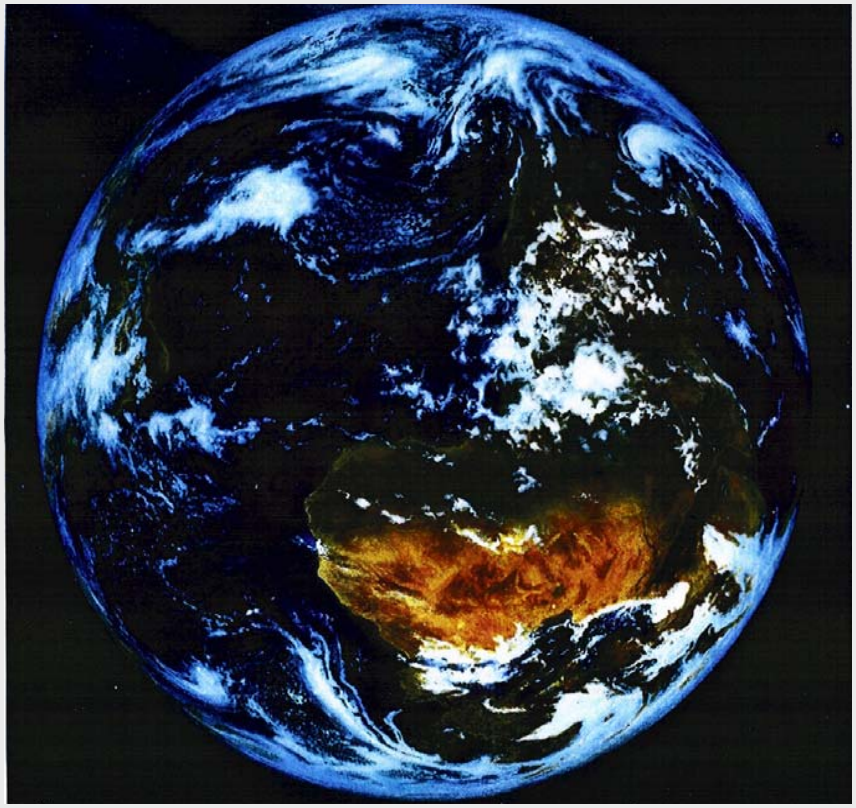
Combustion Generated Aerosol Precursors



Frank Arnold
Max-Planck-Institut für Kernphysik Heidelberg

Talk given at 9 th ETH workshop on ***Combustion Generated Nanoparticles*** ,
15-17 August 2005

