect clouds too...

Location: East of Patagonia (South America)

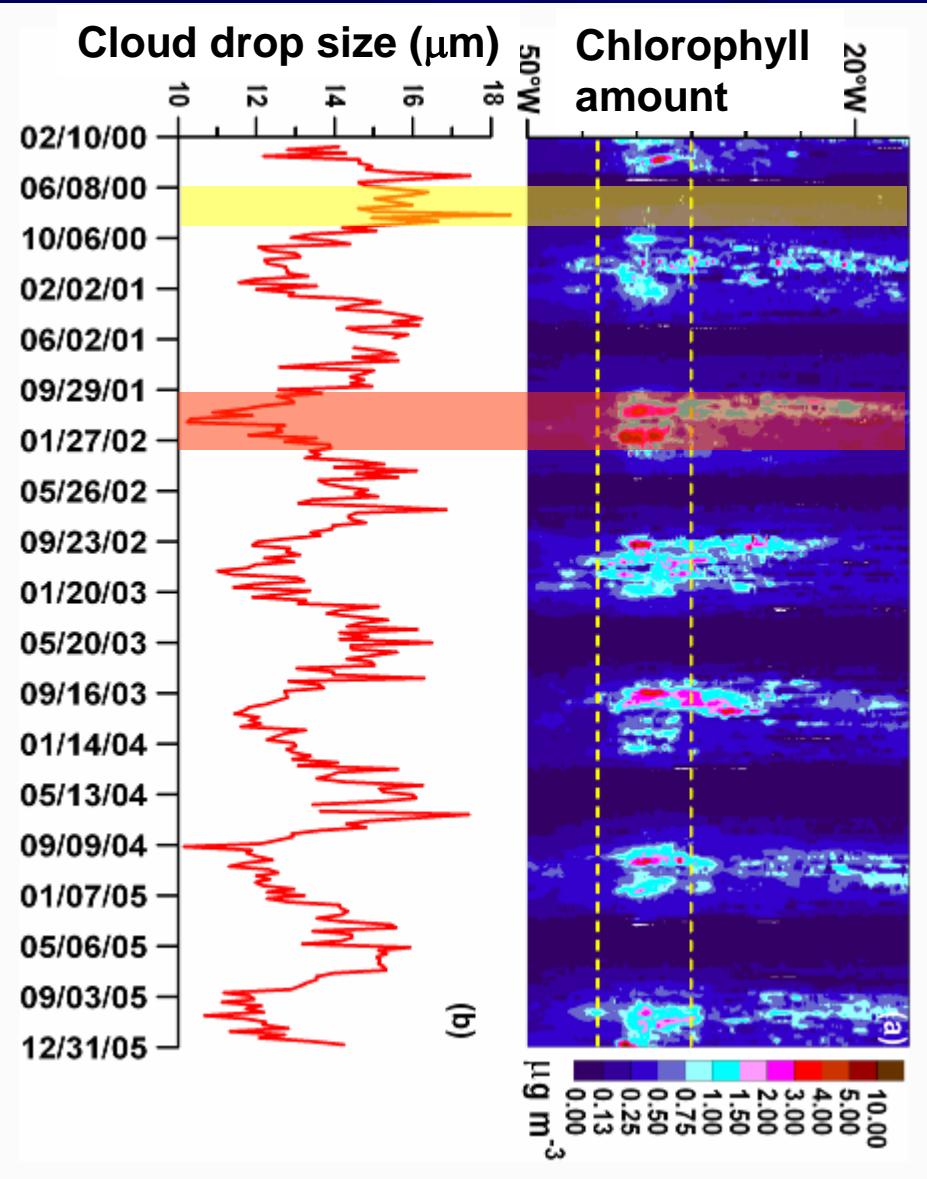
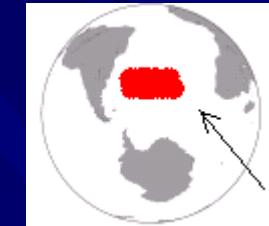


Low chlorophyll period,  
clouds have large drops  
(not very reflective)

High Chlorophyll period,  
Clouds have small drops  
(very reflective)

# Phytoplankton affect clouds too...

Location: East of Patagonia (South America)



Low chlorophyll period,  
clouds have large drops  
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High Chlorophyll period,  
Clouds have small drops  
(very reflective)

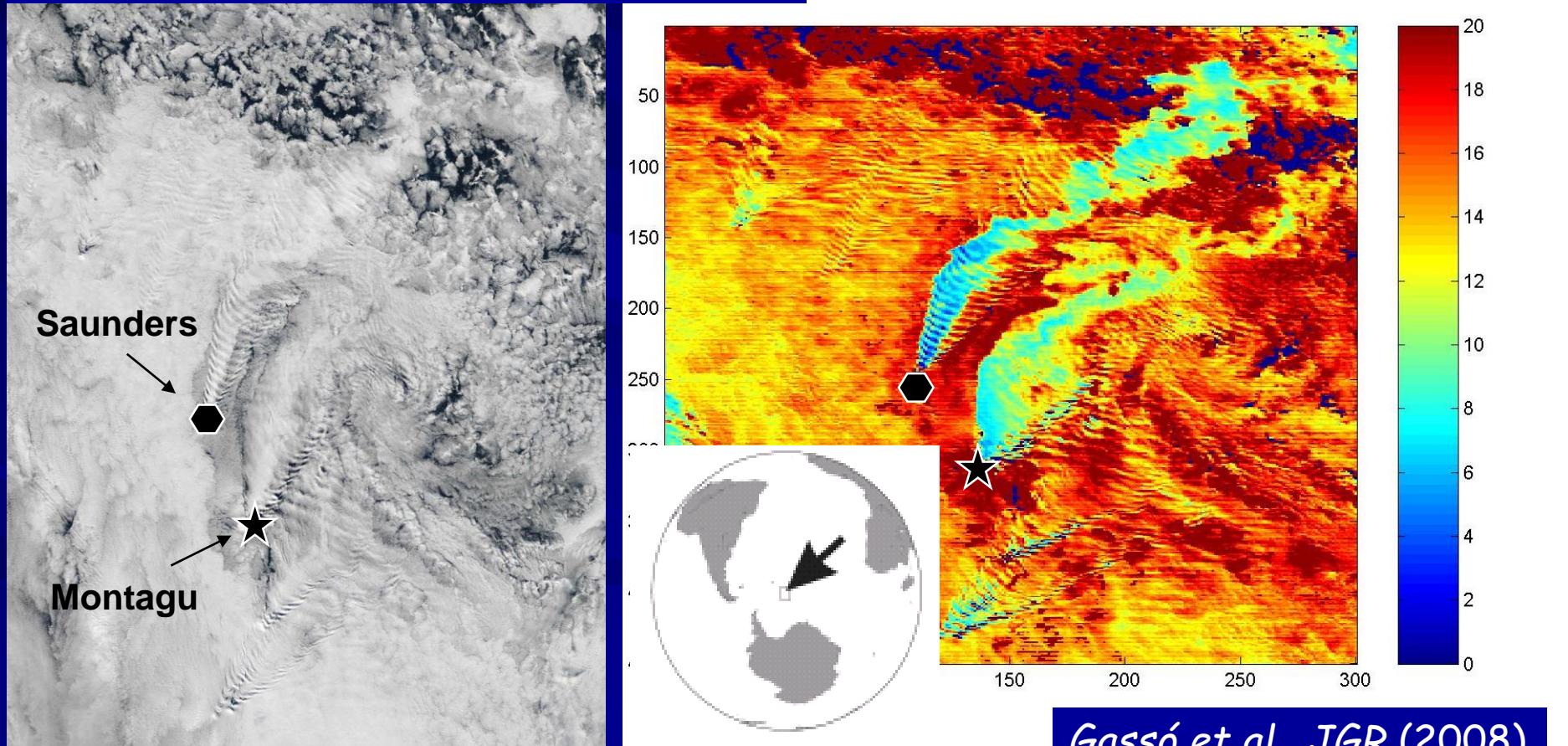
Phytoplankton emissions  
increase particle loads, and  
strongly impact clouds.

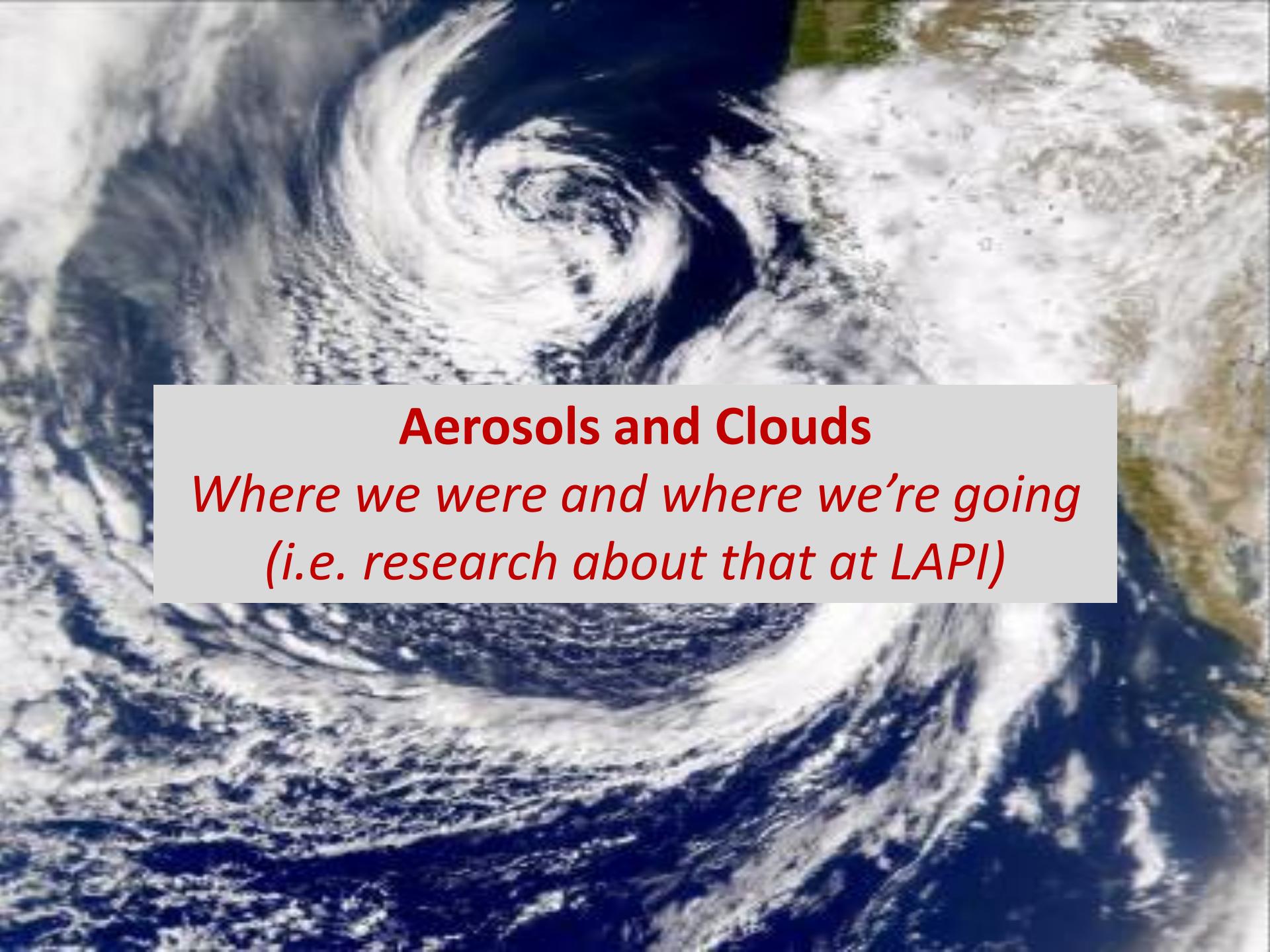
Biology-cloud interactions  
affect radiation in the region.

# So do volcanoes (even when “sleeping”) ...

Volcanoes continuously emit  $\text{SO}_2$  which becomes sulfate aerosol. The aerosol can substantially increase CCN in volcanic plumes. Clouds in the plume are much more reflective than outside.

Location: *Sandwich Islands*,  $\sim 55^\circ\text{S}$ ,  $\sim 30^\circ\text{W}$

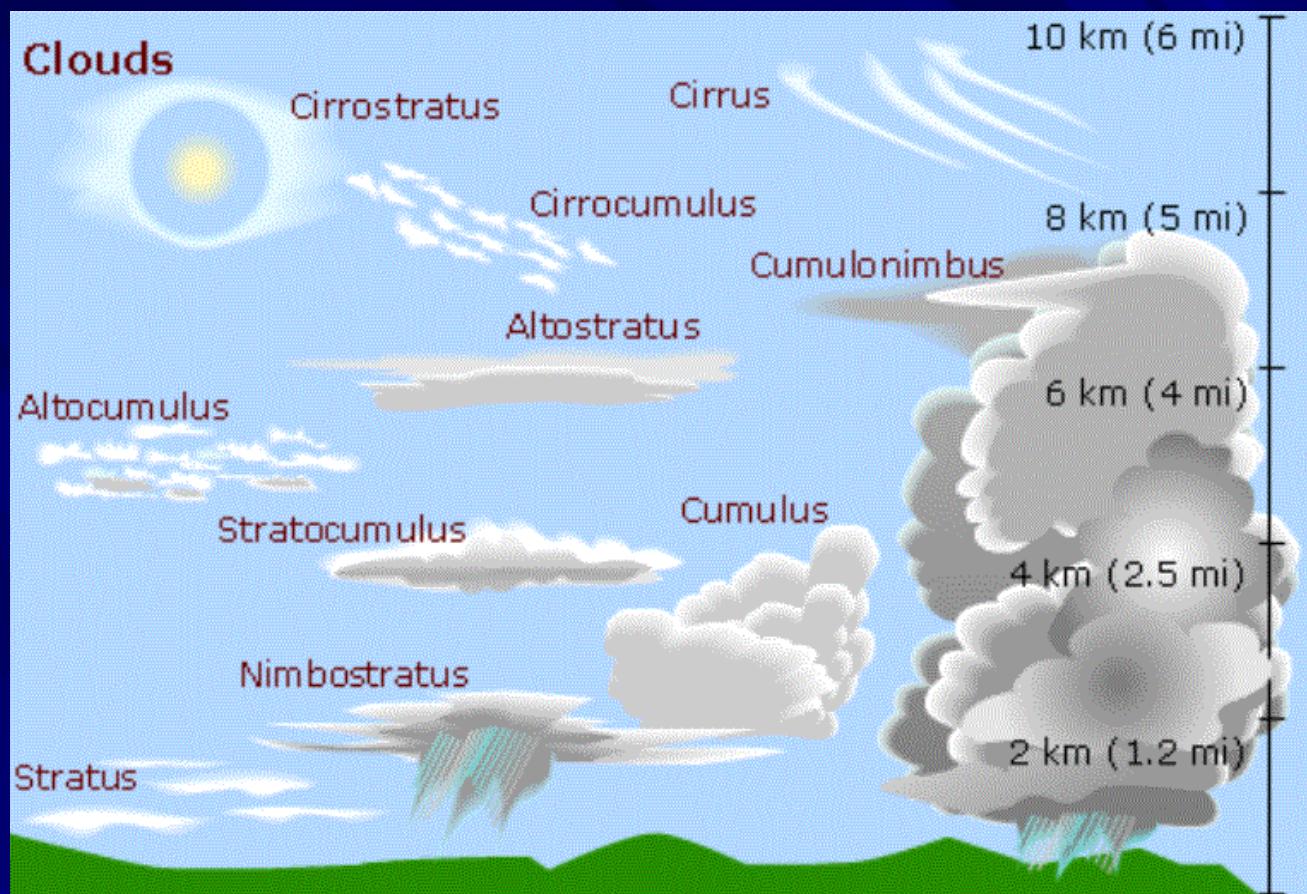




## Aerosols and Clouds

*Where we were and where we're going  
(i.e. research about that at LAPI)*

# Cloud types and phase



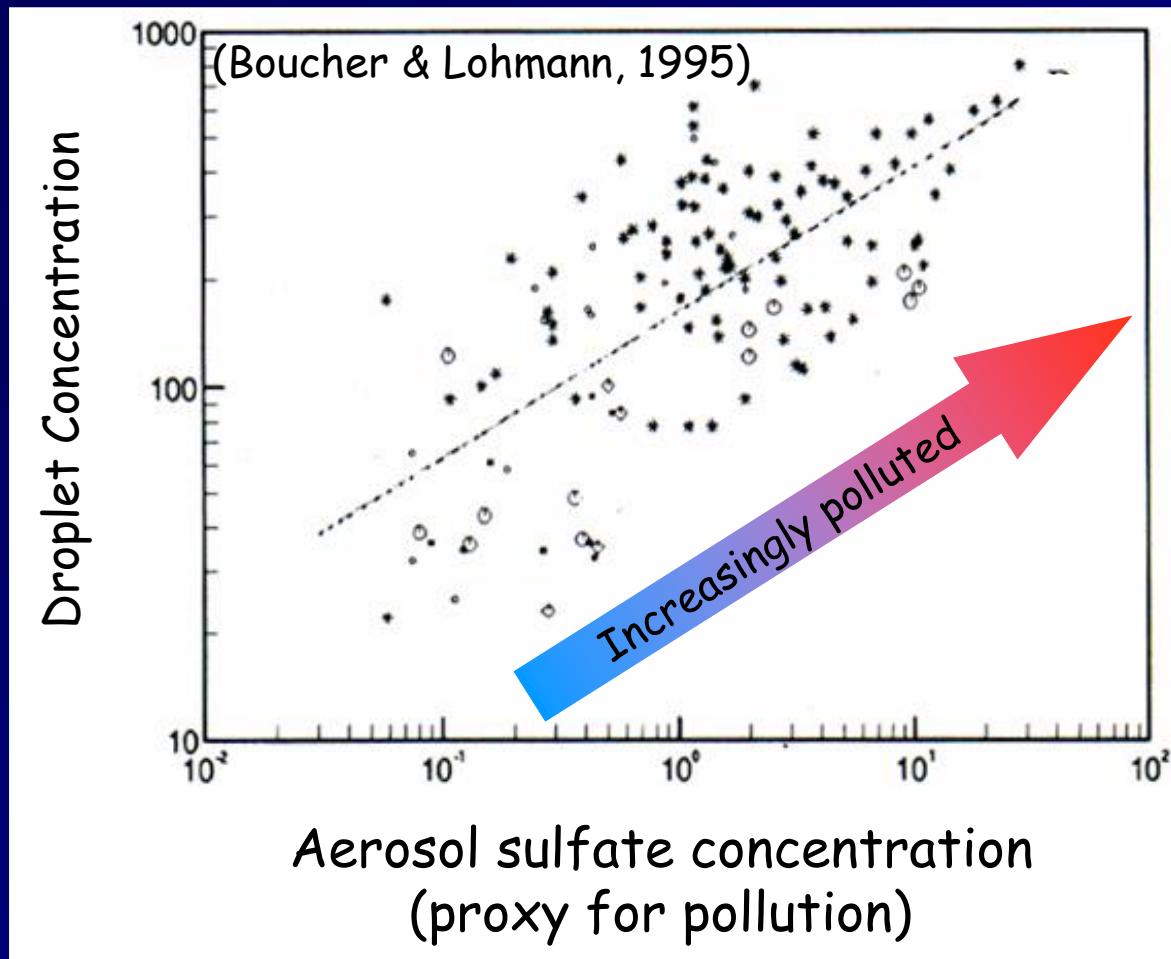
High clouds  
(ice crystals):  
*warm climate*

Mid-level:  
**Warm/cool**

Low clouds  
(liquid drops):  
*cool climate*

Cloud drops/crystals are not created directly from the vapor phase but form upon **airborne particulate matter (aerosol particles)**

# Aerosol-cloud interaction relationships: Major source of climate prediction uncertainty



Empirical approaches used....

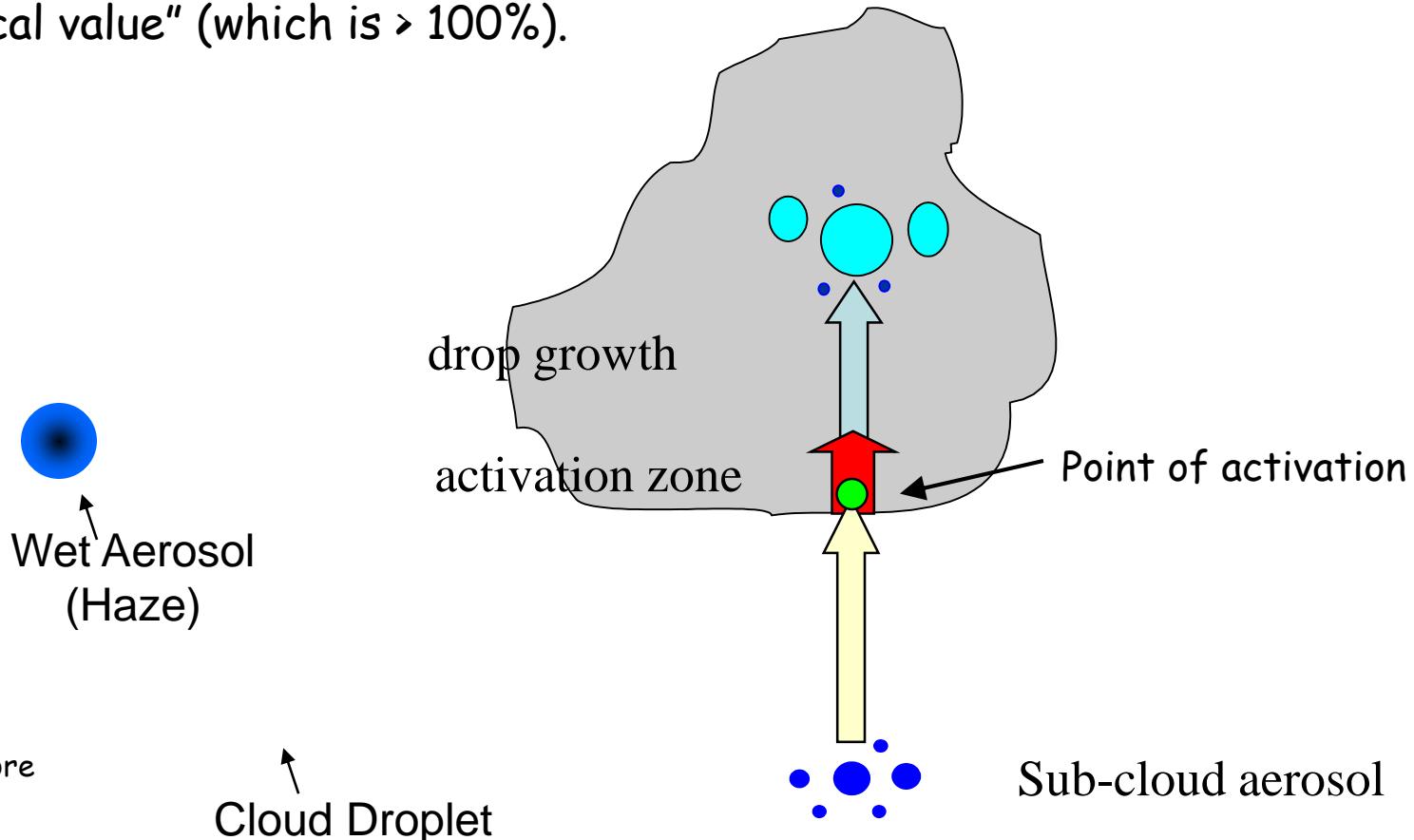
Large uncertainty by not accounting for:

- Meteorology
- Cloud microphysics
- Composition
- ...