Combustion Related Aerosol Particles are important due to their

- Health effects
- Climate effects

Climate Effects of Combustion Related Aerosol Particles

Climate Effects of Combustion Related Aerosol Particles

- direct effect: scattering and absorption of sunlight

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- direct effect: scattering and absorption of sunlight
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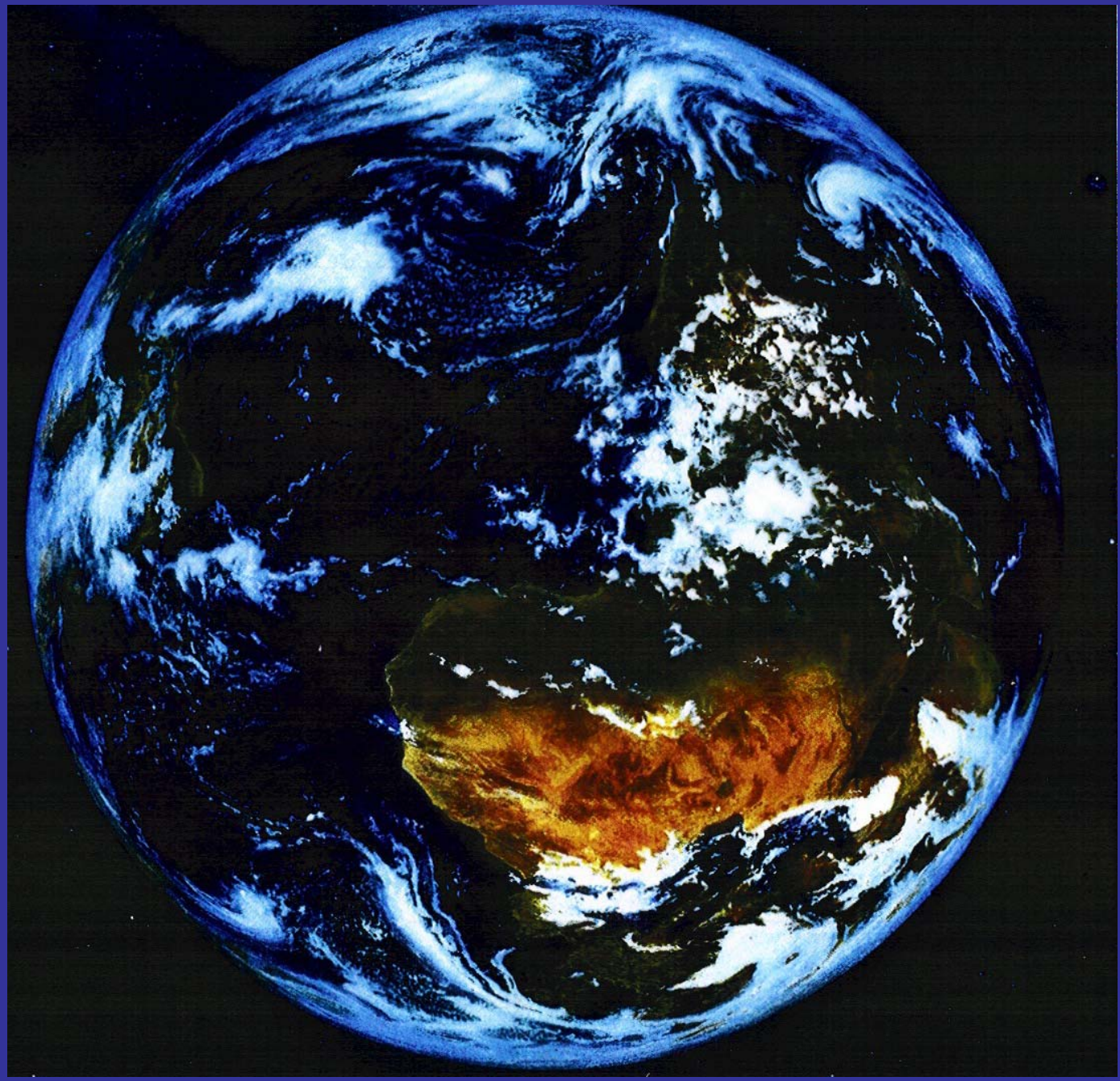
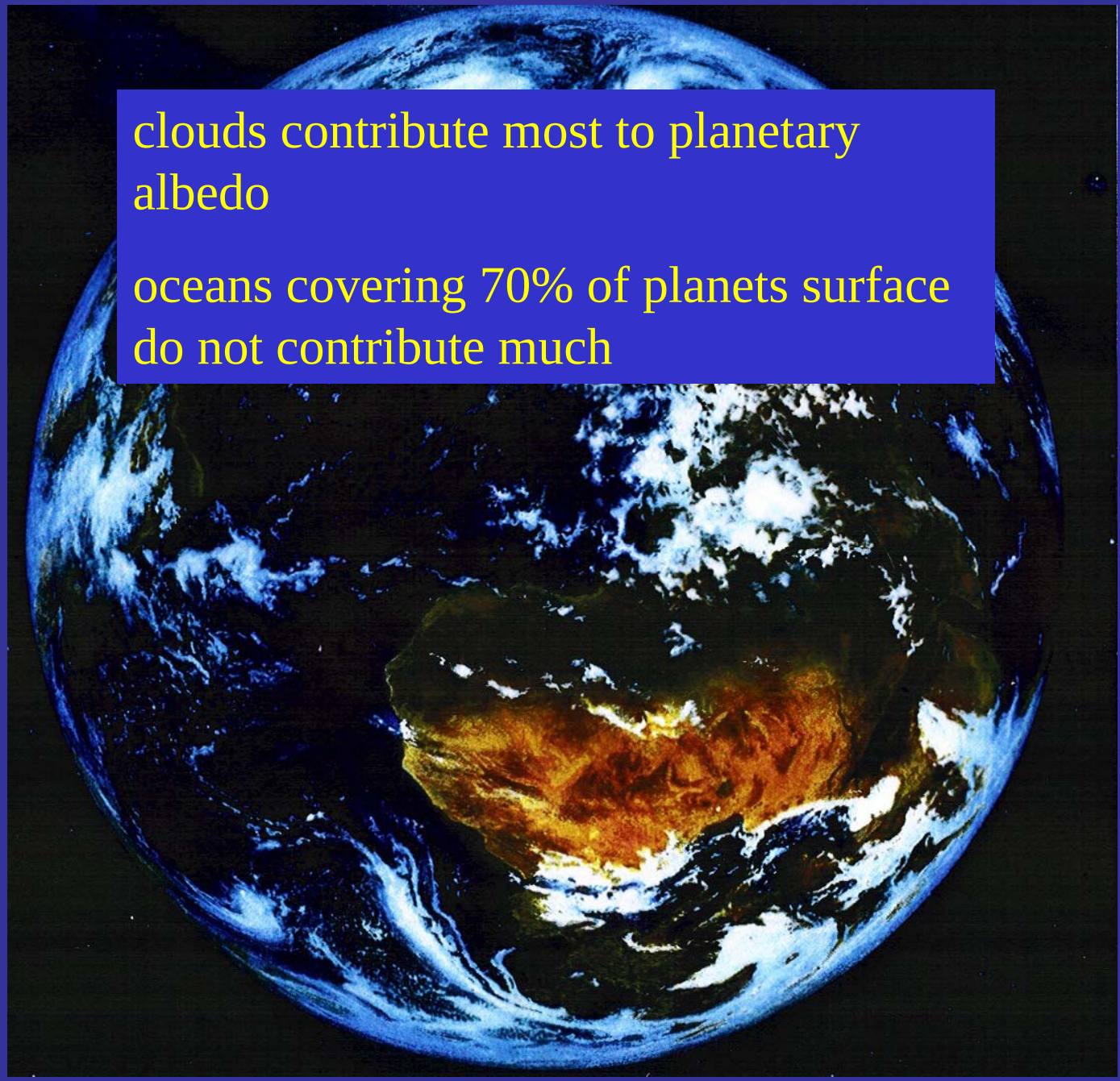
- albedo increase counteracts additional greenhouse warming

Climate Effects of Combustion Related Aerosol Particles

- direct effect: scattering and absorption of sunlight
- indirect effect: particles act as CCN and CFN
- particles tend to increase planetary albedo
- albedo increase counteracts additional greenhouse warming
- albedo increase is not well quantified →