For ice clouds, crystal numbers were simply prescribed ("tuned") to match satellite data

# “Mechanistic parameterization” provide the physical links required.

- To act as a CCN, each particle requires exposure to relative humidity above a “critical value” (which is  $> 100\%$ ).

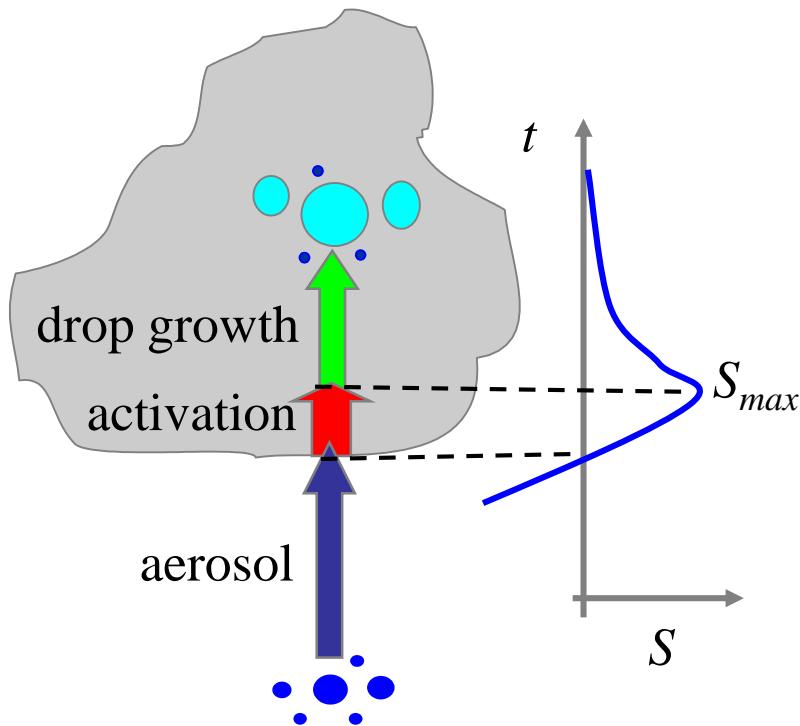


- For all this to work, you need to know the composition and size of each particle to get the CCN concentrations “right”.

# “Mechanistic parameterization” provide the physical links required.

Algorithm for calculating  $N_d$  :  
(Mechanistic parameterization)

1. Calculate  $s_{max}$  (approach-dependent)
2.  $N_d$  is equal to the CCN with  $s_c < s_{max}$



## Mechanistic Parameterizations:

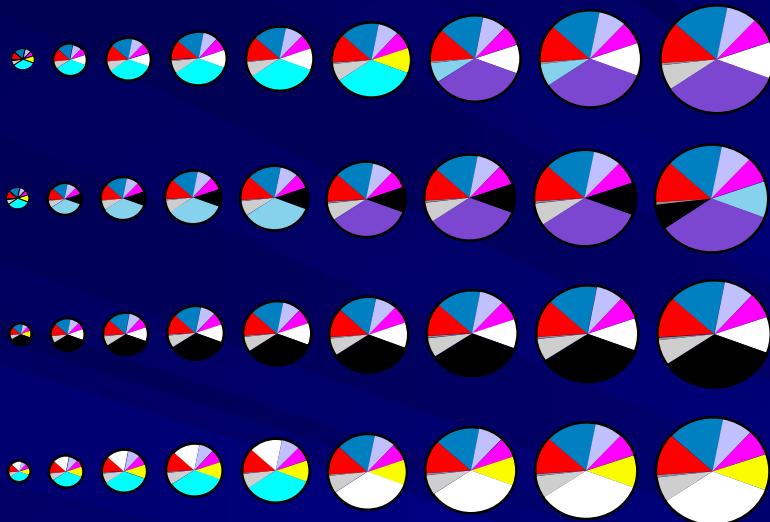
Twomey (1959); Abdul-Razzak et al., (1998); Nenes and Seinfeld, (2003); Fountoukis and Nenes, (2005); Kumar et al. (2009), Morales and Nenes (2014), and others.

**Input:** P, T, vertical wind, particle size distribution, composition.

**Output:** Cloud properties (droplet number, size distribution).

We have also done the same for ice (cirrus) clouds (Barahona et al., 2008, 2009ab) and doing it for mixed-phase clouds & secondary ice (Sotiropoulou et al., 2020, 2021; Georgakaki et al., in prep

# Aerosol Problem: Complexity



courtesy: S.Pandis

An integrated “soup” of

- Inorganics, organics (1000's)
- Particles can have uniform composition with size...
- ... or not
- Can vary vastly with space and time (esp. near sources)

## Organic species are a headache

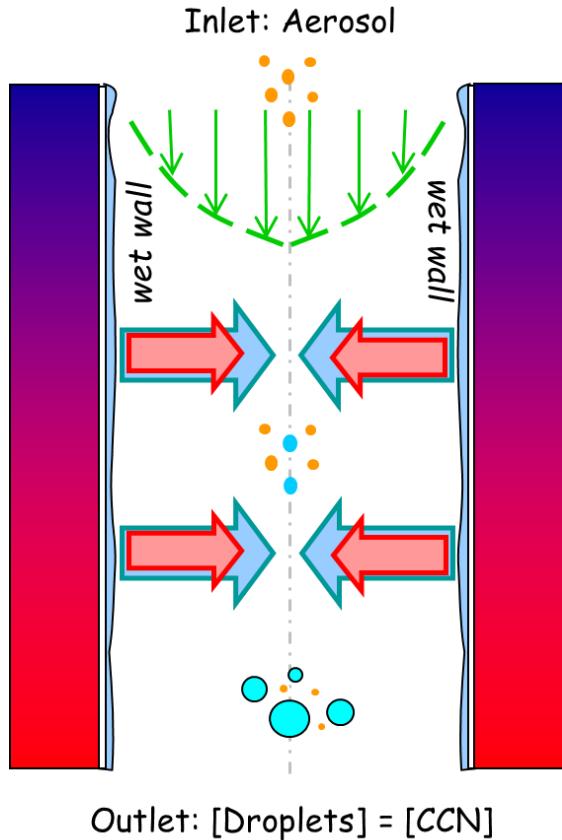
- They can facilitate cloud formation by acting as surfactants and adding solute (hygroscopicity)
- Oily films can form and delay cloud growth kinetics

## In-situ data to study the aerosol-CCN link:

Usage of CCN activity measurements to “constrain” the above “chemical effects” on cloud droplet formation.

# Continuous-Flow Streamwise Thermal Gradient Chamber

## CFSTGC... aka “DMT CCN Counter”



- Metal cylinder with wetted walls
- Streamwise Temperature Gradient
- Water diffuses faster than heat
- Supersaturation,  $S$ , generated at the centerline =  $f$  (Flowrate, Pressure, and Temp. Gradient)

Roberts and Nenes (2005), US Patent 7,656,510  
Lance et al., (2006), Lathem and Nenes (2011),  
Raatikainen et al. (2012)

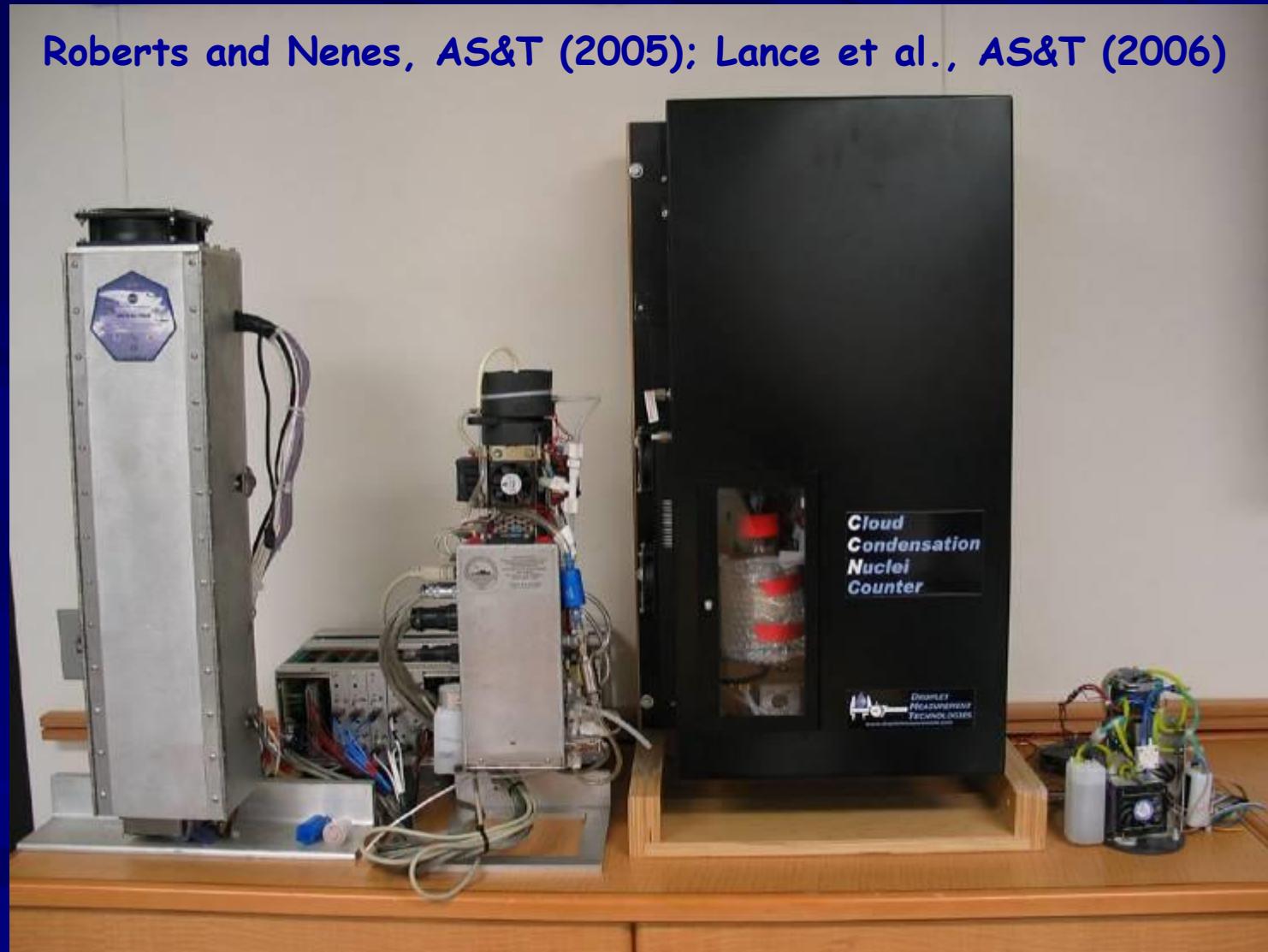
Interesting story on how all this happened...

# Development phases of cloud chamber

scale = 1 m



Roberts and Nenes, AS&T (2005); Lance et al., AS&T (2006)



1<sup>st</sup> version

2<sup>nd</sup> version

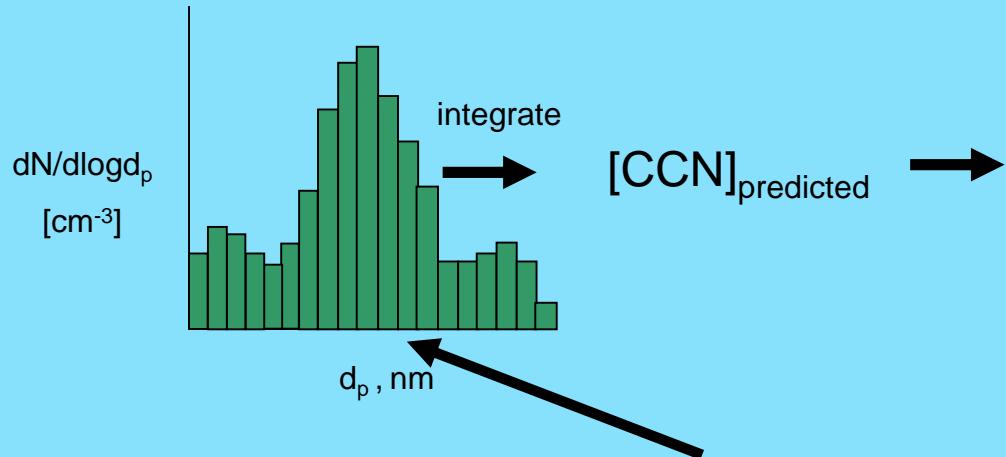
Commercial ver.

Mini-instruments

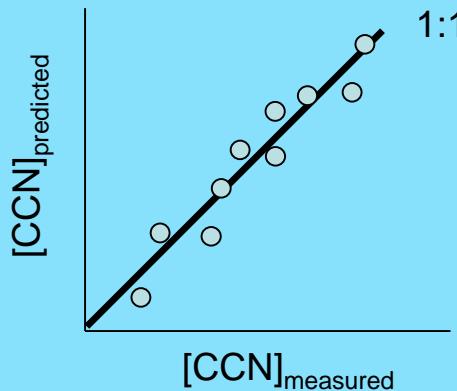
# Testing CCN activation theory: CCN “Closure” studies

Compare measurements of CCN to predictions using theory and a simple description of molar volume for organics

Aerosol Size Distribution



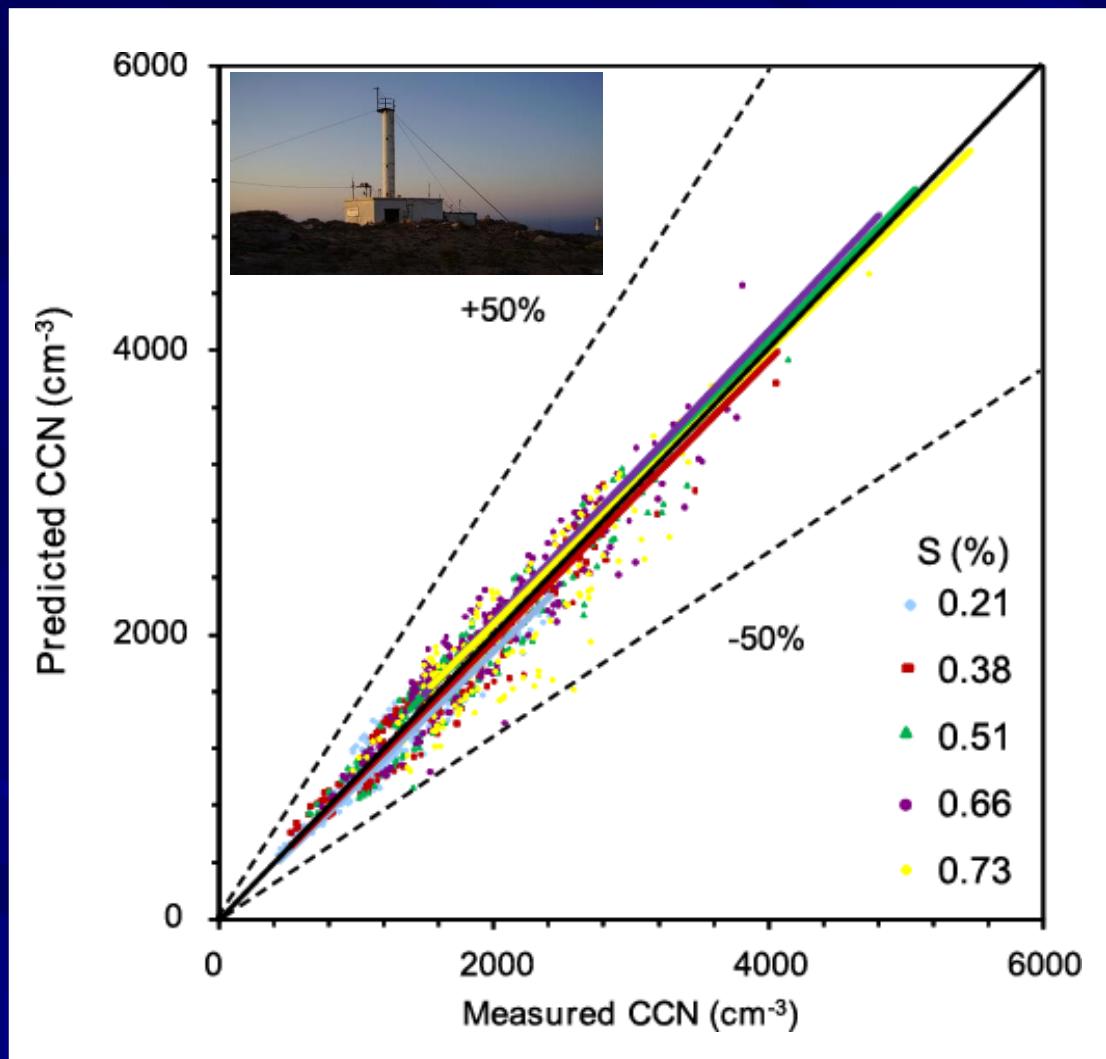
CCN Closure



Use theory to determine the **size** of particles above which they can act as CCN based on instrument supersaturation.

This size then can determine the “bulk” composition of particles using the concept of *hygroscopicity*.

# Example: Finokalia Aerosol Measurement Campaigns (Crete, Greece)



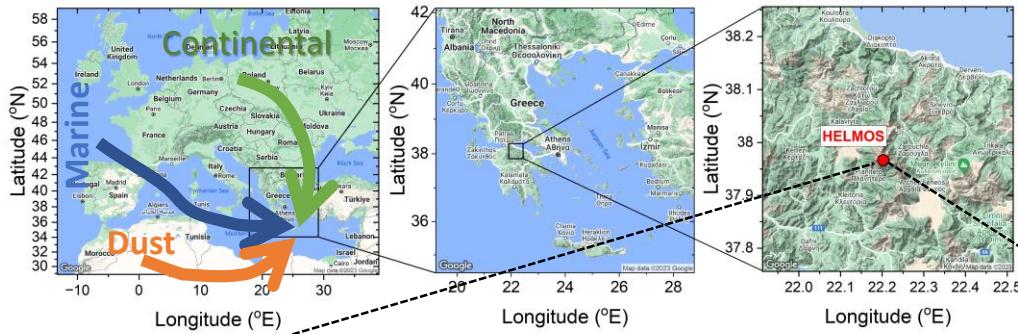
2% overprediction  
(on average).

CCN/Droplet  
prediction theory  
really works.

Simple treatment  
of organic aerosol  
hygroscopicity  
really works too.

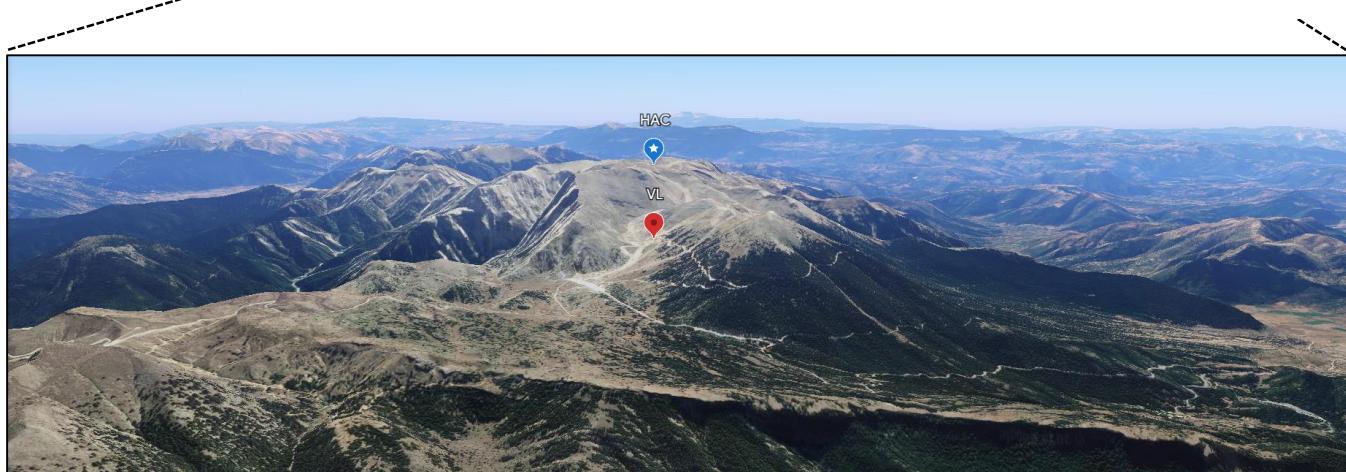
(Bougiatioti et al., ACP, 2009; 2012  
and many other studies)

# Mt. Helmos: Where mythology, aerosols & clouds meet



## Some facts:

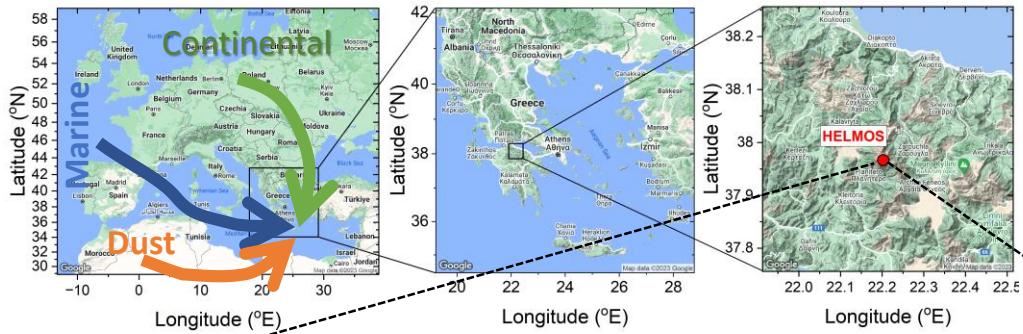
- River Styx near (HAC)<sup>2</sup> – back entrance to Hades
- Hermes was born there and had a home there too.
- Achilles was bathed by the fairy Thetis there and... almost became immortal.