large uncertainties in climate model predictions

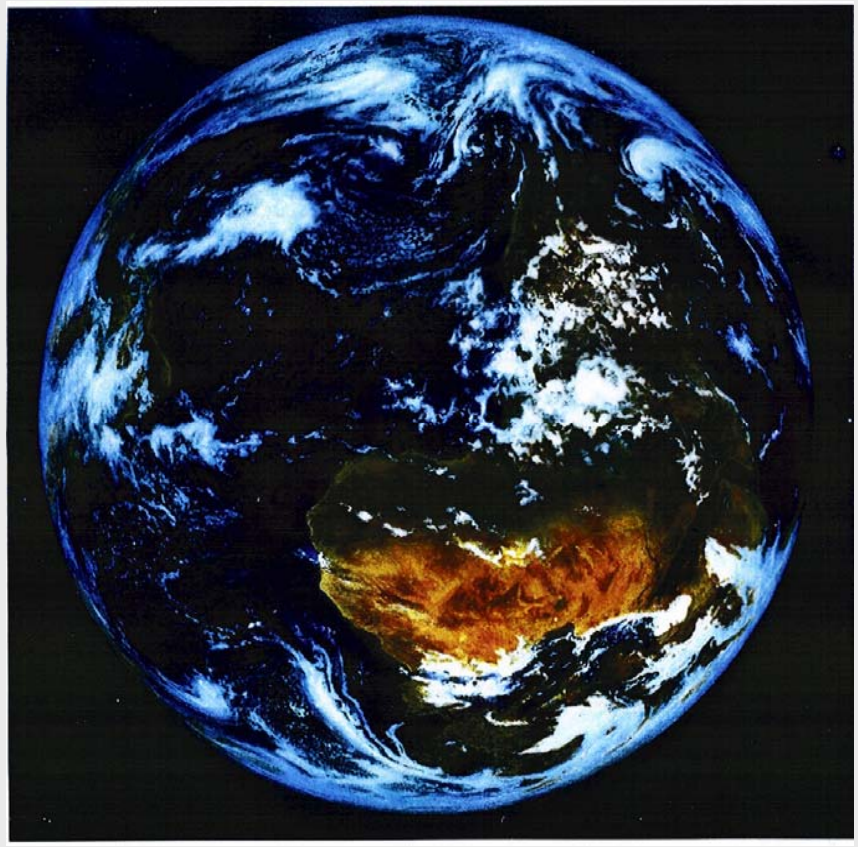


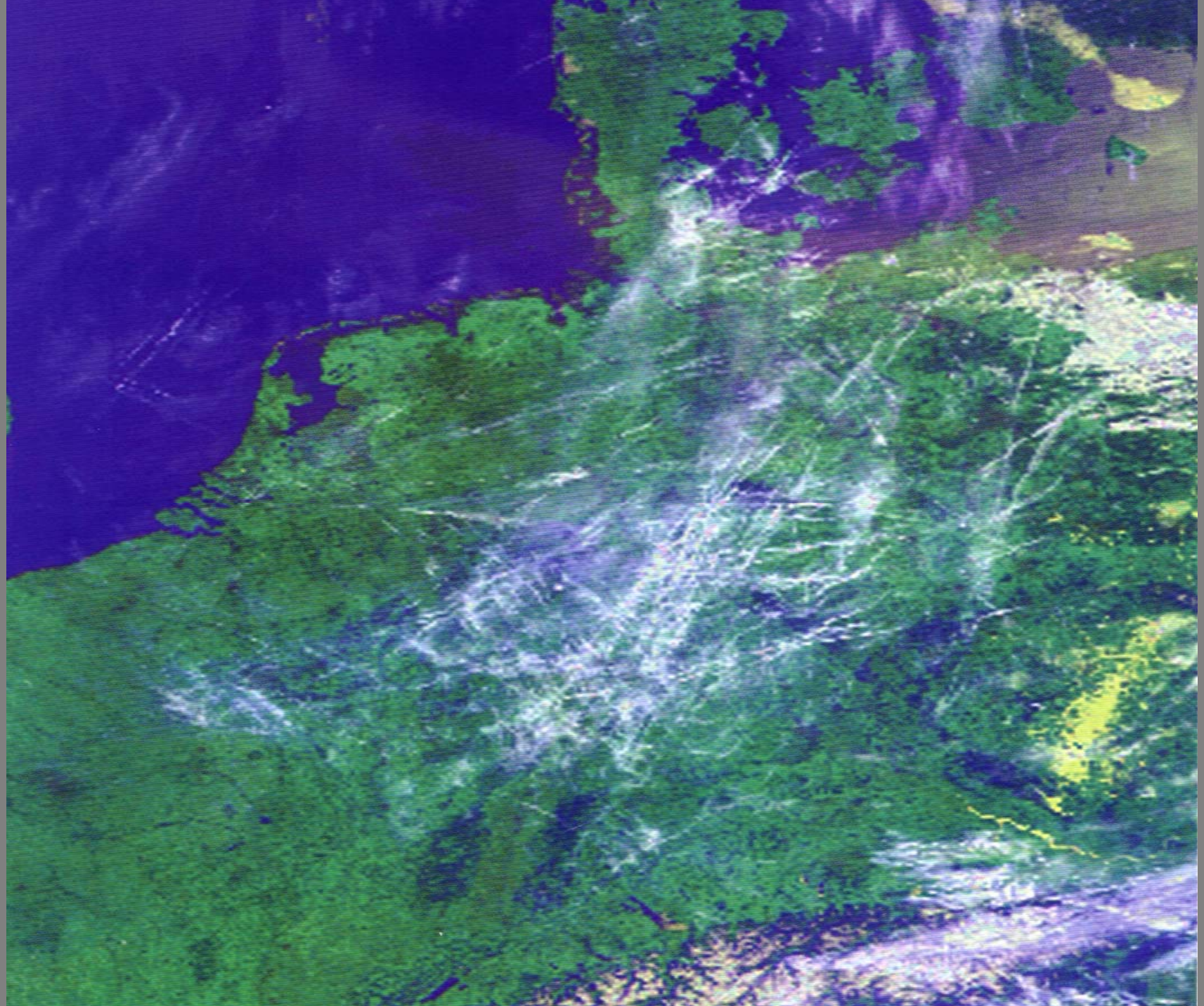


clouds contribute most to planetary albedo

oceans covering 70% of planets surface do not contribute much

Visual Manifestations of Combustion Effects on the Atmosphere





Ship Tracks

Combustion Generated Aerosol Precursors

- **NUCLEATING GASES:**
- **CHEMIIONS:**
- **CONDENSING GASES:**
- **GASEOUS PRECURSORS of NUC. and COND. GASES:**

Combustion Generated Aerosol Precursors

- **NUCLEATING GASES:**
  H_2SO_4
- **CHEMIIONS:**
  $HSO_4^-(H_2SO_4)_a(H_2O)_w$
- **CONDENSING GASES:**
  H_2SO_4, **organics**
- **GASEOUS PRECURSORS of NUC. and COND. GASES:**
  SO_2