## Modern history:

- Played significant role in Greek War of Independence, serving as hideout for guerrilla fighters

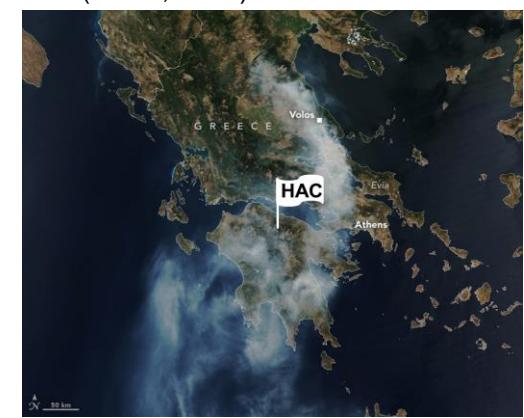
# Mt. Helmos: Where mythology, aerosols & clouds meet



Dust transportation over  
Peloponnese  
(Feb 1, 2015)

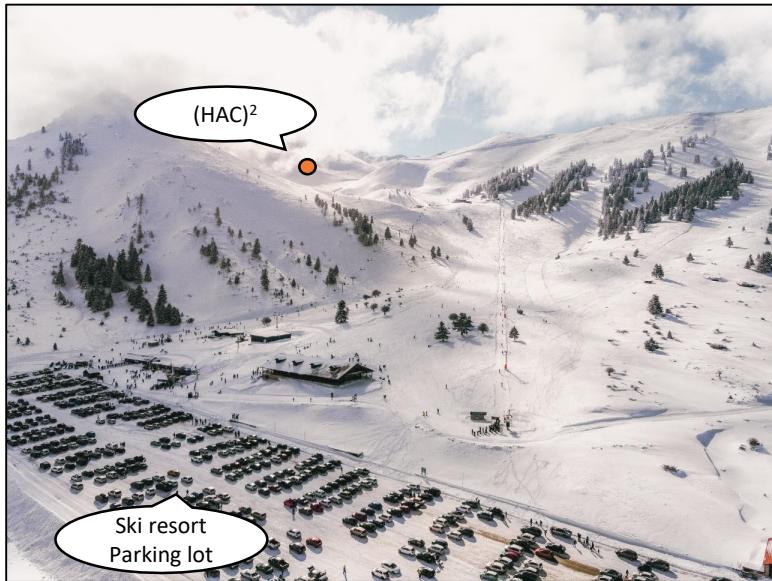


Images: Google Earth; NASA (<https://earthobservatory.nasa.gov>)



Wildfire smoke over Peloponnese  
(Aug 8, 2021)

# CHOPIN: Cleancloud Helmos OrograPhic site experimeNt



## Some facts:

- *Period:* Oct.2024 – Jan 2025 (and later for some instruments)
- *Coordination:* NCSR Demokritos (K.Eleftheriadis) with scientific coordination from FORTH (A.Nenes) and EPFL/NTUA (A.Papayannis)



AARHUS  
UNIVERSITY



FINNISH METEOROLOGICAL  
INSTITUTE



KIT  
Karlsruhe Institut für Technologie



°CICERO



THE CYPRUS  
INSTITUTE  
RESEARCH TECHNOLOGY INNOVATION



## Key goals of the experiment:

1. Understand processes and drivers of cloud formation (build upon CALISHTO).
2. Evaluate, improve and develop remote sensing algorithms for aerosols and clouds

# CHOPIN instruments (not complete list... a lot more in place)



Drones

We invite the whole community to participate!

# Moments from CHOPIN



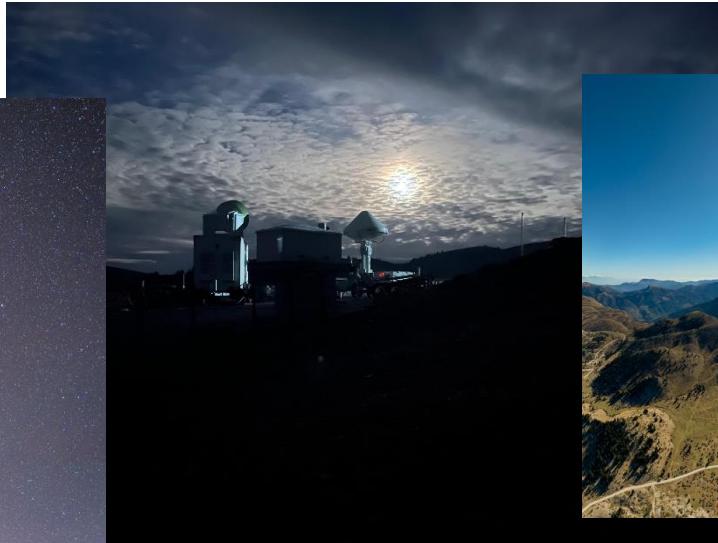
# Moments from CHOPIN



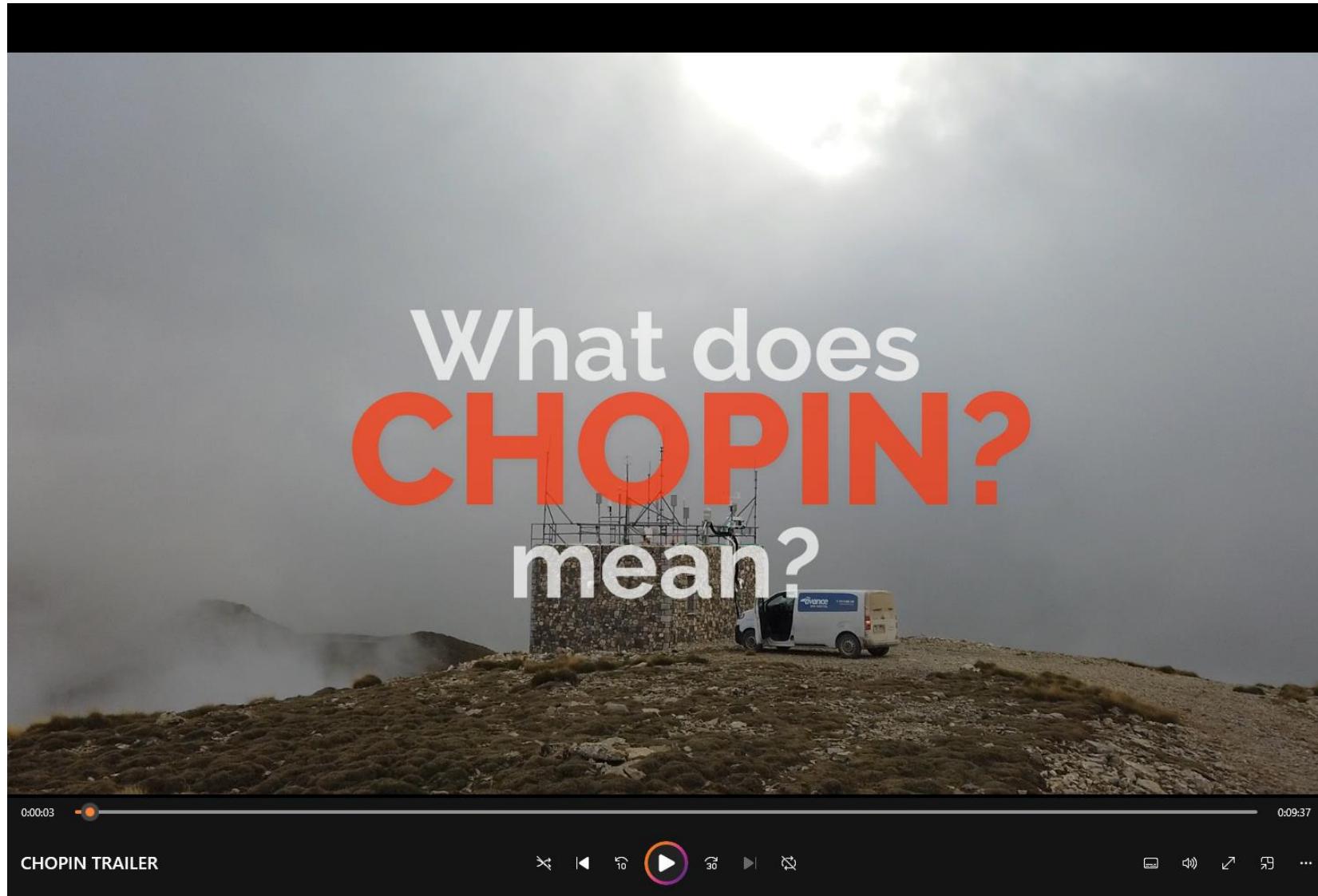
# Moments from CHOPIN



# Moments from CHOPIN



Let's watch CHOPIN science and researchers in action!



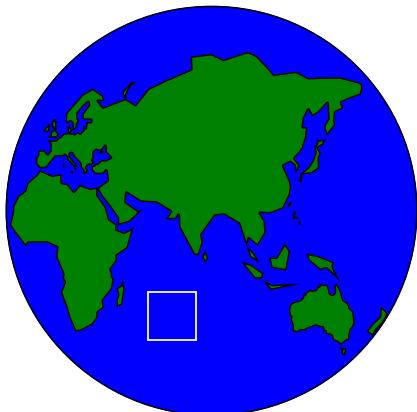
Lets take a break to stretch our legs ... but keep the nice visuals going on!



<https://mediaspace.epfl.ch/channel/CleanCloud/70678>

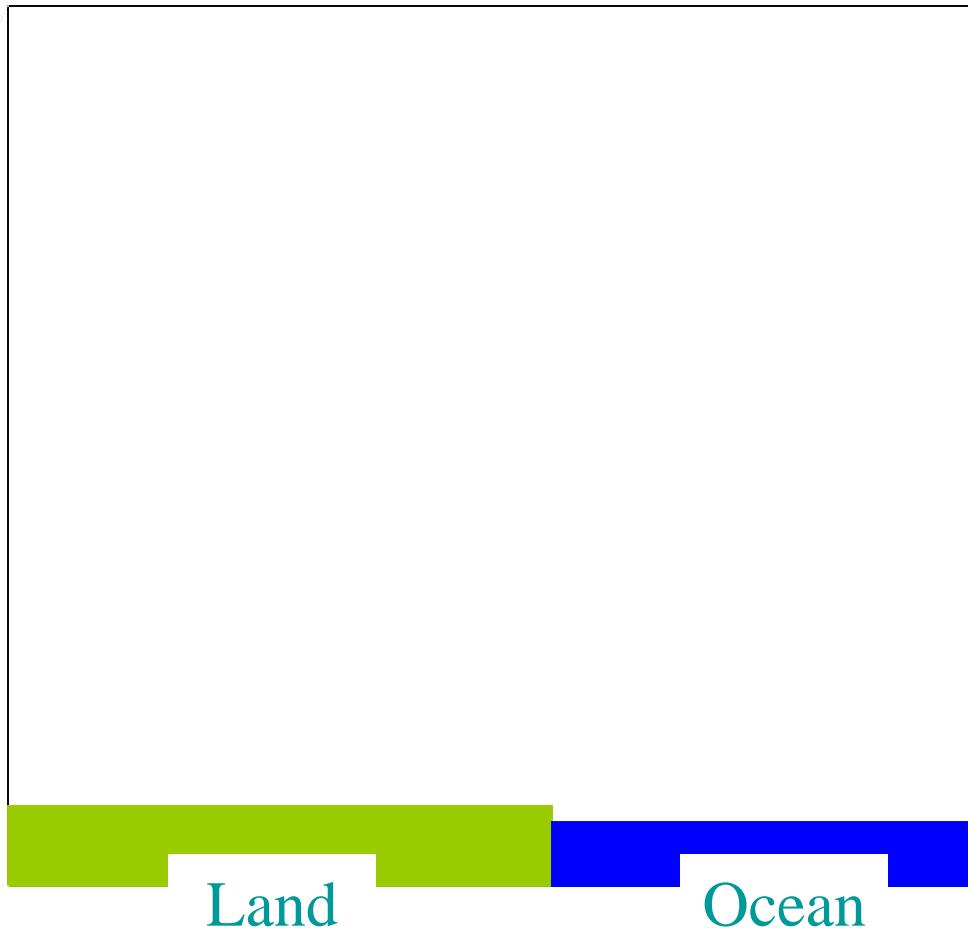
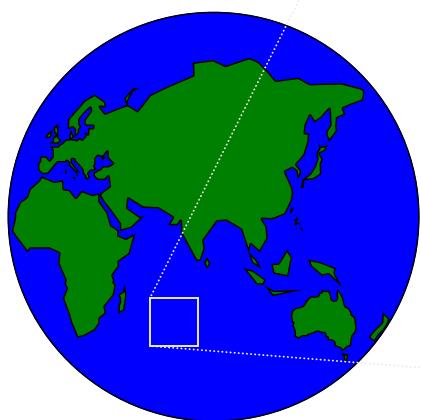
# Global Climate Models: Tools of understanding

- Divide the Earth into small parts ("grid cells").
- Write equations describing
  - Conservation of Energy, Water, chemical constituents
  - Evolution of aerosol size distribution
  - Interactions of land/ocean with atmosphere
  - ... etc.
- Prescribe initial conditions (e.g., climatology).
- Integrate the equations (numerically) over time.



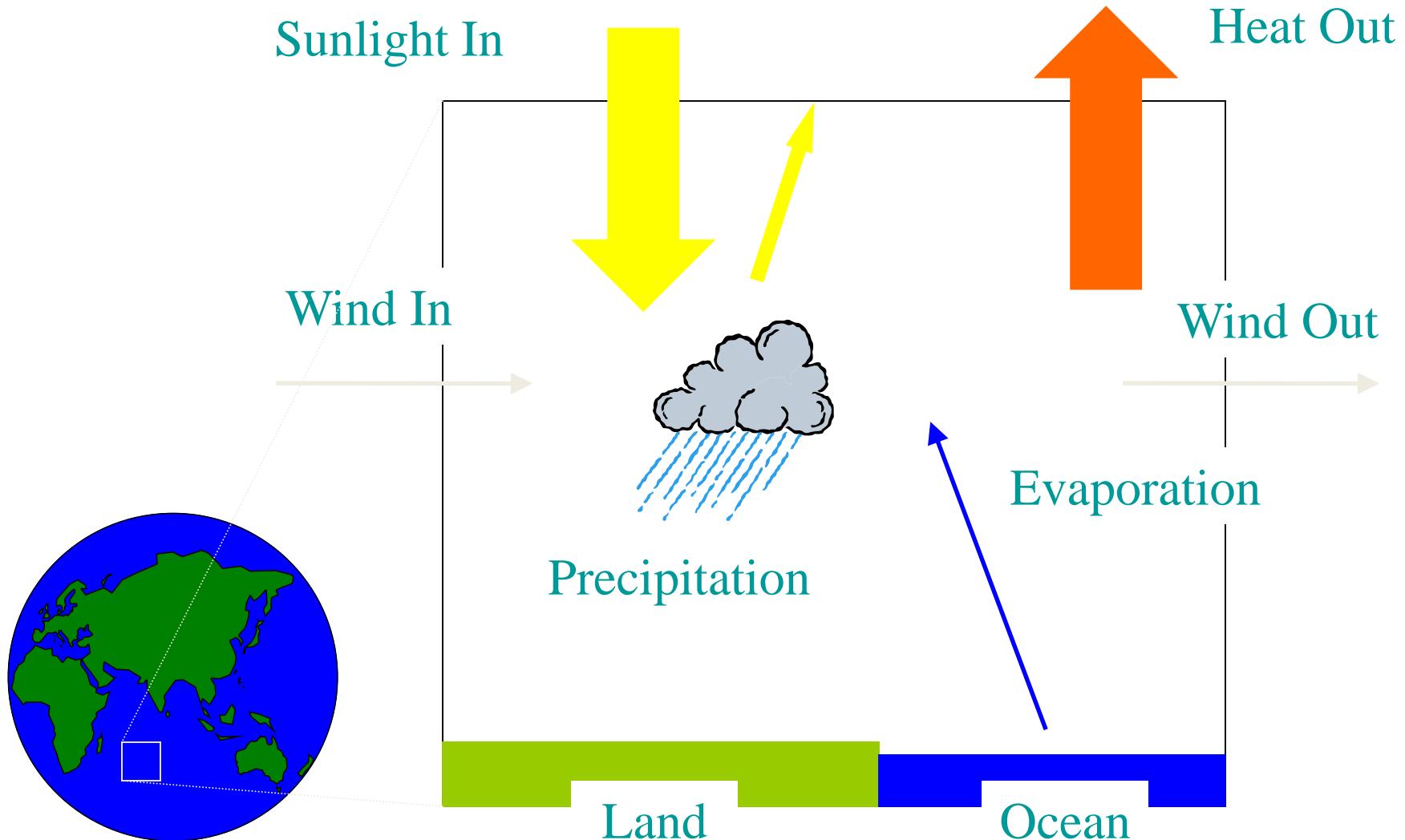
# How Computer Climate Models Work

Example: conservation of energy in the atmosphere



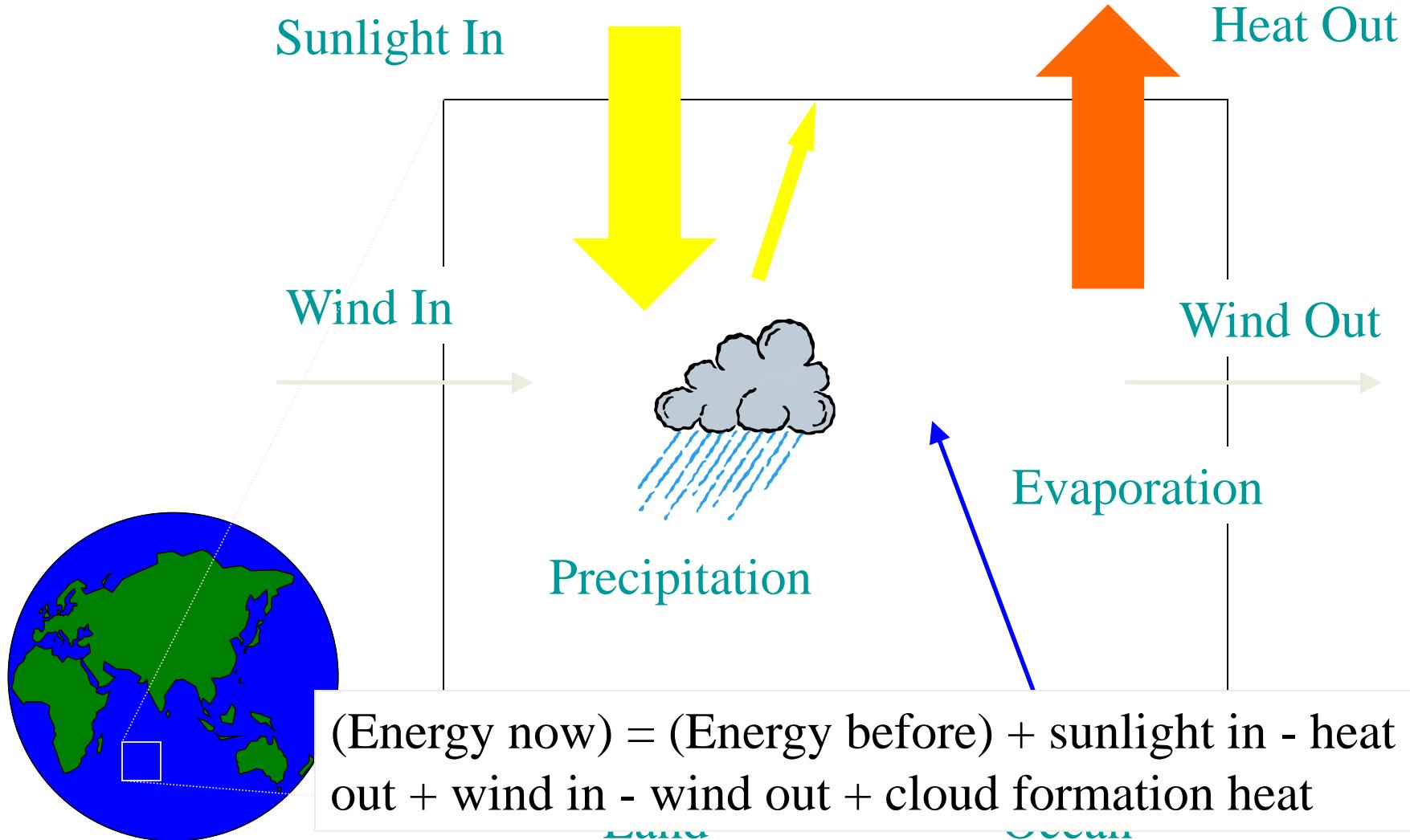
# How Computer Climate Models Work

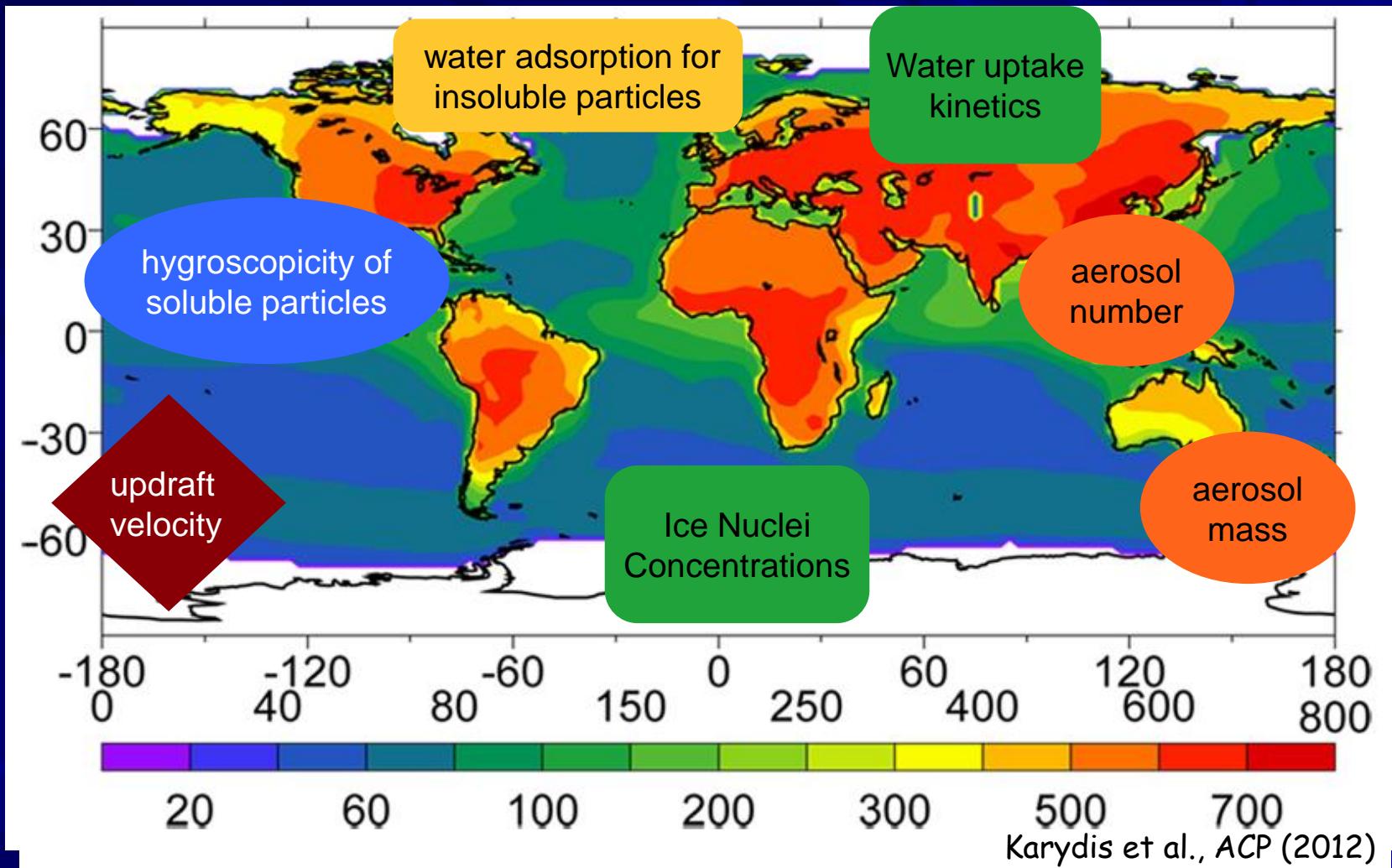
Example: conservation of energy in the atmosphere



# How Computer Climate Models Work

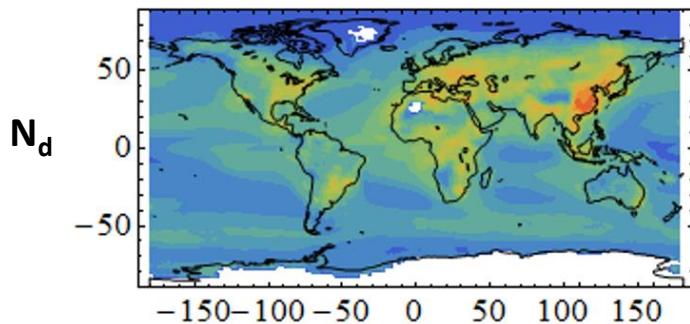
Example: conservation of energy in the atmosphere





How important is each parameter? What causes most variability? What needs to be best understood?