Focus of present talk

nucleating and condensing gas

H2SO4

Aerosol Precursor Measurements: Environments

- Free Atmosphere
- Atmospheric Boundary Layer
- Air Craft Wakes (in flight)
- Air Craft Engine Exhaust (at ground level)
- Ship Plumes
- Automobile Exhaust
- Burner Exhaust (laboratory)
- Flow Reactor (laboratory)

Aerosol Precursor Measurements: Environments

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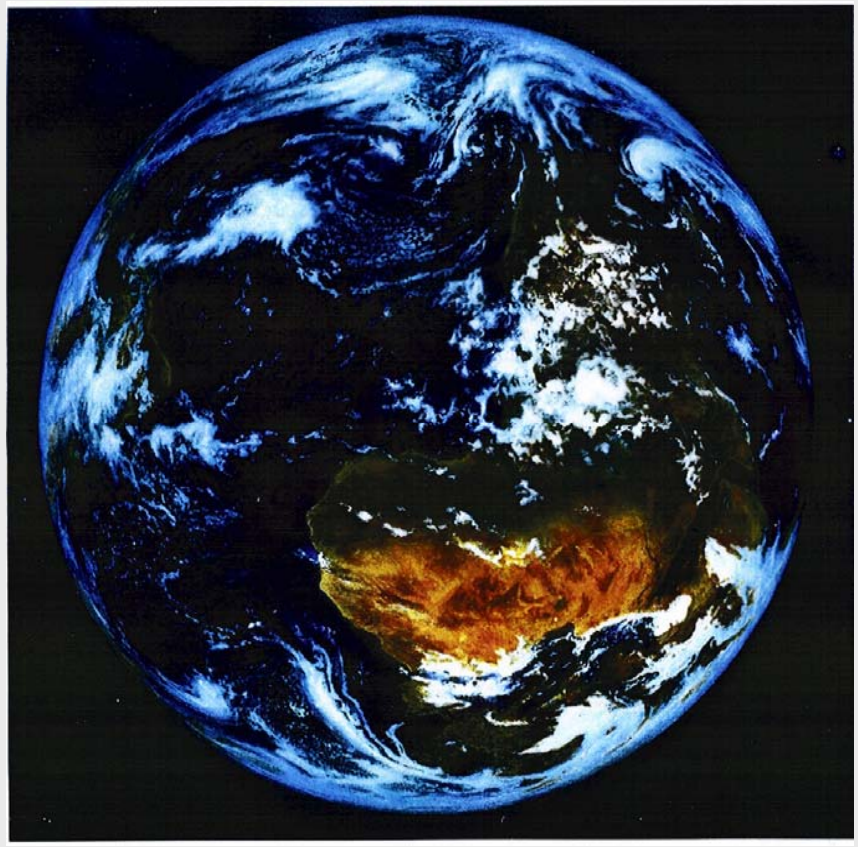
Atmospheric Gaseous Sulfuric Acid



SULFURIC ACID MOLECULE
H_2SO_4

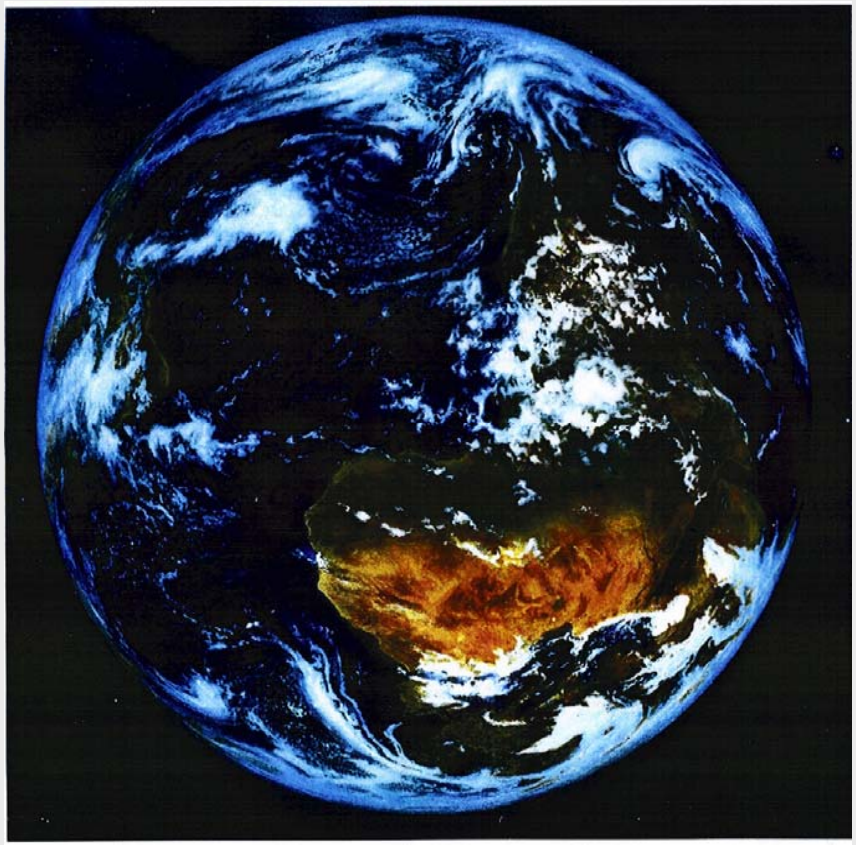
- Most important property : large **GA**
  → proton transfer to other molecule with large **PA**
  (Atmosphere : **H_2O**)
  → Gas-Phase Hydrates **$H_2SO_4(H_2O)_n$**

SULFURIC ACID MOLECULE
H2SO4

- Most important property : large **GA**
  → proton transfer to other molecule with large **PA**
  (Atmosphere : **H2O**)
  → Gas-Phase Hydrates **H2SO4(H2O)n**

- Atmosphere :
  Secondary H2SO4: formed in Atmosphere from **SO2**
  Primary H2SO4 : released from combustion
  Example: Aircraft

Sources and Sinks of Atmospheric H2SO4



Atmospheric Gaseous Sulfuric Acid Sources and Sinks

SO2

Atmospheric Gaseous Sulfuric Acid Sources and Sinks

SO2

SO2-Sources (megatons per year):

Fossil Fuel combustion : 78.1

Oceanic Plankton : 15.4

Volcanism : 9.4

Atmospheric Gaseous Sulfuric Acid Sources and Sinks

clouds

SO2

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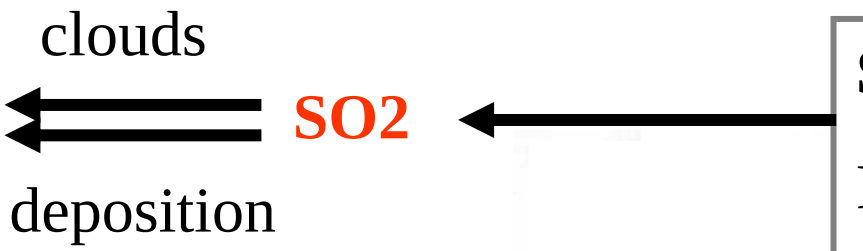
Atmospheric Gaseous Sulfuric Acid Sources and Sinks



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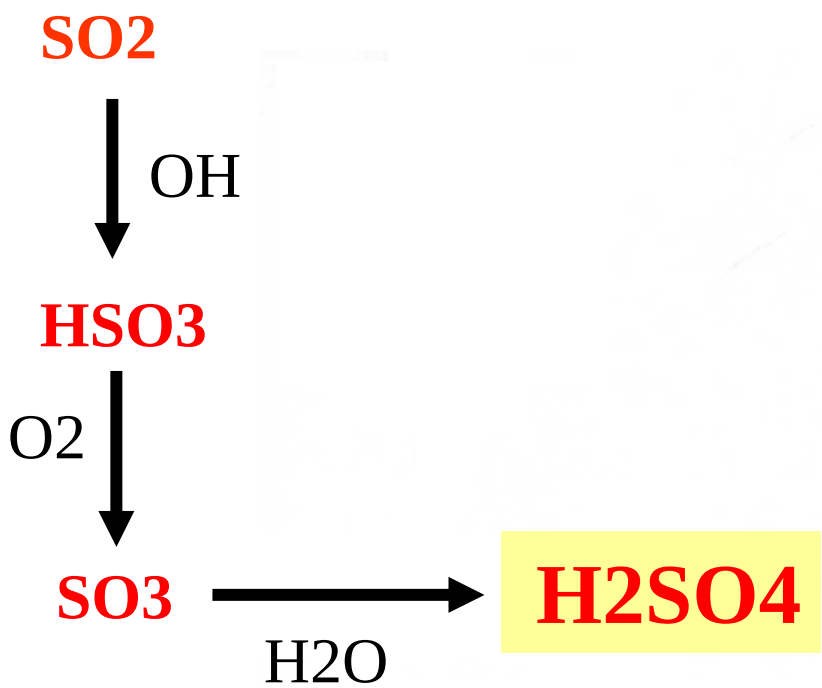
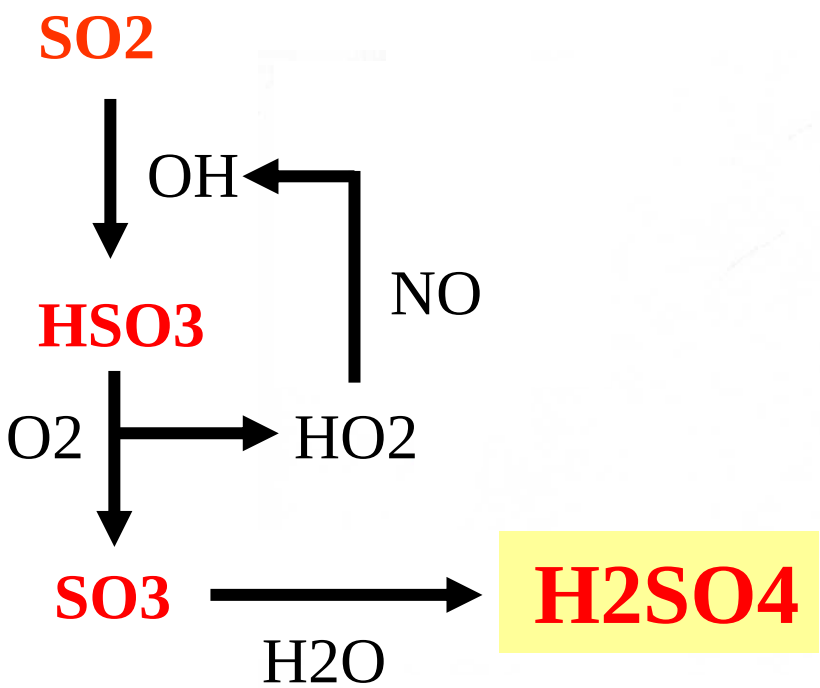
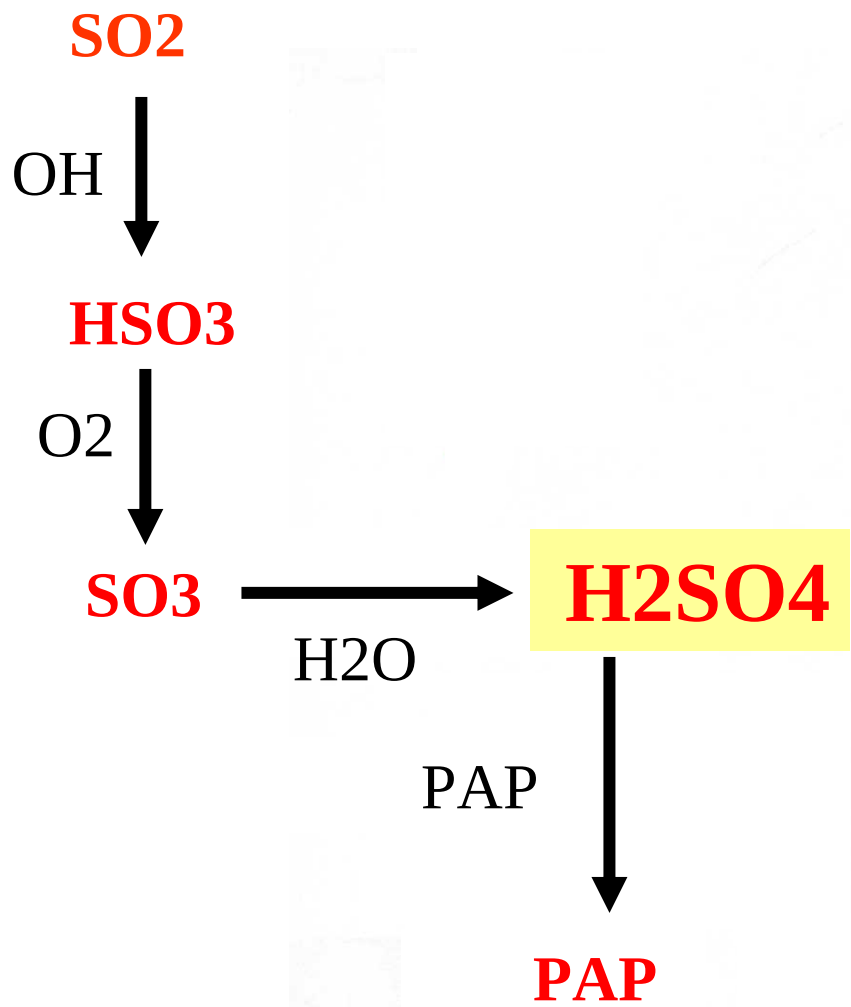
Atmospheric Gaseous Sulfuric Acid Sources and Sinks