# Our group contributes to climate models

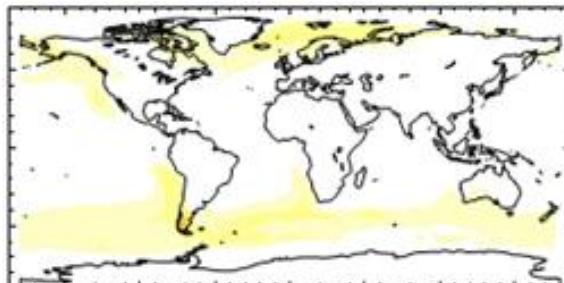
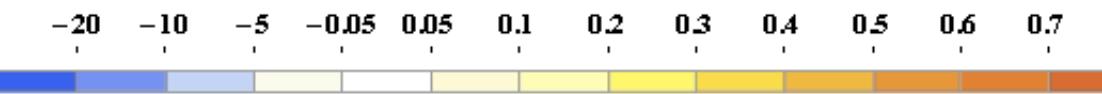


Present Day Emissions (2000), NCAR 5.1

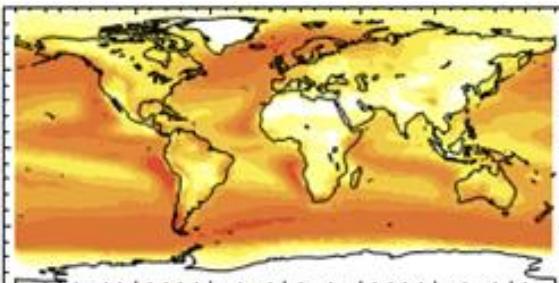
$\text{cm}^{-3}$

15 45 75 100 150 200 250 300 350 400 500 600 700

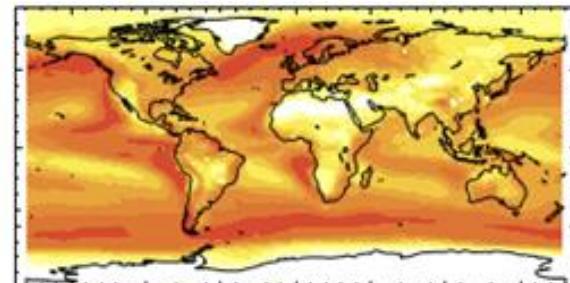
Morales and Nenes, ACP (2014)



$\frac{\partial N_d}{\partial n_{ai}}$   
Ultrafine particles

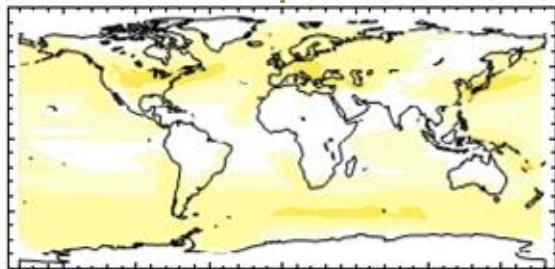


Fine particles

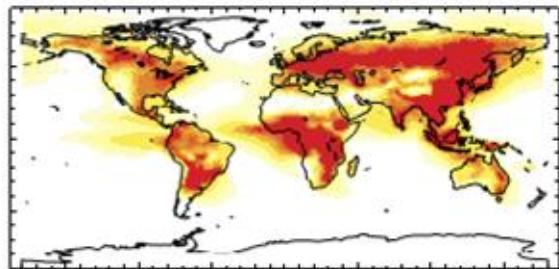


Coarse particles

$$\frac{\partial N_d}{\partial n_{ai}}$$



$$\frac{\partial N_d}{\partial \kappa_i}$$



-100 -80 -60 -40 -20 -10 -5 5 10 20 40 60 80 100 [  $\text{cm}^{-3}$  ]

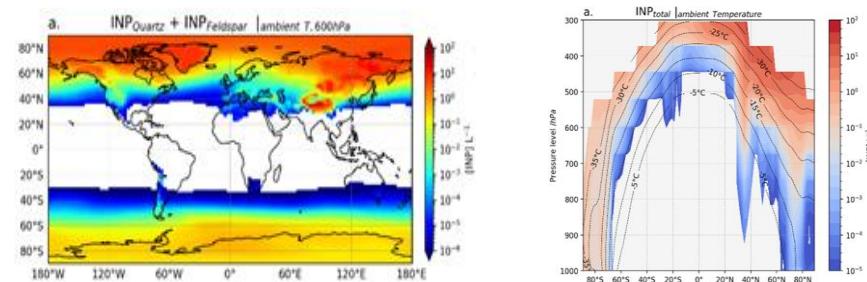
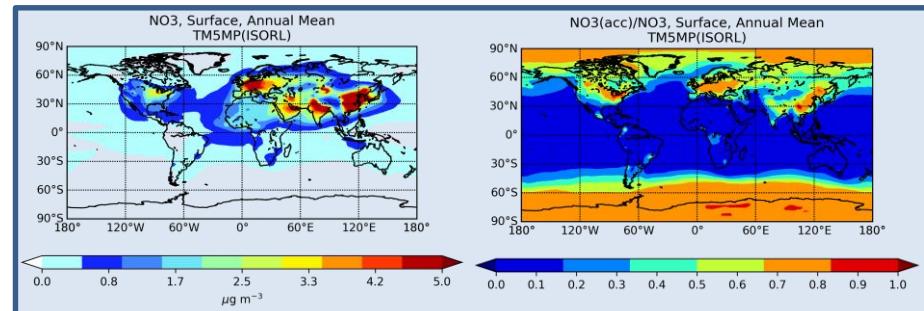
15 45 75 100 150 200 250 300 350 400 500 600 700

# Climate Modeling: EC-Earth Consortium Member and contributor to many global and regional climate models

CSTACC contributions to EC-Earth include (blue means contributions to future IPCC runs)

- aerosol representation :
  - coarse mode nitrate and the thermodynamic module isorropia lite
  - Brown carbon (BrC), the absorbing component of organic aerosol
  - K-feldspar & quartz dust minerals with ice nucleating properties (INP)
  - Marine organics and terrestrial bioaerosols with ice nucleating properties
- liquid, ice and mixed-phase clouds (their representation and interaction with aerosols)
- nutrient representation and their atmospheric deposition

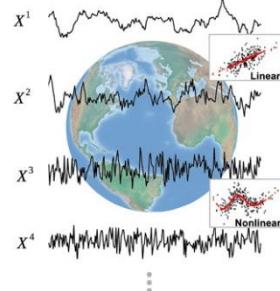
CSTACC modules for aerosol (ISORROPIA) and clouds widely used in other global climate models (NorESM, CESM, GFDL, NASA GEOS, ECHAM-HAM, HadGCM, ICON) & regional climate models (WRF/Polar-WRF)



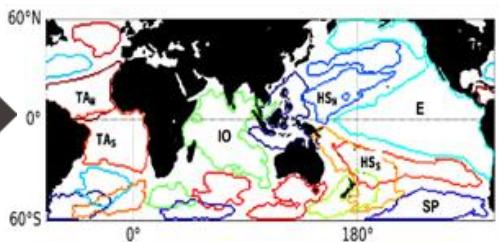
Chatziparaschos et al., to be submitted to ACPD, 2022

# Data mining & Knowledge Discovery to Constrain Climate Sensitivity

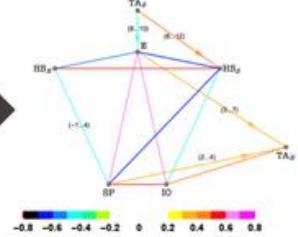
Take climate model output



Dimensionality reduction ( $\delta$ -MAPS)



Network inference ( $\delta$ -MAPS)

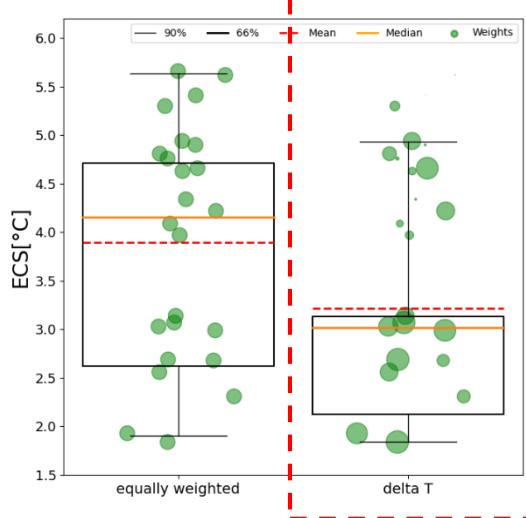


Observations and reanalysis

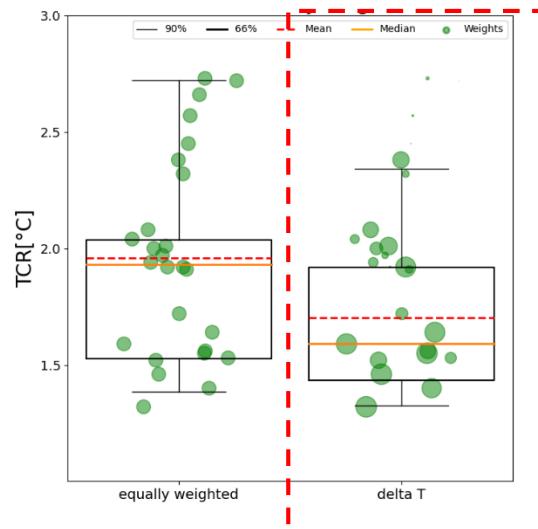
1. HadISST
2. COBEv2



Equilibrium Climate Sensitivity



Transient Climate Sensitivity



Application of Network Analysis ( $\delta$ -MAPS) to climate model simulations reduces the uncertainty range of future projections.

We developed a “robust” emergent constraint the climate community needs.

Ricard et al., in prep

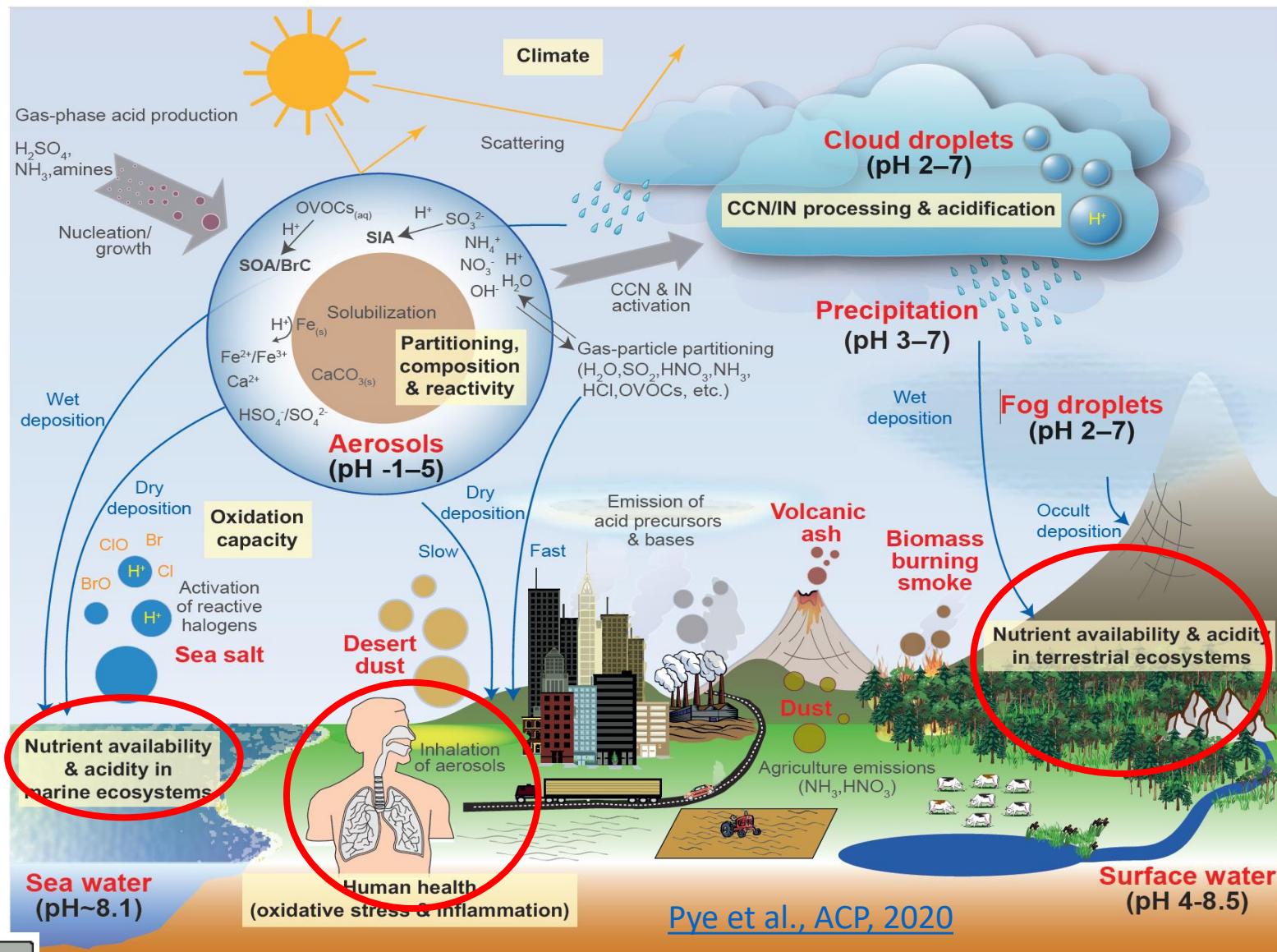




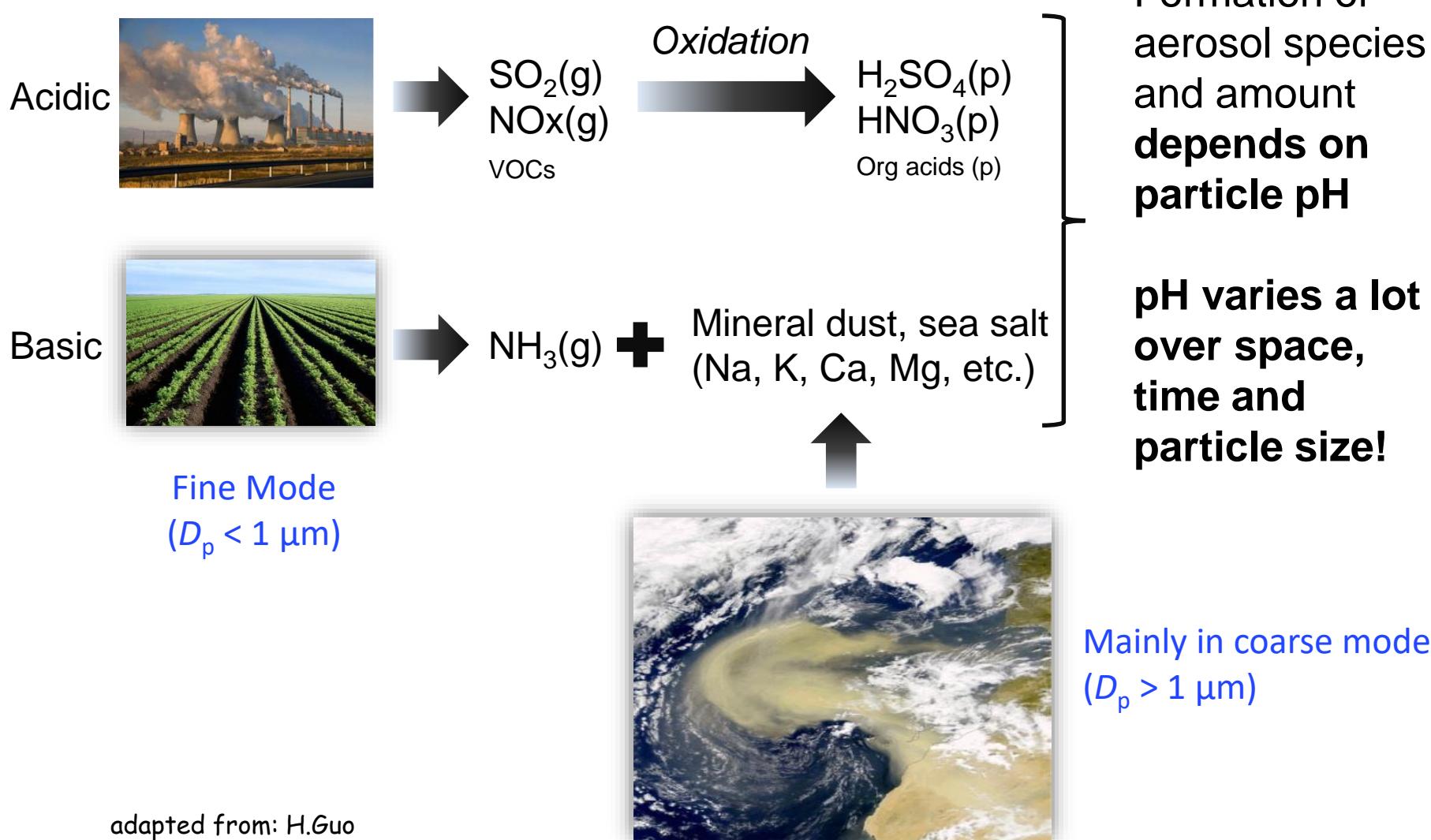
# **Aerosols, Acidity and Impacts**

*An exploding area of research*

# The Acidity of Atmospheric Particles



# Emissions & partitioning affect aerosol acidity



adapted from: H.Guo

# Measuring aerosol pH: The problem

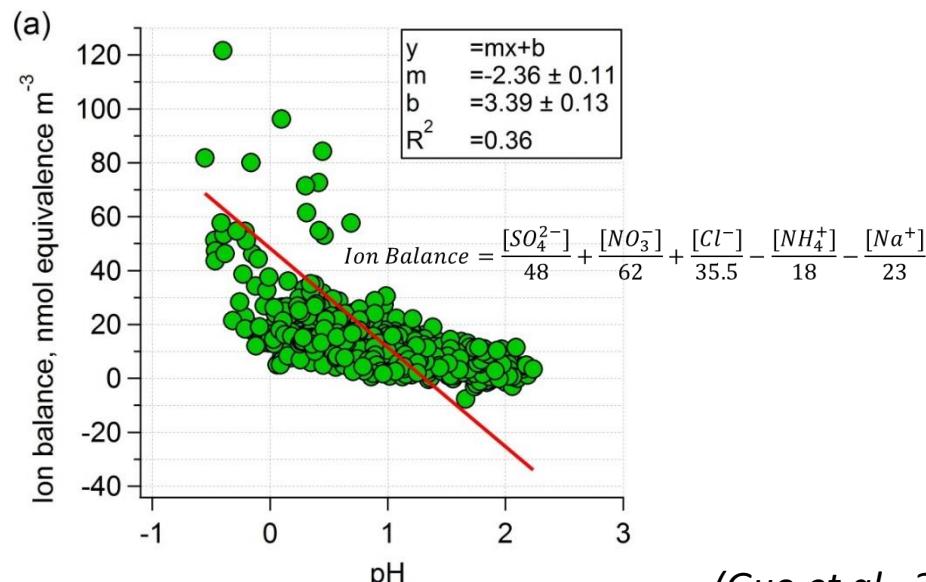
- No direct measurement of pH is available for single particles *in-situ*.
- Emerging offline methods – but a long way to go before they are widely used.
- “pH proxies” (ion balance, molar ratios), **do not strongly correlate with pH**

$$pH = -\log_{10}[H^+] = -\log_{10} \frac{1000H_{air}^+}{LWC}$$

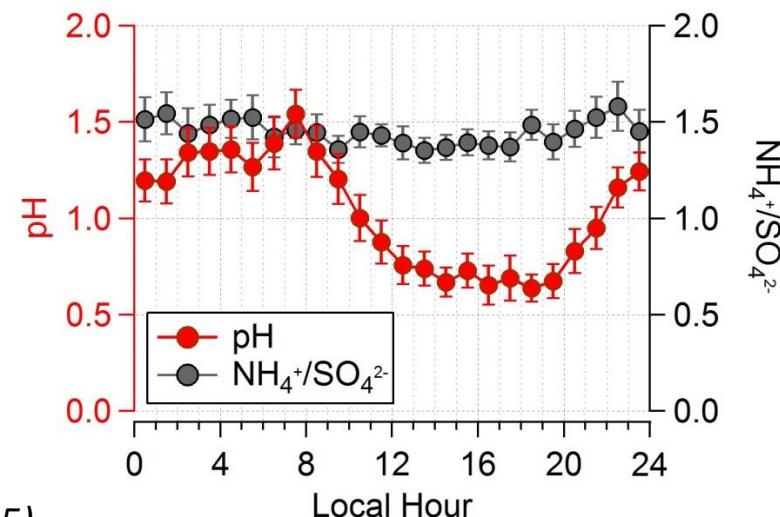
$H_{air}^+$ , LWC units:  $\mu\text{g m}^{-3}$  air