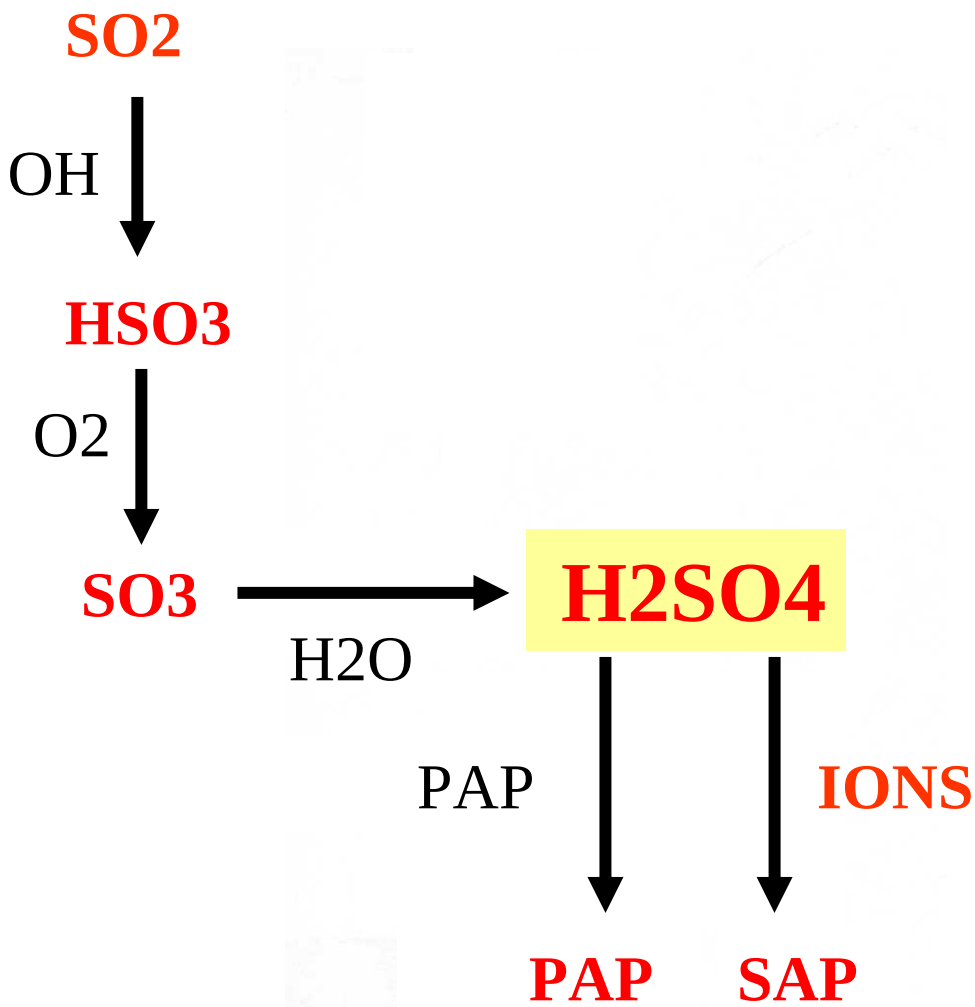
Atmospheric Gaseous Sulfuric Acid Sources and Sinks