- Current gold standard: Measurements + Thermodynamic modeling

## *Ion balance:*



## *NH<sub>4</sub><sup>+</sup>/SO<sub>4</sub><sup>2-</sup> Molar ratio:*



(Guo et al., 2015)

# Determining aerosol pH: The “heart” of it

## ISORROPIA-II calculates:

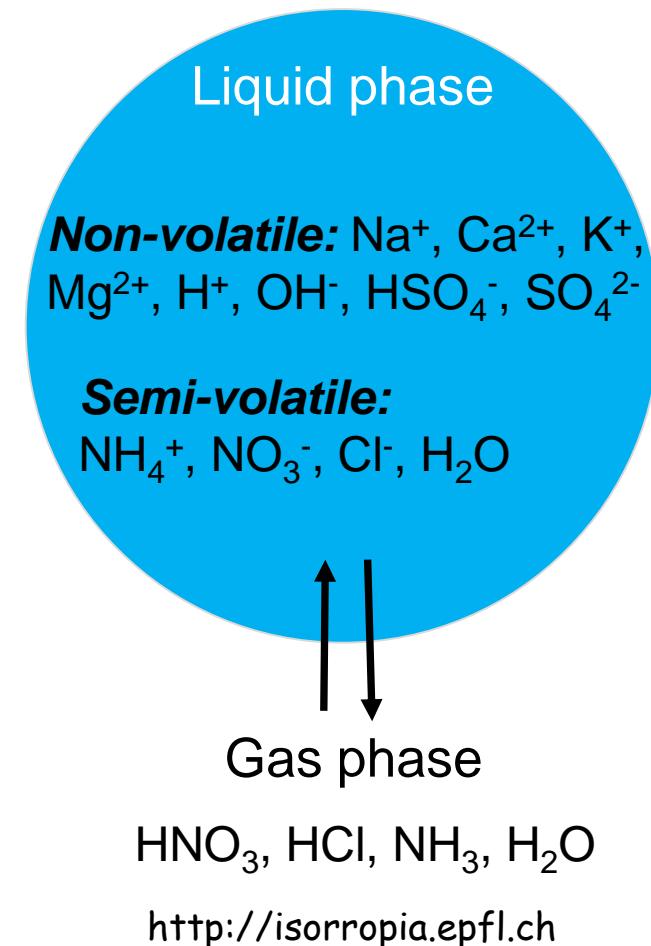
the composition and phase state of an  $\text{NH}_4^+$ -  
 $\text{SO}_4^{2-}$ - $\text{NO}_3^-$ - $\text{Cl}^-$ - $\text{Na}^+$ - $\text{Ca}^{2+}$ - $\text{K}^+$ - $\text{Mg}^{2+}$ -water  
inorganic aerosol in equilibrium with gases

## Assumptions:

- “metastable” aerosol & no phase separation  
(a single aqueous phase)
- $\text{PM}_{2.5}$  in equilibrium with gas

## Forward mode:

calculates equilibrium partitioning given total  
concentration of species (gas + particle)



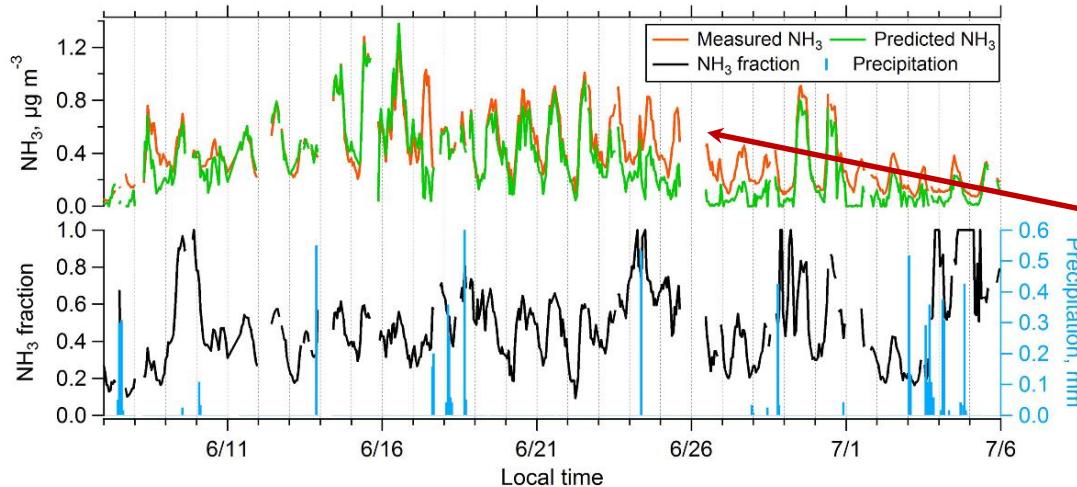
{ Input: Total nitrate ( $\text{HNO}_3 + \text{NO}_3^-$ ) → Output:  $\text{HNO}_3$ ,  $\text{NO}_3^-$

Input: Total ammonium ( $\text{NH}_3 + \text{NH}_4^+$ ) → Output:  $\text{NH}_3$ ,  $\text{NH}_4^+$

<http://isorropia.epfl.ch>

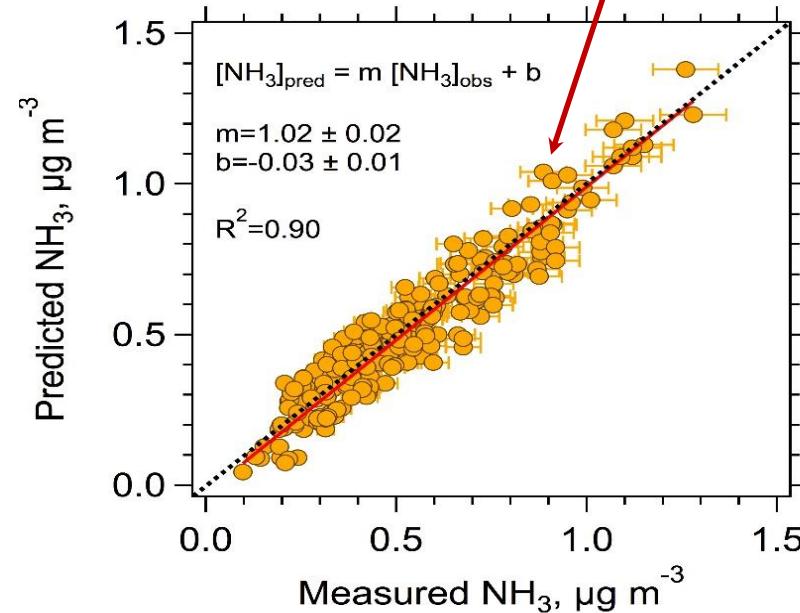
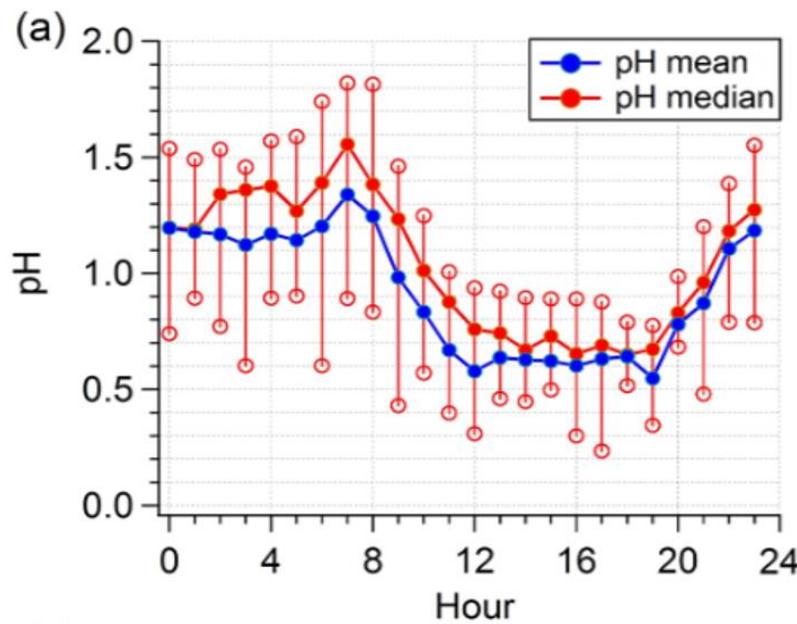
# pH constrained by $\text{NH}_3\text{-NH}_4^+$ partitioning

SOAS: (Southern Oxidant Aerosol Study) 6/7, 2013 Centreville, AL (CTR)



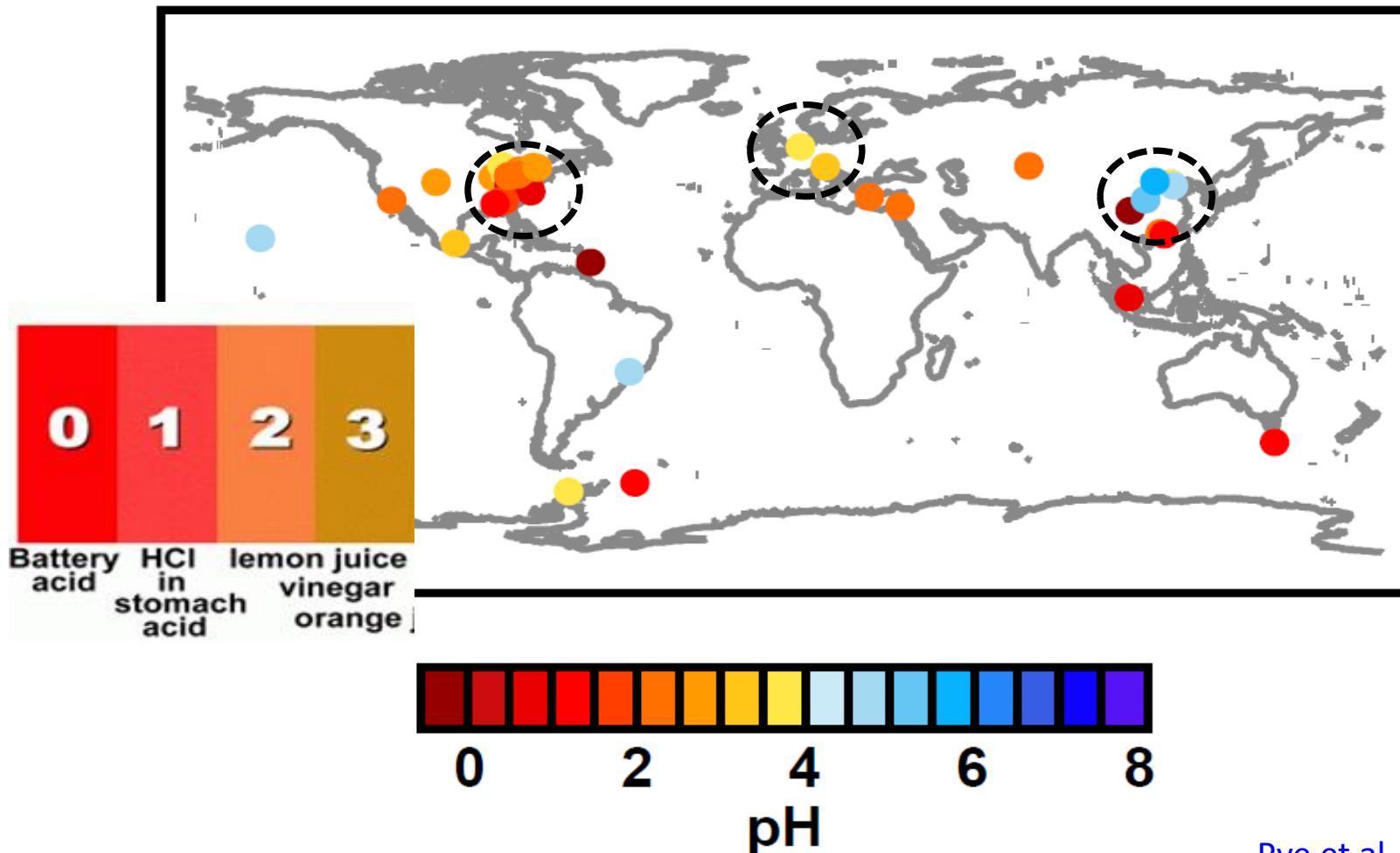
Guo et al., ACP, 2015.

Comparison of  
predicted vs.  
observed gas-  
phase  $\text{NH}_3$ .



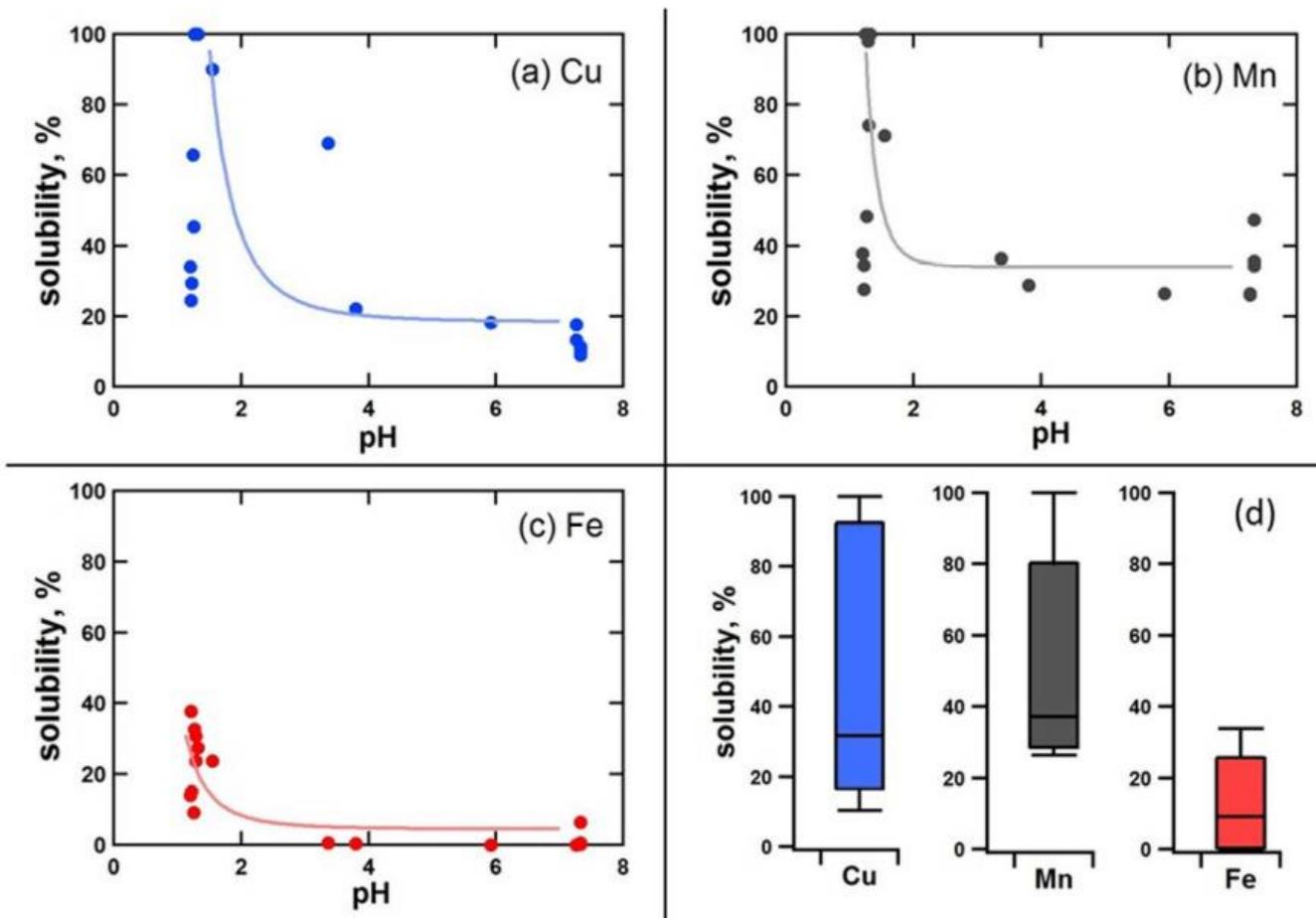
# Acidic aerosol is everywhere

## pH varies a lot



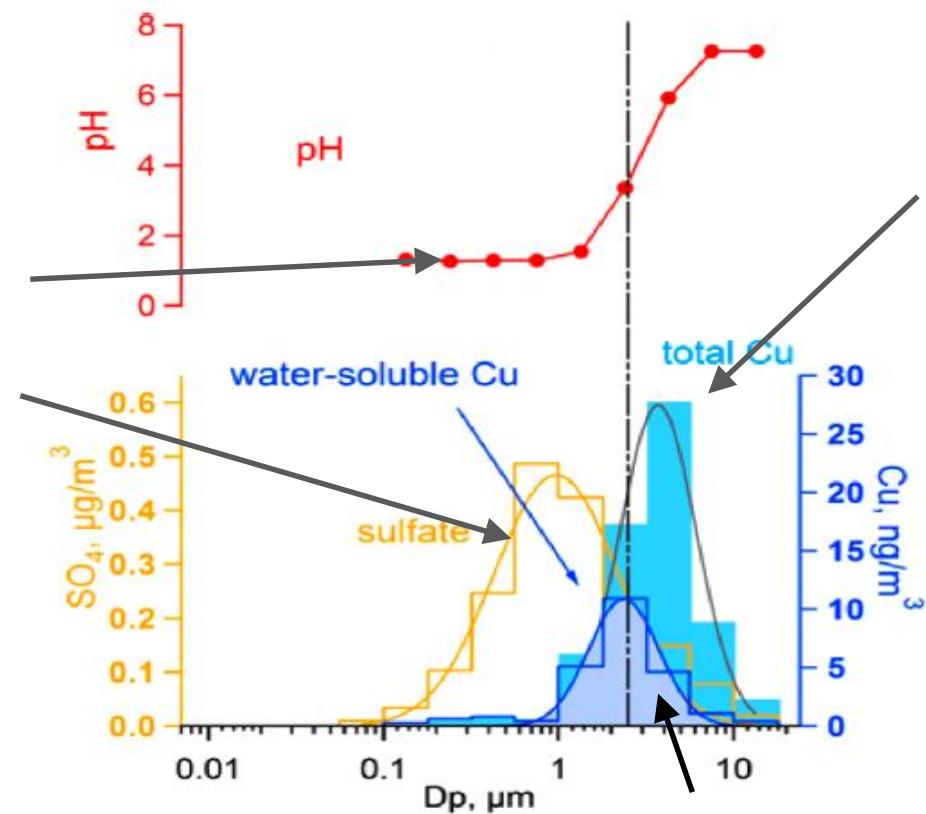
# Health impacts: Acidity dissolves metals

- pH is profound for determining PM2.5 levels.
- Important driver of toxicity – and can explain the association of sulfate, soluble metals etc. with adverse health outcomes.



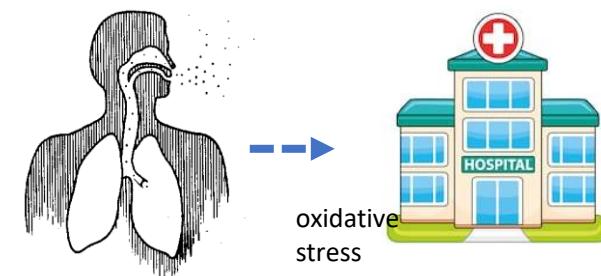
# Health impacts: Acidity dissolves metals

Acidic PM<sub>1</sub> due to presence of sulfate and few NVCs.



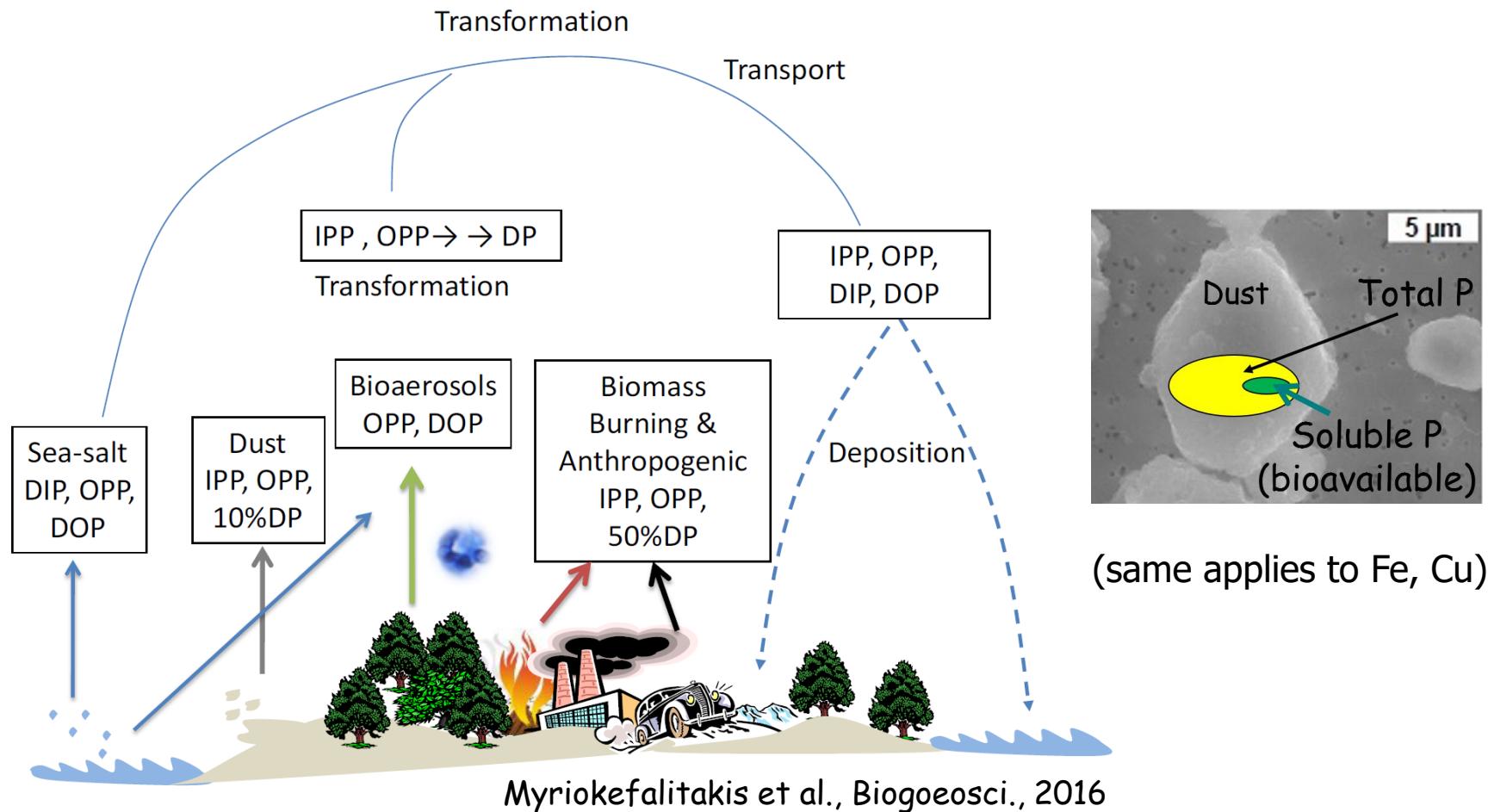
Fang et al. *ES&T* 2017  
Dallenbach et al., *Nature*, 2020

Emitted insoluble metals in coarse mode.