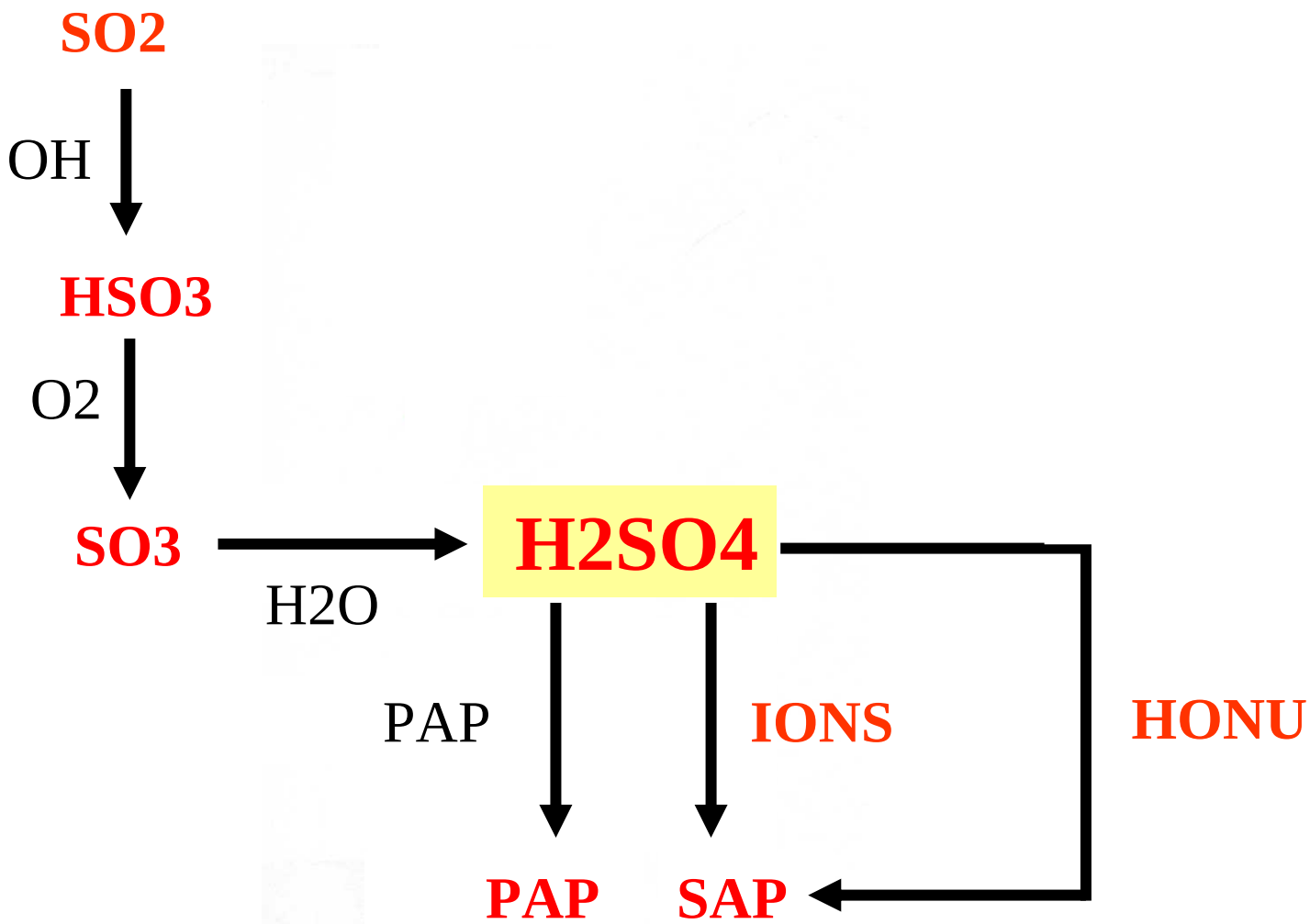
Atmospheric Gaseous Sulfuric Acid Sources and Sinks



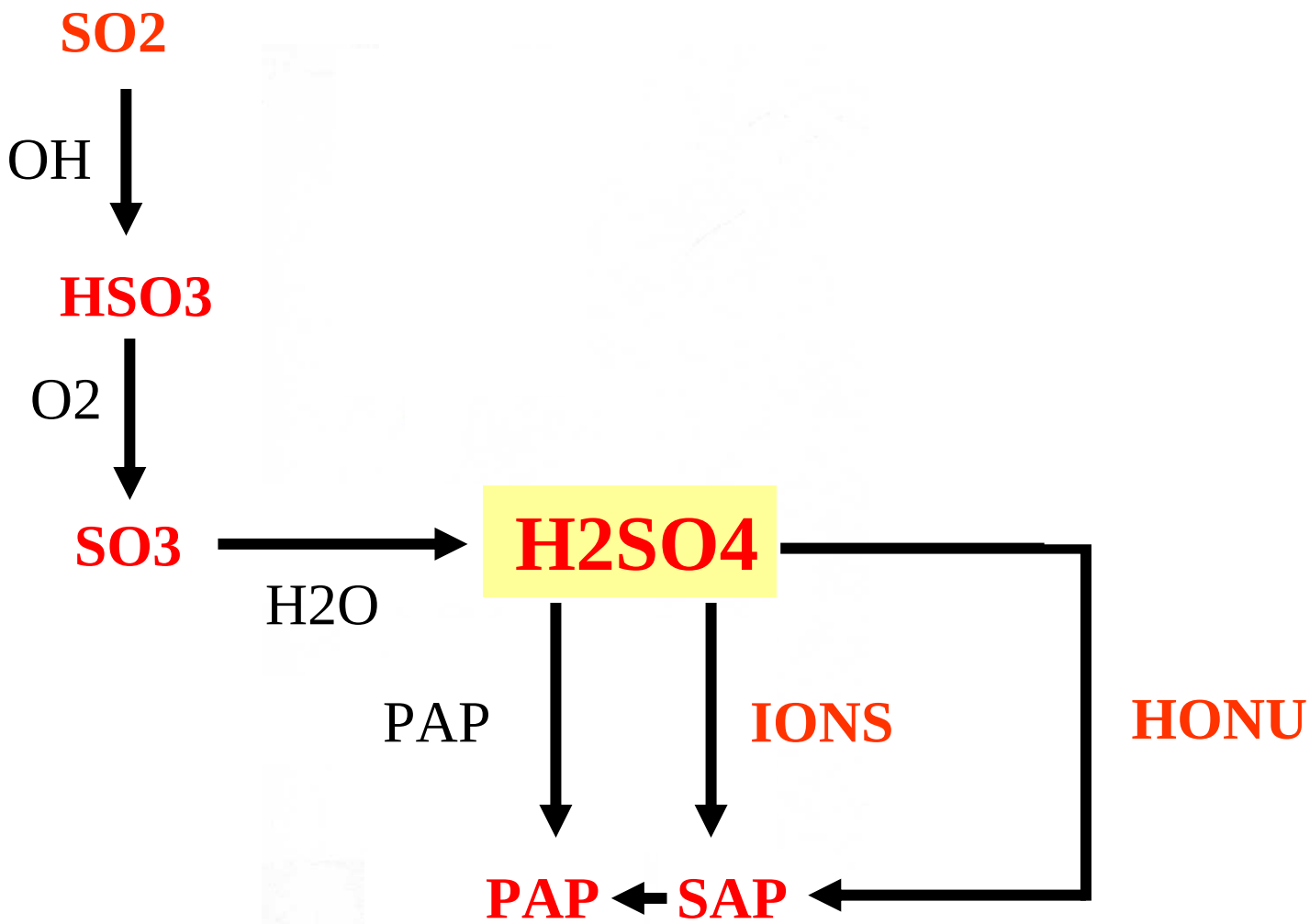
Atmospheric Gaseous Sulfuric Acid Sources and Sinks