- Soluble metals appear where acidity is strong (pH low)
- Toxicity related to inhalation of soluble metals
- ***Mechanism explaining why PM 2.5 sulfate in the environment is associated with toxicity***

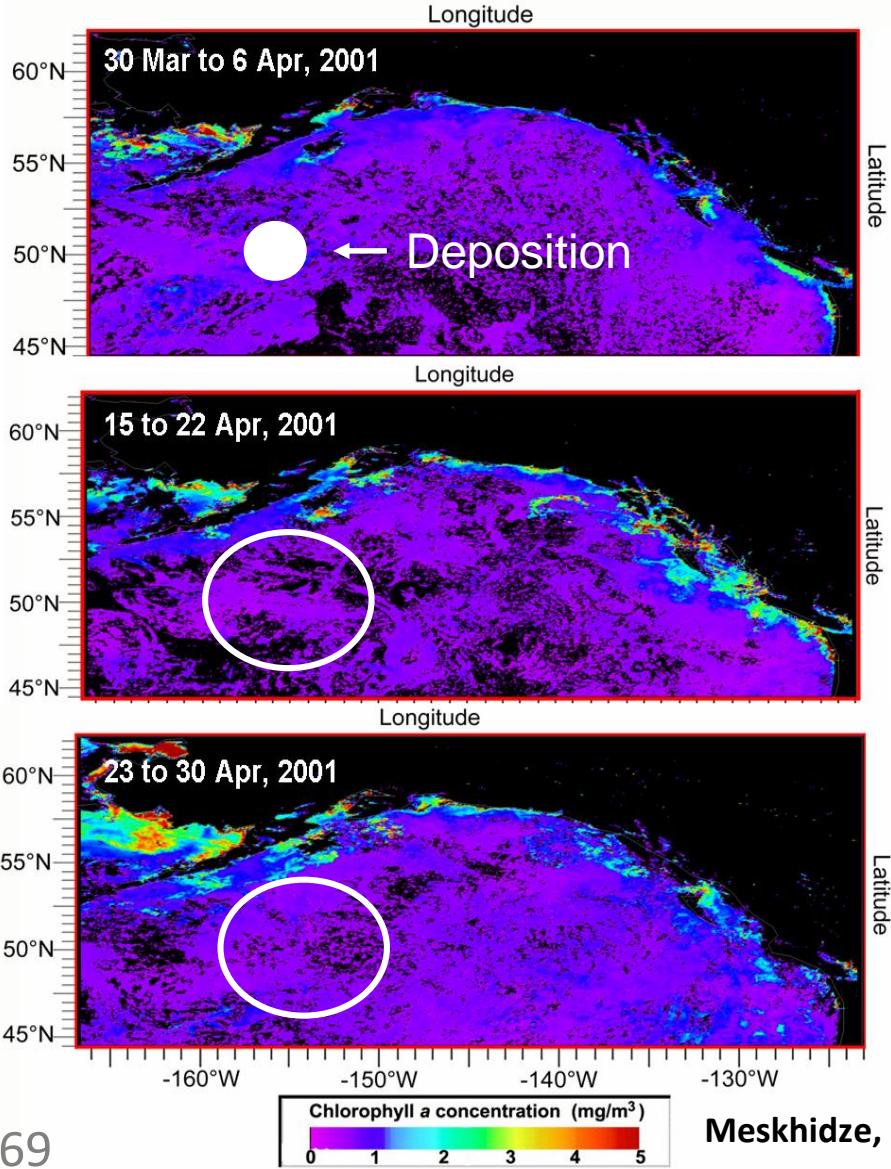
# Particle pH affects global nutrient cycles



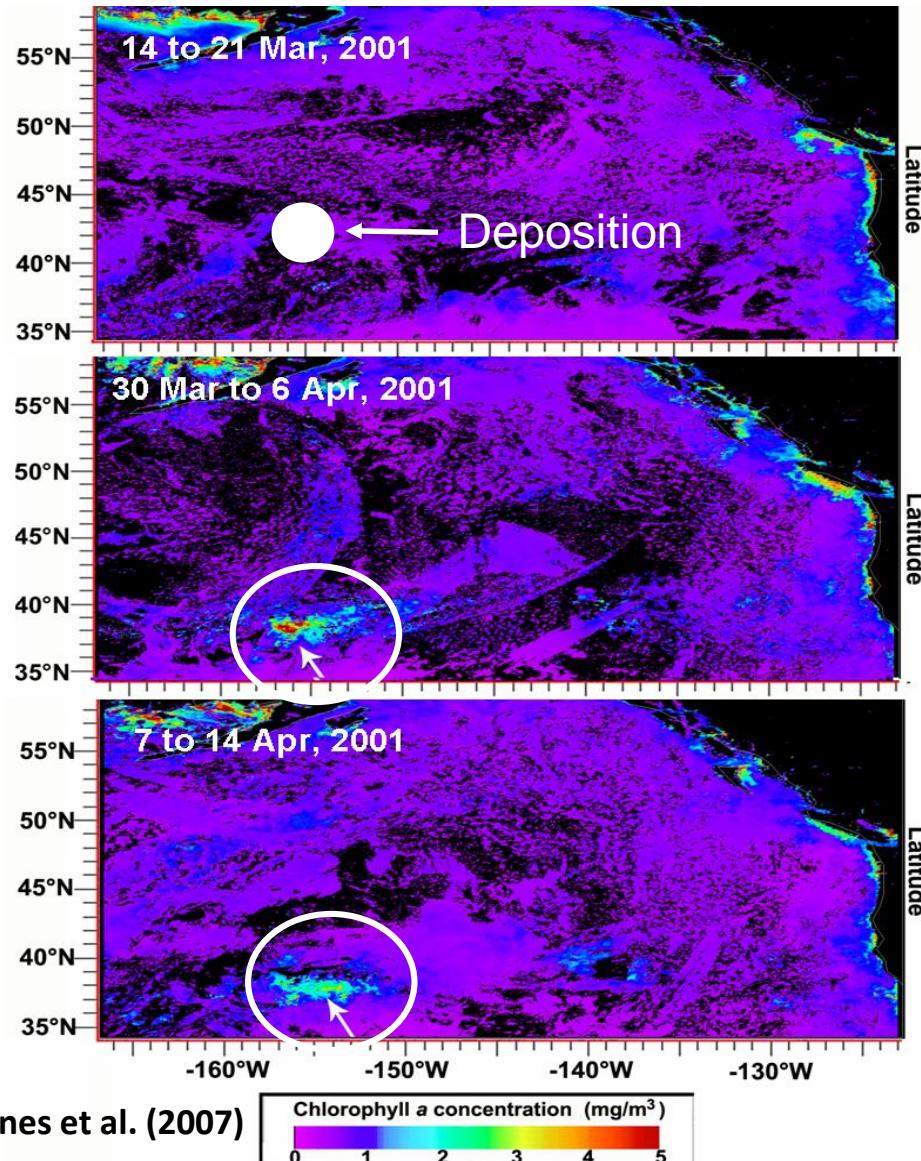
- Mineral dust is a prime source of P, Fe, Cu to the (offshore) ocean.
- Interaction of Dust with pollution affects their soluble (bioavailable) fraction.
- Aging largely occurs by acidification/dissolution of metal-containing minerals.

# Ecosystems respond to increased nutrient deposition

Dust deposition event & weak acidity

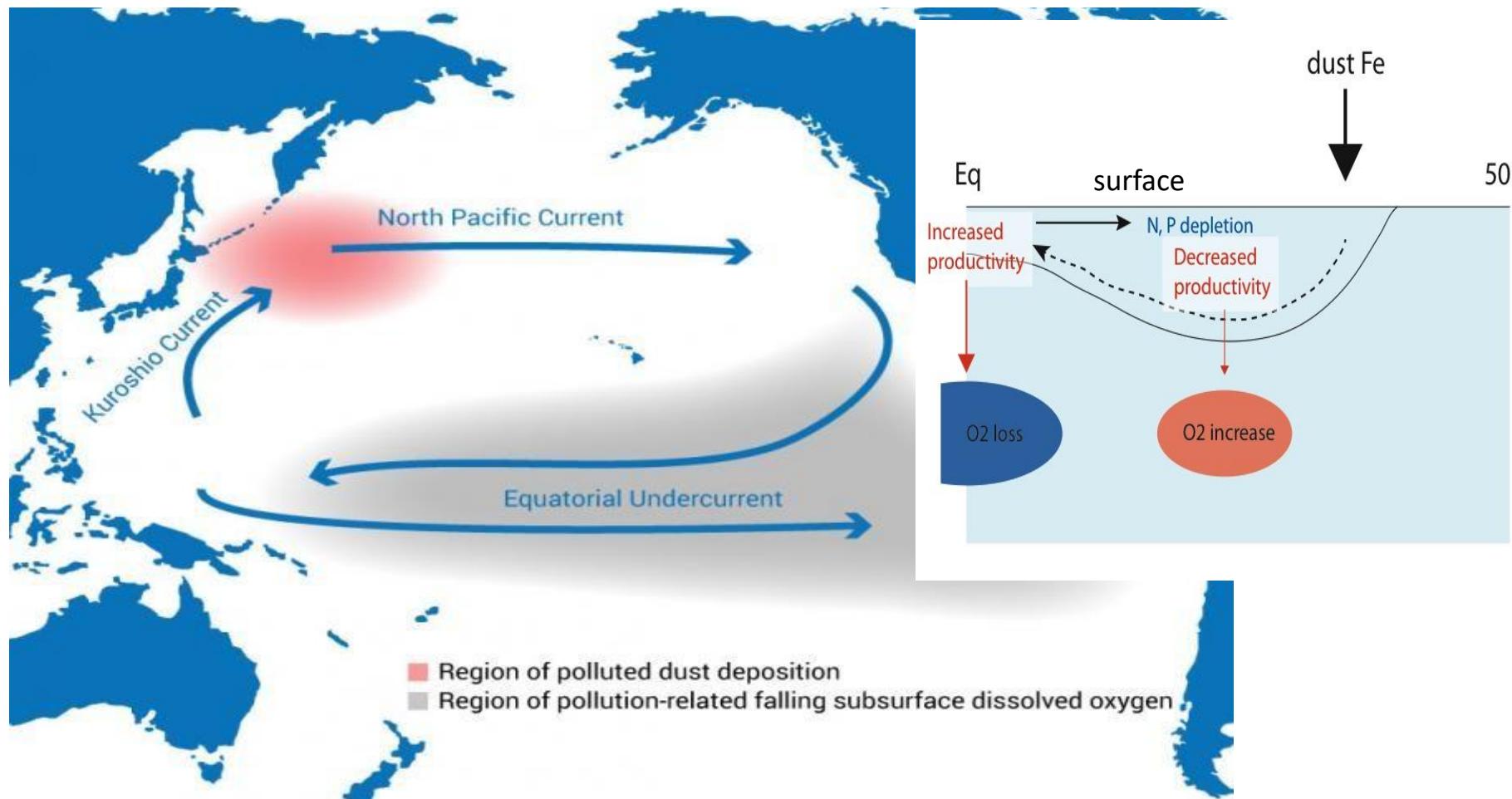


Dust deposition event & strong acidity



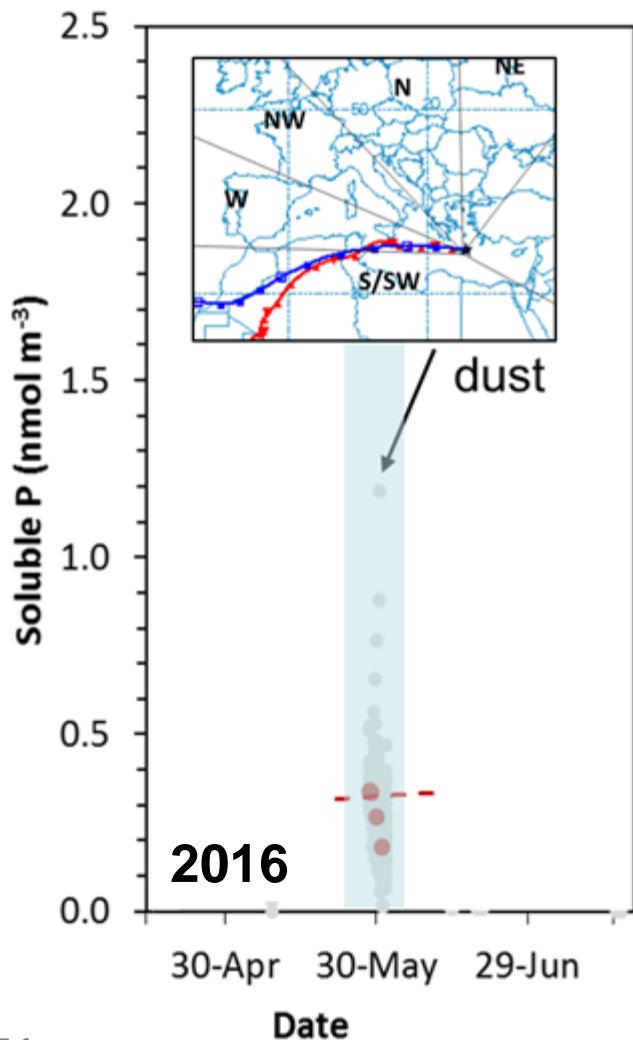
Meskhidze, Nenes et al. (2007)

Fe deposition can profoundly impacts productivity and ocean  $O_2$  levels – far from deposition region.



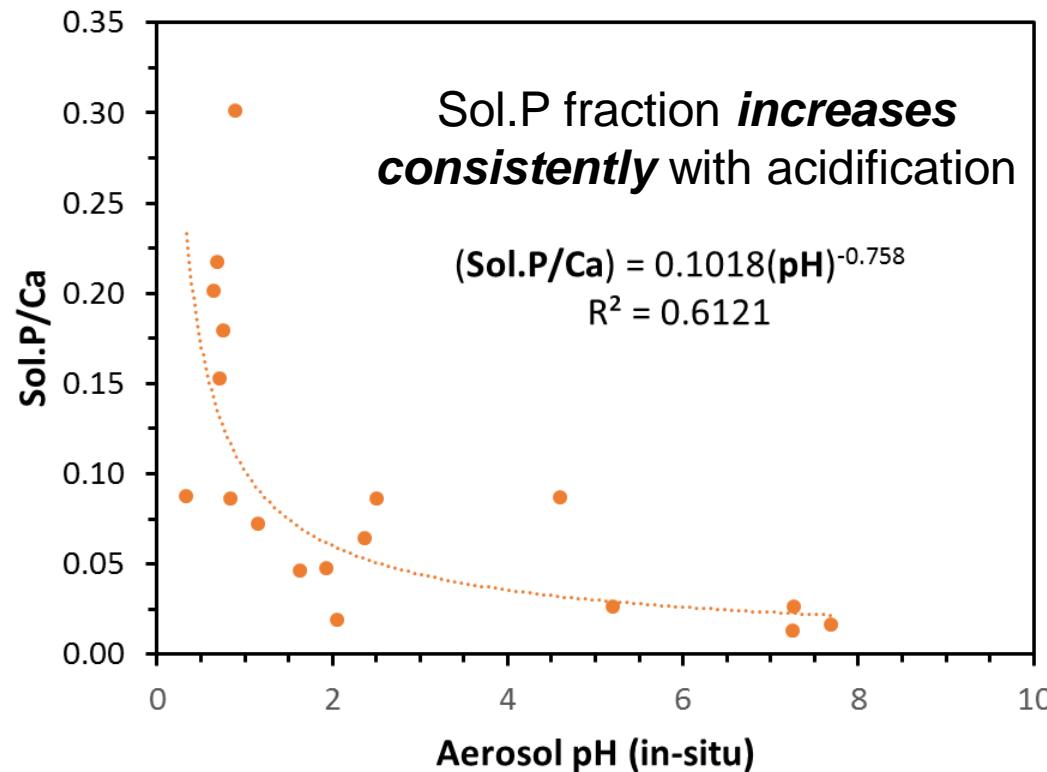
Acceleration of oxygen decline in the tropical Pacific over the past decades by aerosol pollutants

# Acidification solubilizes dust P: evidence from E.Mediterranean data.



## Focus on dust events:

- ISORROPIA (Fountoukis and Nenes, 2007) to obtain aerosol pH for in-situ (fine mode) aerosol
- Express P/Ca vs.aerosol pH (as Nenes et al. 2011)

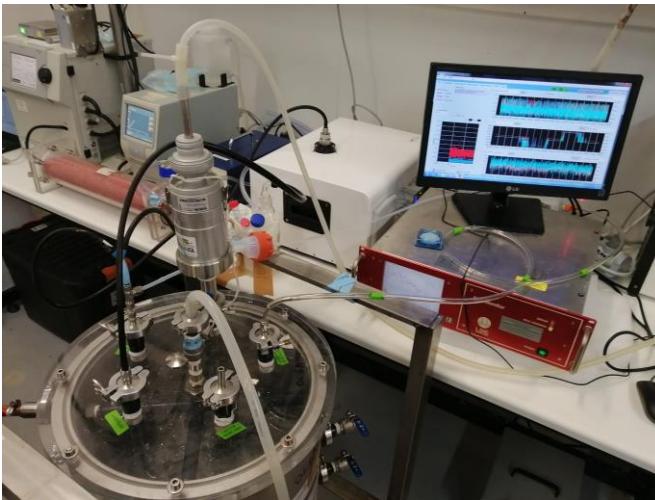


# Fertilization of E.Mediterranean ecosystems from dust (AQUACOSM-plus).

- May-June 2022
- Understand impacts of pollution/bioaerosol/dust deposition on marine ecosystems.
- Collection of aerosol from different sources, and deposit them to seawater collected from the E.Mediterranean

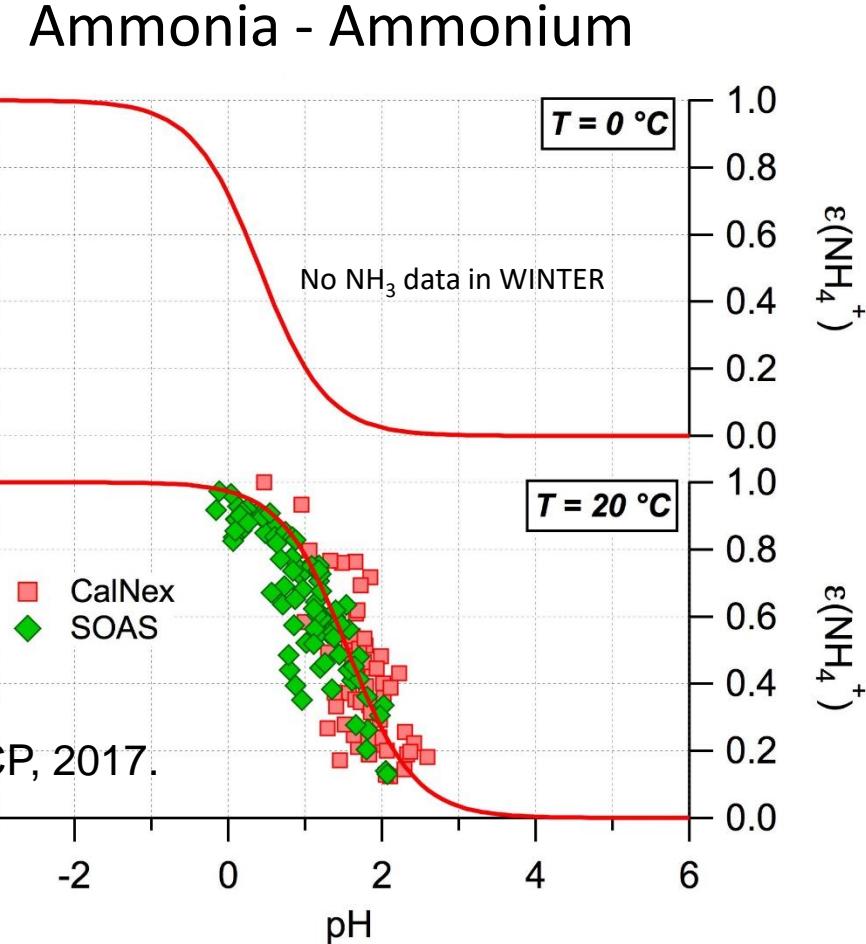
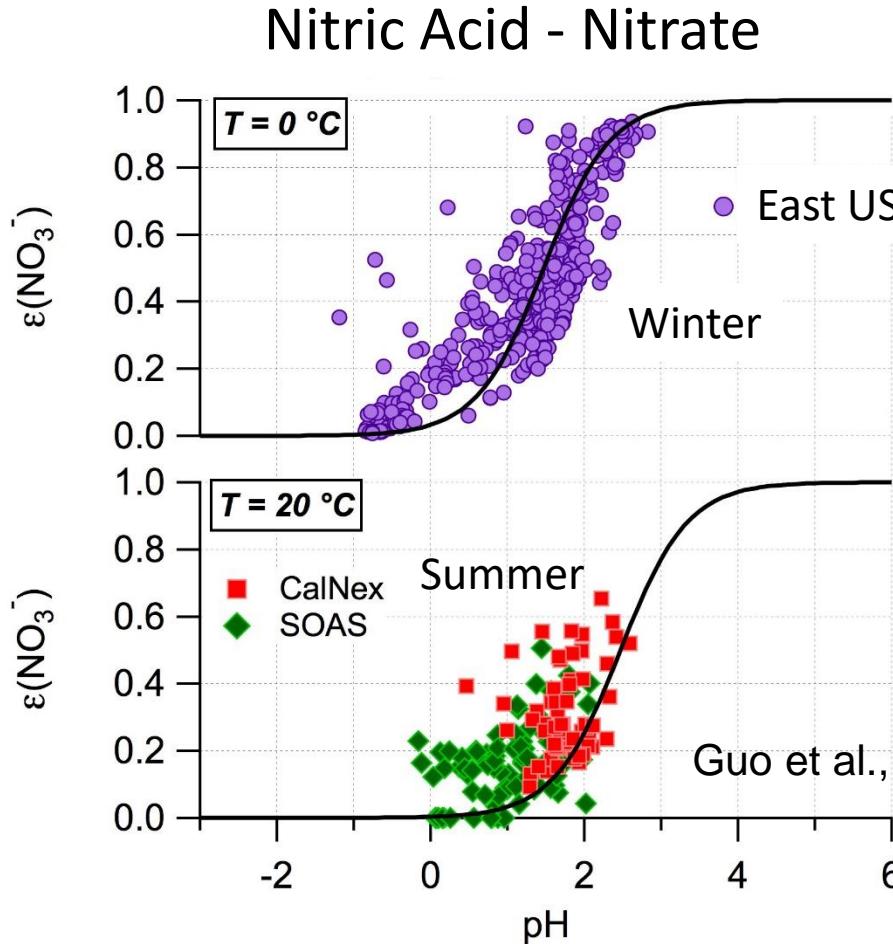


# Fertilization of E.Mediterranean ecosystems from dust (AQUACOSM-plus).



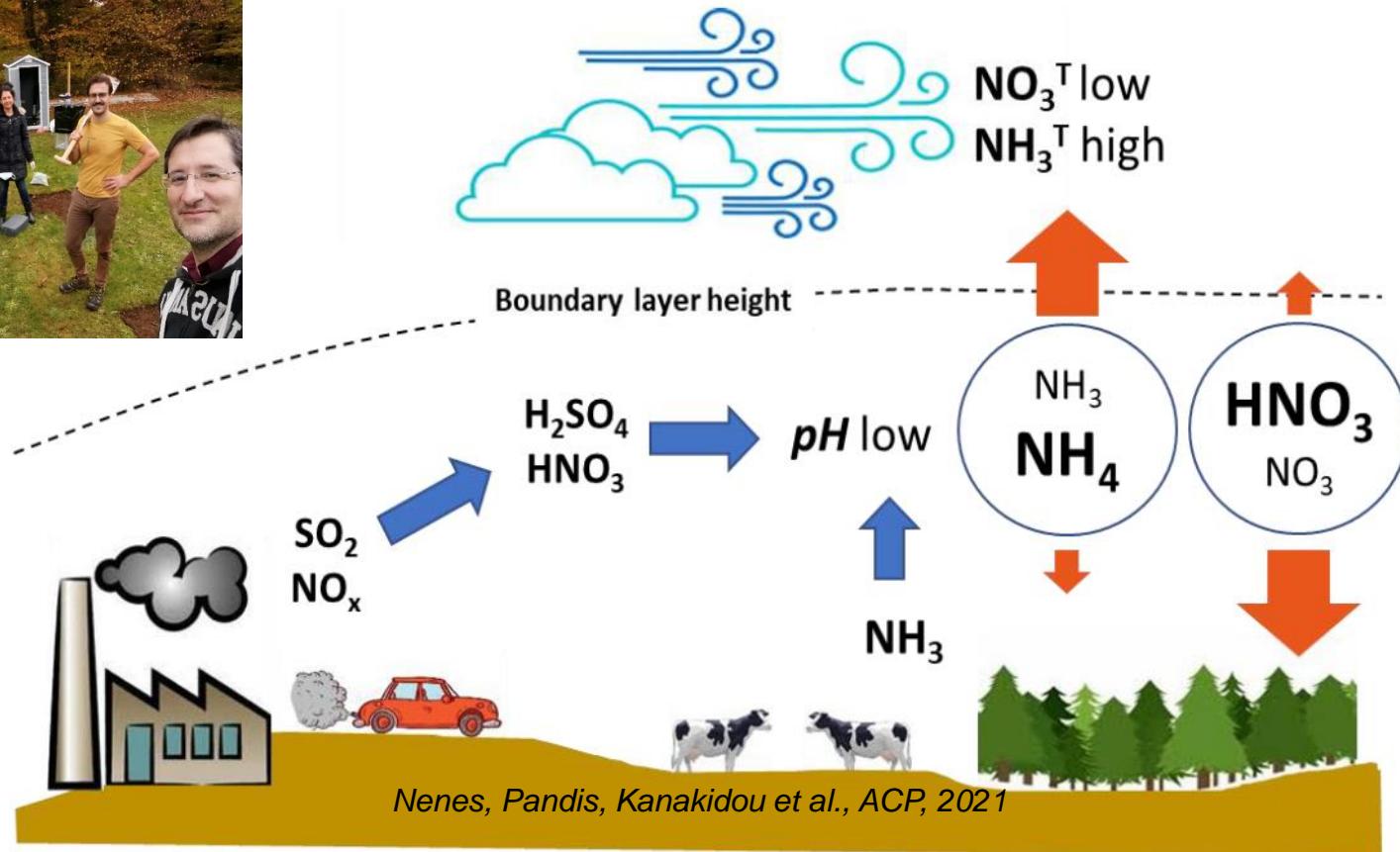
Characterization of aerosol generated from perturbed ecosystems for understanding impacts on clouds.

# pH and observed partitioning of nitrate and ammonium follow “S - curves”



Consistency between predicted and observed partitioning of both species affirms that predicted *acidity levels with models are reasonable*.

Because of its effect on partitioning, acidity impacts even dry nitrogen deposition – impacting ecosystems



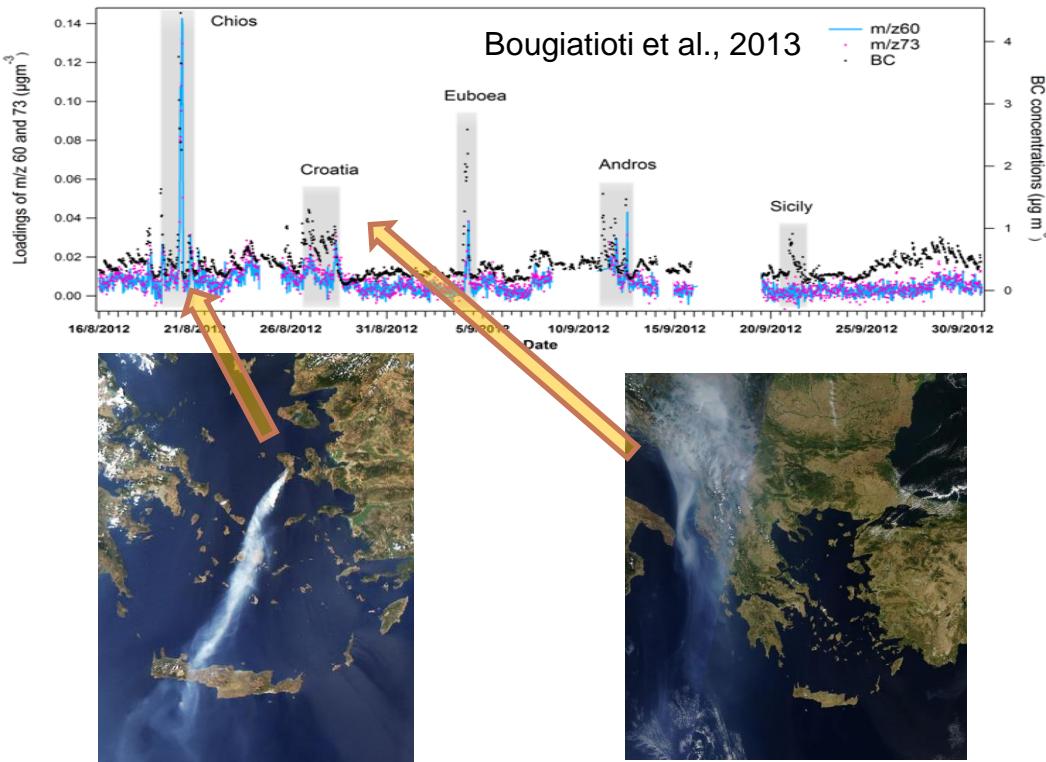
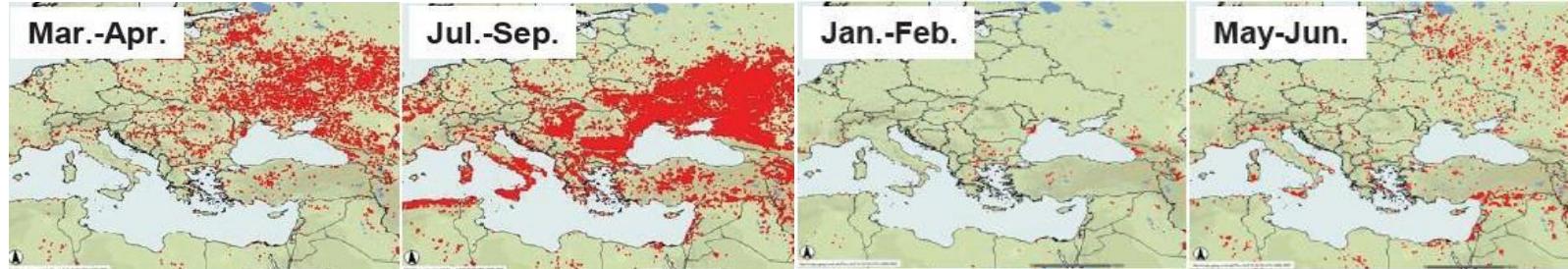
We are now exploring how these deposition pattern changes affect plant growth (ReCLEAN joint initiative lead by our lab)



**Other current exciting areas of research**

# Biomass Burning: a major aerosol source of great influence for our region

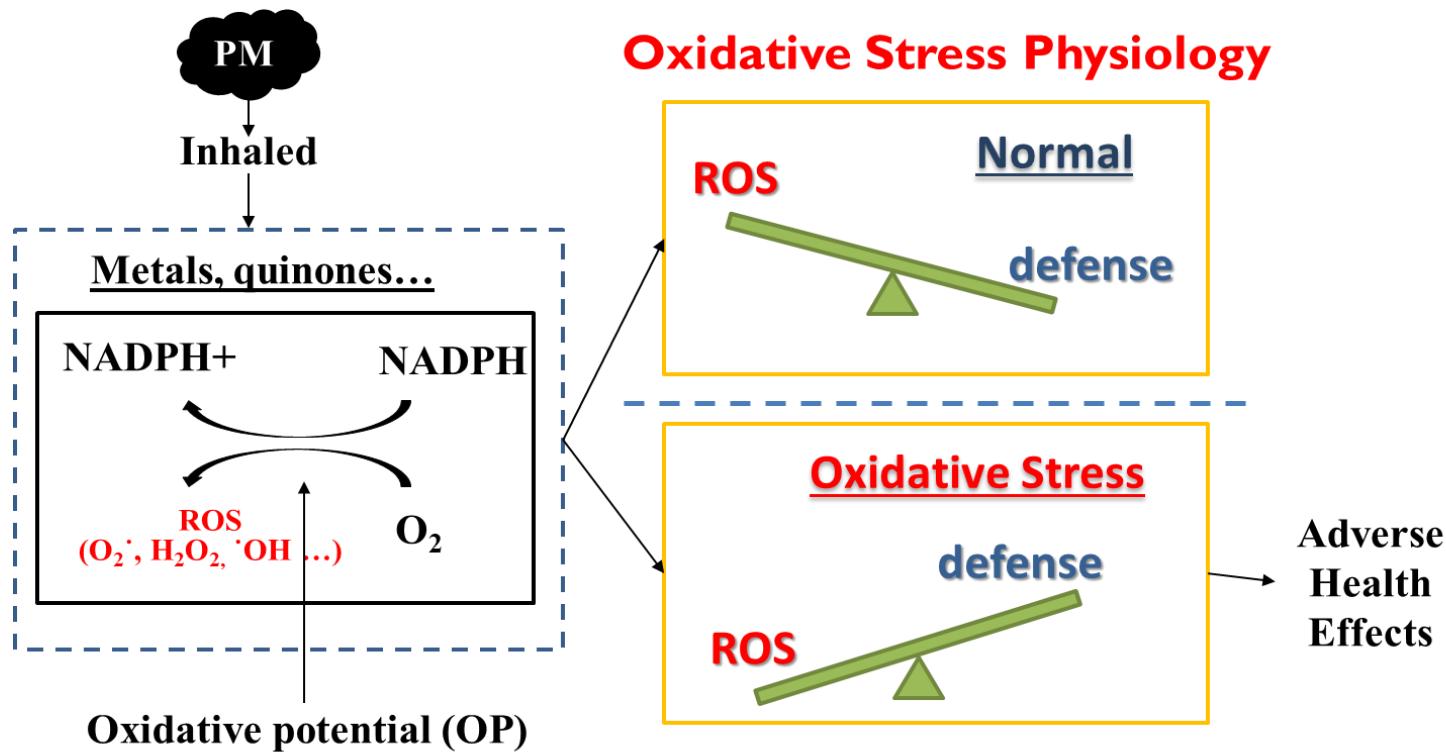
BB smoke is always present in the Eastern Mediterranean.



## BB influence “phases”:

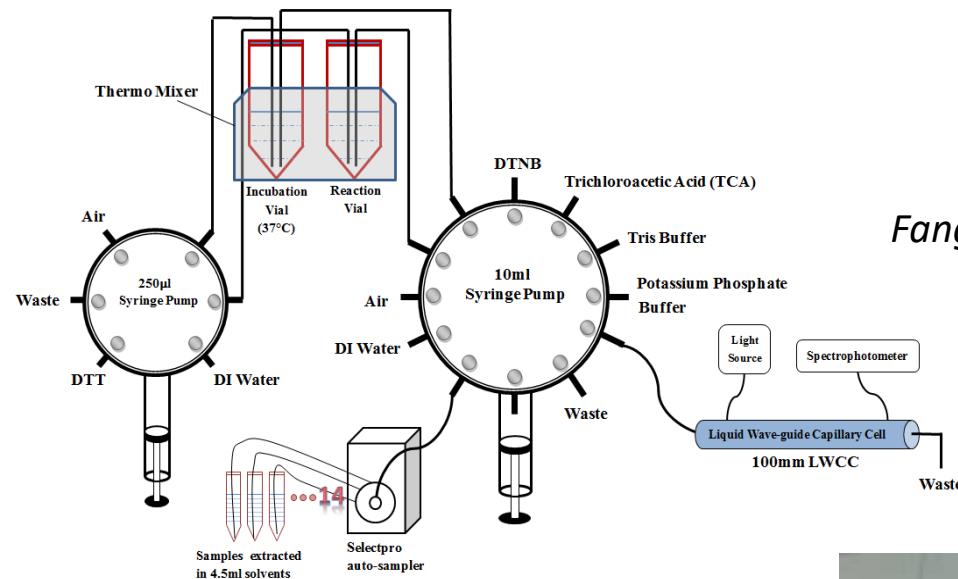
- *Summer/early fall*: wildfire smoke from isolated events that age 0 - 3 days before reaching Greece
- *Winter/early spring*: Fresh BB from domestic burning in urban areas (Athens, Ioannina, etc.).
- Both constitute unique “natural settings” for studying BB aerosol over a range of ages.

# Biomass Burning: Health impacts



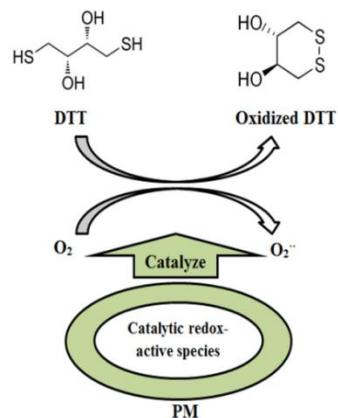
DTT assay	AA assay
chemical surrogate to biological antioxidant	physiological antioxidant in lung lining fluid
$\text{DTT} + \text{O}_2 + \text{H}^+ \xrightarrow[\substack{\text{pH}=7.4 \\ \text{T}=37^\circ\text{C}}]{\text{PM}} \text{DTT}_{\text{ox}} + \text{H}_2\text{O}_2$ <p>[Cho et al., 2005]</p>	$\text{AA} + 2 \text{O}_2 \xrightarrow[\substack{\text{pH}=7.4 \\ \text{T}=37^\circ\text{C}}]{\text{PM}} \text{AA}_{\text{ox}} + 2 \text{O}_2^\cdot$ <p>[Ayres et al., 2008]</p>

# Dithiothreitol (DTT) Assay – An abiotic assay of Oxidative Potential



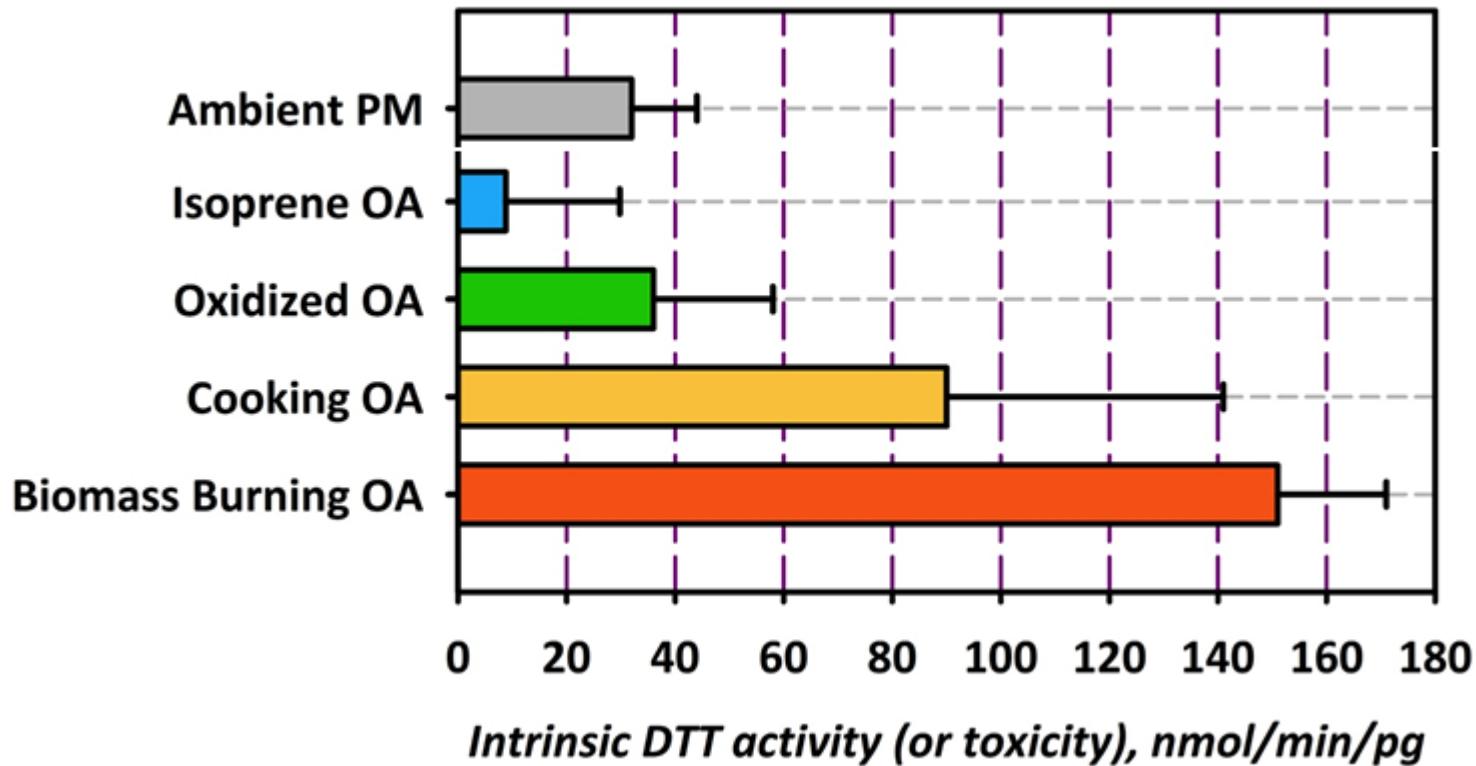
Fang et al. Atmos. Meas. Tech., 2014

DTT Assay at CSTACC-FORTH, Greece



# Intrinsic Potential of OAs to generate ROS

Verma et al., (2017).



- **Highest Toxicity for BBOA (and Cooking OA)**
- Very low toxicity for Isoprene Organic Aerosol (from trees)
- **How do these toxicities evolve as BB ages?**

# ERC CoG PyroTRACH: identify smoke particles and their impacts from emission to deposition

## In-situ sampling & Processing



Finokalia, Crete



Athens, Greece

- Highly populated urban area with fresh BB emissions.
- Remote site exposed to BB plumes 0-3 days old.
- Age *in-situ* BB aerosol in portable environ.chamber
- Characterize aerosol properties



Portable env.chamber

## Lab Generation & Processing

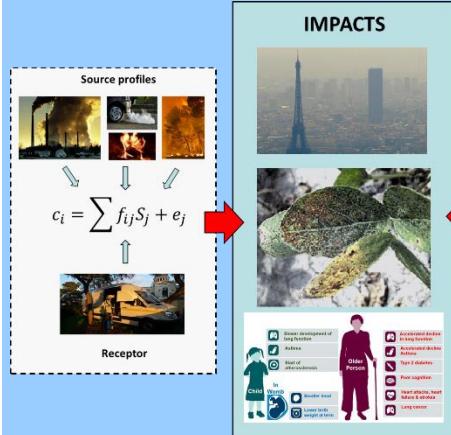


ICE-HT/FORTH Environmental Chamber

- Generate fresh smoke from a variety of BB types.
- Age aerosol in the ICE-HT/FORTH environmental chamber for characteristic oxidation pathways and atmospheric conditions.
- Characterize aerosol properties

- Lab data used for unraveling ambient data.
- Focus on decay of ROS/BrC and stability of chemical markers.

## Impacts & Implications



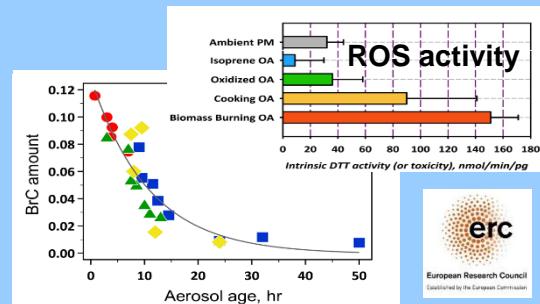
## BBOA Ageing Properties Impacts



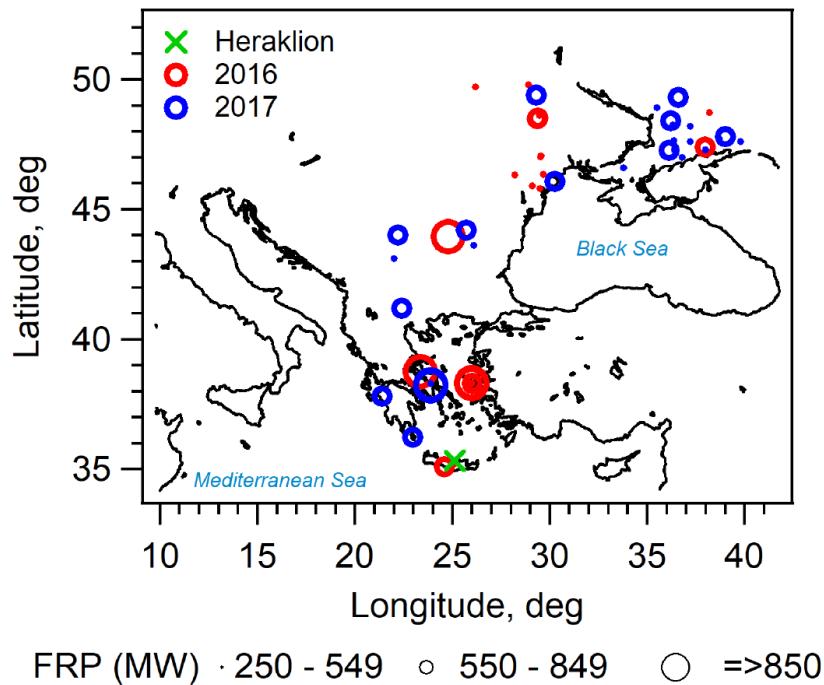
ROS/BrC

## Parameterization

- Determine optical parameters for BrC.
- Intrinsic ROS activity for each BBOA type
- Volatility distributions for BrC, ROS.
- Factor analysis for BBOA contribution to OA.



European Research Council  
Co-funded by the European Commission



**Fig.1:** Locations and sizes of relevant fire events in 2016 (red) and 2017 (blue), as detected by MODIS.

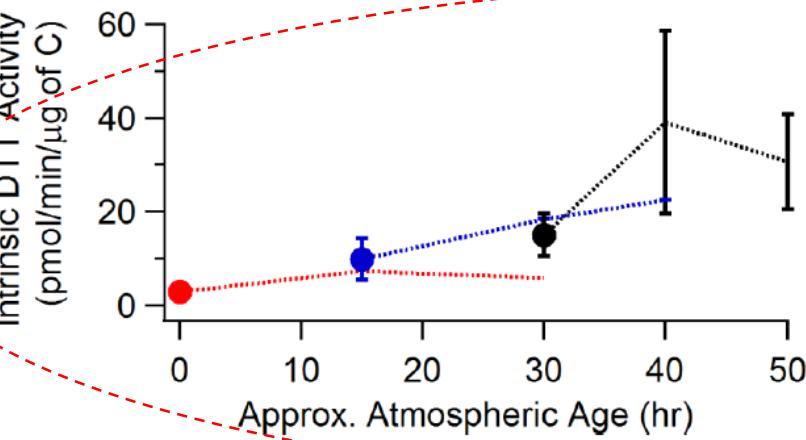
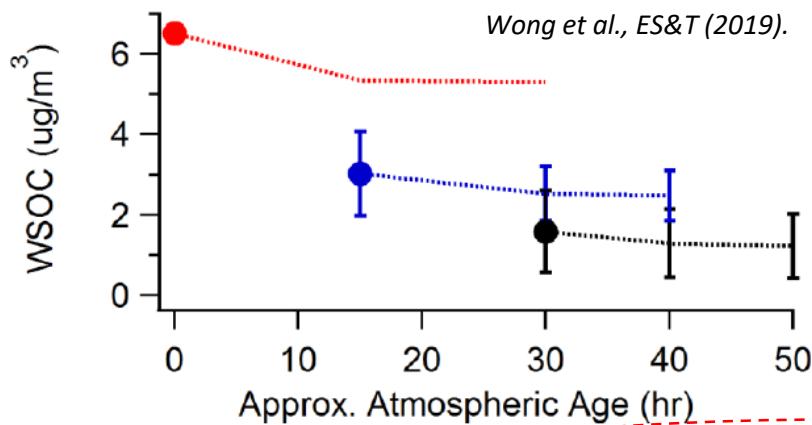


**Apply now to ambient samples:**  
**Filters collected at E. Mediterranean site (Finokalias, Crete) with BB influence**

## Further UVA Light Exposure for Crete Filters (15 and 30 hours)

("Fresh", "Intermediate", and "More Aged" Samples – just WS components)

Analysis: WSOC, WS-DTT, Bulk and Size Separated BrC



RESEARCH & INNOVA

European Commission

Home > Research and innovation > News > Horizon magazine

# HORIZON

The EU Research & Innovation Magazine

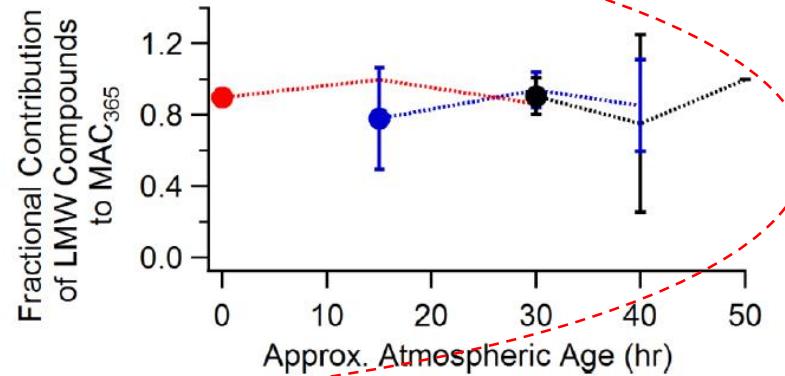
HOME VIEWS TOPICS VIDEOS

ENVIRONMENT

22 July 2020

**'Four times more toxic': How wildfire smoke ages over time**

Air pollution travels globally and affects the health of people around the world. →



As smoke ages: It becomes **more TOXIC** and LESS Brown overall. The large molecules however **REMAIN**.



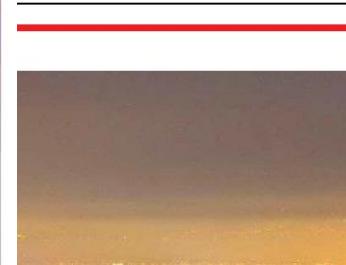
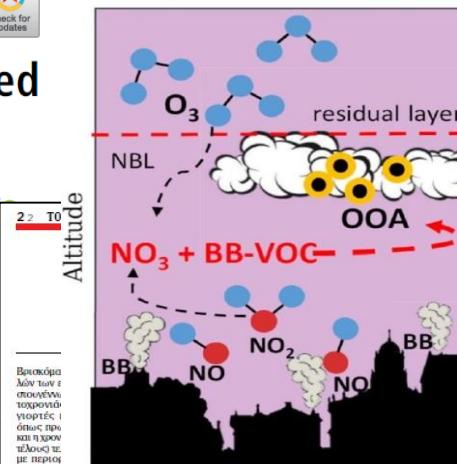
# Rapid dark aging of biomass burning as an overlooked source of oxidized organic aerosol

John K. Kodros<sup>a</sup>, Dimitrios K. Papanastasiou<sup>a,1</sup>, Marco Paglione<sup>a,b</sup>, Mauro Masoli<sup>a,2</sup>, Stefania Squizzato<sup>a</sup>, Kalliopi Florou<sup>a</sup>, Ksavousta Skyllakou<sup>a</sup>, Christos Kaltsonoudis<sup>a</sup>, Athanassios Nenes<sup>a,c,3</sup>, and Spyros N. Pandis<sup>a</sup>

<sup>a</sup>Institute of Chemical Engineering Sciences, Foundation for Research & Technology-Hellas, Patras 26504, Greece; <sup>b</sup>Institute of Atmospheric Sciences & Climate, Italian National Research Council, Bologna 40129, Italy; <sup>c</sup>School of Architecture, Civil and Environmental Engineering, Swiss Federal Institute of Technology Lausanne, Lausanne 1015, Switzerland; and <sup>d</sup>Department of Chemical Engineering, University of Patras, Patras 26504, Greece

Edited by Mark Thiemens, University of California San Diego, La Jolla, CA, and approved October 20, 2020 (received for review May 22, 2020)

Oxidized organic aerosol (OOA) is a major component of ambient particulate matter, substantially impacting climate, human health, and ecosystems. OOA is readily produced in the presence of sunlight, and requires days of photooxidation to reach the levels observed in the atmosphere. High concentrations of OOA are thus expected in the summer; however, our current mechanistic understanding fails to explain elevated OOA during wintertime periods of low photochemical activity that coincide with periods of intense biomass burning. As a result, atmospheric models underpredict As stricter controls on fossil fuel combustion are implemented, residential biomass burning (BB) as a source of heating or cooking is becoming an increasingly important source of OA in urban environments (1, 11, 12). Further, increasing rates of wildfire from climate change are increasing the frequency of smoke-impacted days in urban areas (12–14). BB emissions in high concentrations of POA, SVOCs, IVOCs, and VOCs (15) thus making BB a key source of OOA. Previous research has focused on quantifying the concentration of OOA formed by



# ΓΙΑΤΙ ΤΑ TZAKIA ΕΙΝΑΙ Η... PIZA ΤΟΥ ΤΟΞΙΚΟΥ ΝΕΦΟΥΣ ΤΗ ΝΥΧΤΑ

- New chemistry rapidly changes BB smoke at night (surprise).
- Inclusion in models explains large biases.
- *Important impacts* on the BB toxicity as it ages.
- Reshapes our understanding on how BB evolves over time, especially in urban centers and populated regions.



εξαπίσια πλαις διαδικασίας με πρωταγωνιστή έναν... δράκοντα (της Χιμέας). Το μήνυμα της μελέτης είναι σαφές προς τους κατόκους των ήδη επιβαρυν-



**Φυσικό εργαστήριο**  
Η νέα μελέτη που δημοσιεύθηκε την εβδομάδα που μας πέρασε στην έκτη πεντάετη στην *Proceedings of the National Academy of Sciences*, (PNAS), δι-

διακός Πολυτεχνικής Σχολής της Λοζάννης (École Polytechnique Fédérale de Lausanne) οπουτακά τις διεργασίες στην απόδοση - για την αύξηση της απόδοσης της παραγωγής.

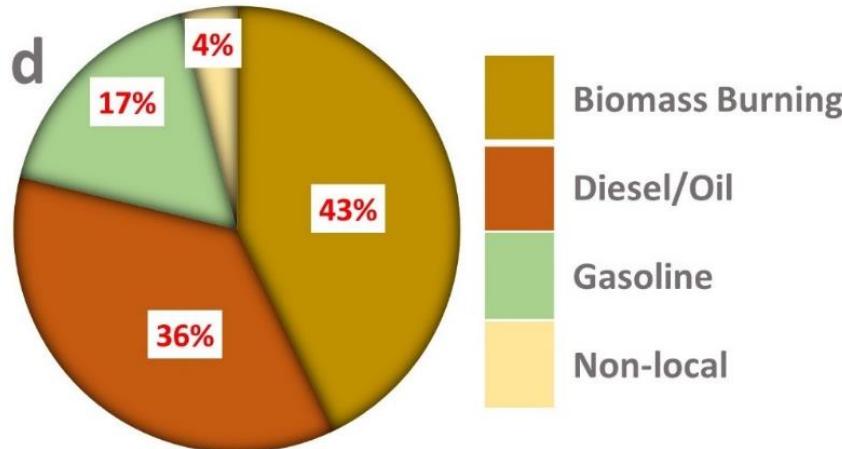
Lausanne - EPFL). Όπως εξηγεί μαλώνιας στο BH-  
και η Ελλάδα θεωρεί-  
ται το καλύτερο φυσικό  
Νυκτόβιος ρύπος!

τα χαράματα» περιγράφει  
ο καθηγητής.

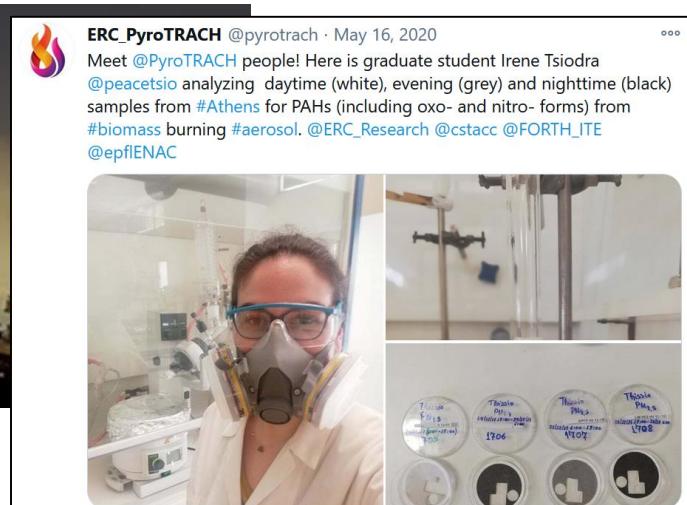
# PAHs from BB during wintertime biomass burning episodes



## estimated annual carcinogen exposure for Athens



**Wintertime exposure dominates annual exposure to carcinogens (PAHs). BB is > 50% responsible**



A screenshot of a The Guardian news article. The headline reads: 'Wood burners cause nearly half of urban air pollution cancer risk - study'. The article discusses how wood smoke is a more important carcinogen than vehicle fumes, based on an Athens analysis. It includes a photograph of smoke rising from a chimney and a quote from Damian Carrington, Environment editor.

Tsiodra, I., Grivas, G., Tavernaraki, K., Bougiatioti, A., Apostolaki, M., Paraskevopoulou, D., Gogou, A., Parinos, C., Oikonomou, K., Tsagkaraki, M., Zarmpas, P., Nenes, A., and Mihalopoulos, N.: Annual exposure to PAHs in urban environments linked to wintertime wood-burning episodes, *Atmos. Chem. Phys.*, 2021.



**Other current exciting areas of research**

# Bioaerosol research: Quantifying airborne microbes, their interactions and impacts

npr

## Bird, Plane, Bacteria? Microbes Thrive In Storm Clouds

by VÉRONIQUE LACAPRA

January 29, 2013 3:38 AM ET

Online Impact



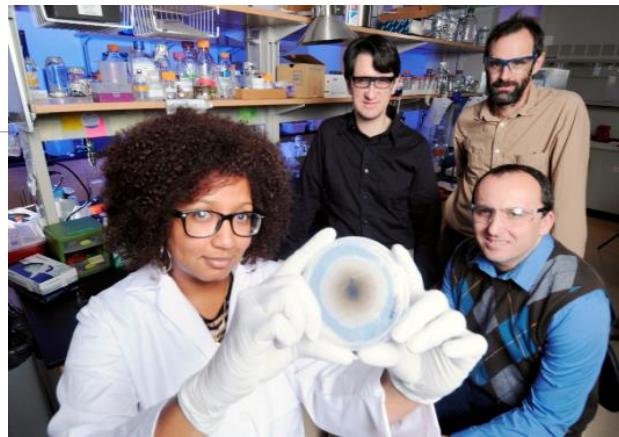
PNAS

## Microbiome of the upper troposphere: Species composition and prevalence, effects of tropical storms, and atmospheric implications

Natasha DeLeon-Rodríguez<sup>a</sup>, Terry L. Lathem<sup>b</sup>, Luis M. Rodríguez-R<sup>a</sup>, James M. Barazesh<sup>c</sup>, Bruce E. Anderson<sup>d</sup>, Andreas J. Beyersdorf<sup>d</sup>, Luke D. Ziemba<sup>d</sup>, Michael Bergin<sup>b,c</sup>, Athanasios Nenes<sup>b,e,1</sup>, and Konstantinos T. Konstantinidis<sup>a,c,1</sup>

<sup>a</sup>School of Biology, <sup>b</sup>School of Earth and Atmospheric Sciences, <sup>c</sup>School of Civil and Environmental Engineering, <sup>d</sup>School of Chemical and Biomolecular Engineering, Georgia Institute of Technology, Atlanta, GA 30332; and <sup>e</sup>Chemistry and Dynamics Branch/Science Directorate, National Aeronautics and Space Administration Langley Research Center, Hampton, VA 23681

Edited by W. Ford Doolittle, Dalhousie University, Halifax, NS, Canada, and approved December 19, 2012 (received for review July 15, 2012)



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ARTICLE OPEN

### Bioaerosols and dust are the dominant sources of organic P in atmospheric particles

Kalliopi Violaki<sup>1,2\*</sup>, Athanasios Nenes<sup>2,3</sup>, Maria Tsagkarakis<sup>4</sup>, Marco Paglione<sup>2,5</sup>, Stéphanie Jacquet<sup>1</sup>, Richard Sempère<sup>1</sup> and Christos Panagiotopoulos<sup>3</sup>

Several studies assessed the impact of inorganic P in fertilizing oligotrophic areas, however, the importance of organic P in such fertilization processes received far less attention. In this study, the amount and origin of organic P delivered to the eastern Mediterranean Sea were characterized in atmospheric particles using the positive matrix factorization model (PMF). Phospholipids together with other chemical compounds (sugars, metals) were used as tracers in PMF. The model revealed that dominant sources of organic P are bioaerosols and dust. The amount of organic P from bioaerosols ( $\sim 4 \text{ Gg P yr}^{-1}$ ) is similar to the amount of soluble inorganic P originating from dust aerosols; this is especially true during highly stratified periods when surface waters are strongly P-limited. The deposition of organic P from bioaerosols can constitute a considerable flux of bioavailable P—even during periods of dust episodes, implying that airborne biological particles can potentially fertilize marine ecosystems.

*npj Climate and Atmospheric Science* (2021) 4:63; <https://doi.org/10.1038/s41612-021-00215-5>

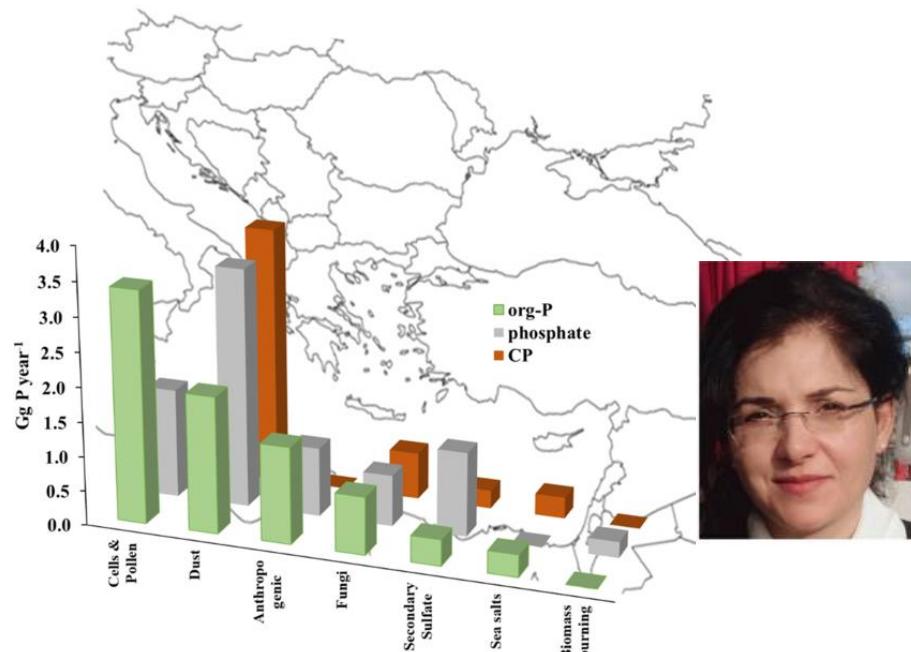
**INTRODUCTION**

Phosphorus (P) is critical to life on Earth, and its distribution in marine<sup>1</sup> and terrestrial ecosystems<sup>2</sup> is shaped by many biogeochemical processes. Inorganic P species (e.g., mono- or diprotonated orthophosphate) comprise the most bioavailable P forms and have been studied for many decades. Organic phosphorus-containing compounds (org-P), such as nucleic acids, phospholipids, inositol phosphates, phosphoamides, phosphonates, phosphoproteins, sugar phosphates, and phosphonic acids, are thought to play a critical role in driving cell growth and metabolism, as well as the community composition of microorganisms<sup>3,4</sup>.

The org-P compounds are ubiquitous in organisms and thus

On the other hand, Wang et al. (2015)<sup>12</sup> indicated that combustion-related emissions of atmospheric P ( $1.8 \text{ Tg P yr}^{-1}$ ) represent over 50% of global atmospheric sources of P ( $3.5 \text{ Tg P yr}^{-1}$ ), suggesting that the perturbation of the global P cycle by anthropogenic emissions is greater than previously thought; however, the estimates are based on a limited number of studies. These assessments highlight the uncertainties in understanding the role of atmospheric P in global biogeochemistry.

The Mediterranean Sea region has been identified as one of the most climate-sensitive marine ecosystems, with increased vulnerability owing to the effects of the increasing demographic and economic development occurring throughout its coastal zone. The long-term impacts on biogeochemical cycles and the ecosystem





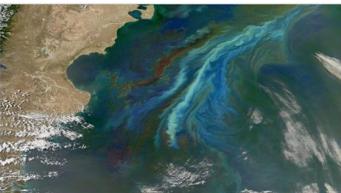
## LAPI – Athanasios (Thanos) Nenes

### Laboratory of atmospheric processes and their impacts



<http://lapi.epfl.ch>

<http://cstacc.iceht.forth.gr>



Biogeochemical Cycles



Aerosol – Cloud – Climate Interactions



Air Quality and Health



Aerosol Chemistry and Impacts



ReCLEAN





## LAPI – Athanasios (Thanos) Nenes

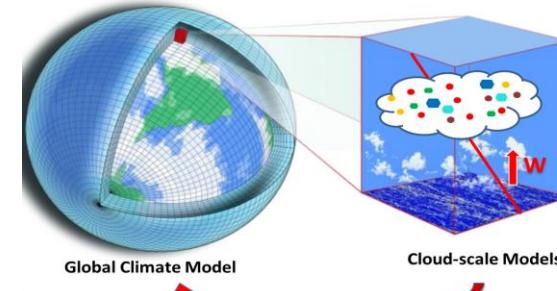
### Laboratory of atmospheric processes and their impacts



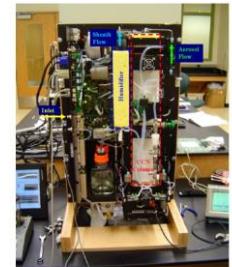
### Field and Laboratory Observations



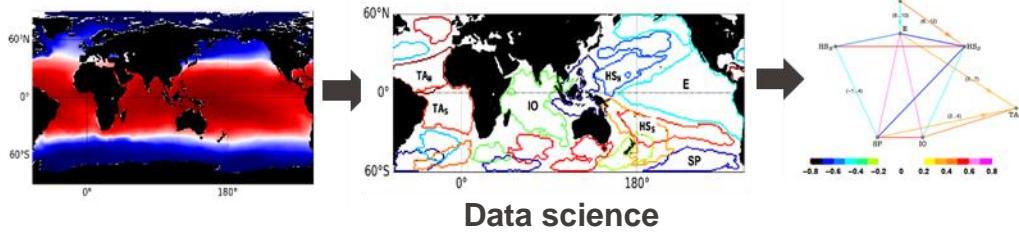
### Modeling

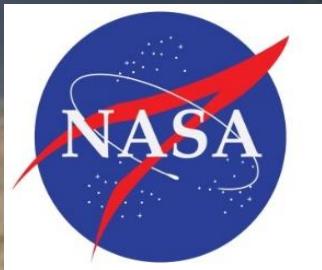


### Instrumentation



Cloud Condensation Nuclei Counter, US Patent 7,656,510





# Thank you!!



For more information, please visit  
<http://lapi.epfl.ch> - @LAPI\_epfl  
And don't forget to visit the LAPI video  
channel at <http://mediaspace.epfl.ch> !