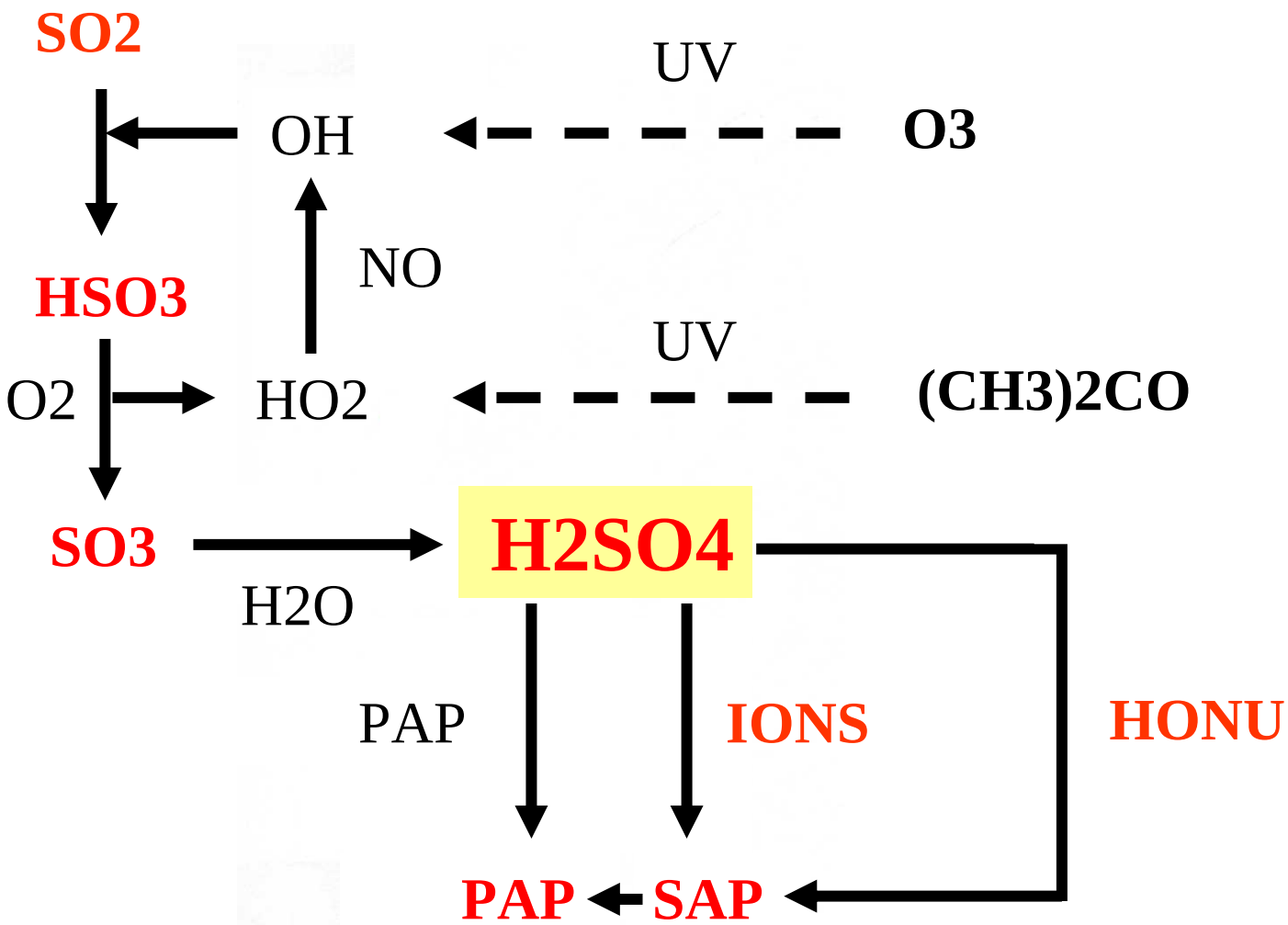
Atmospheric Gaseous Sulfuric Acid Sources and Sinks



Atmospheric Gaseous Sulfuric Acid Sources and Sinks



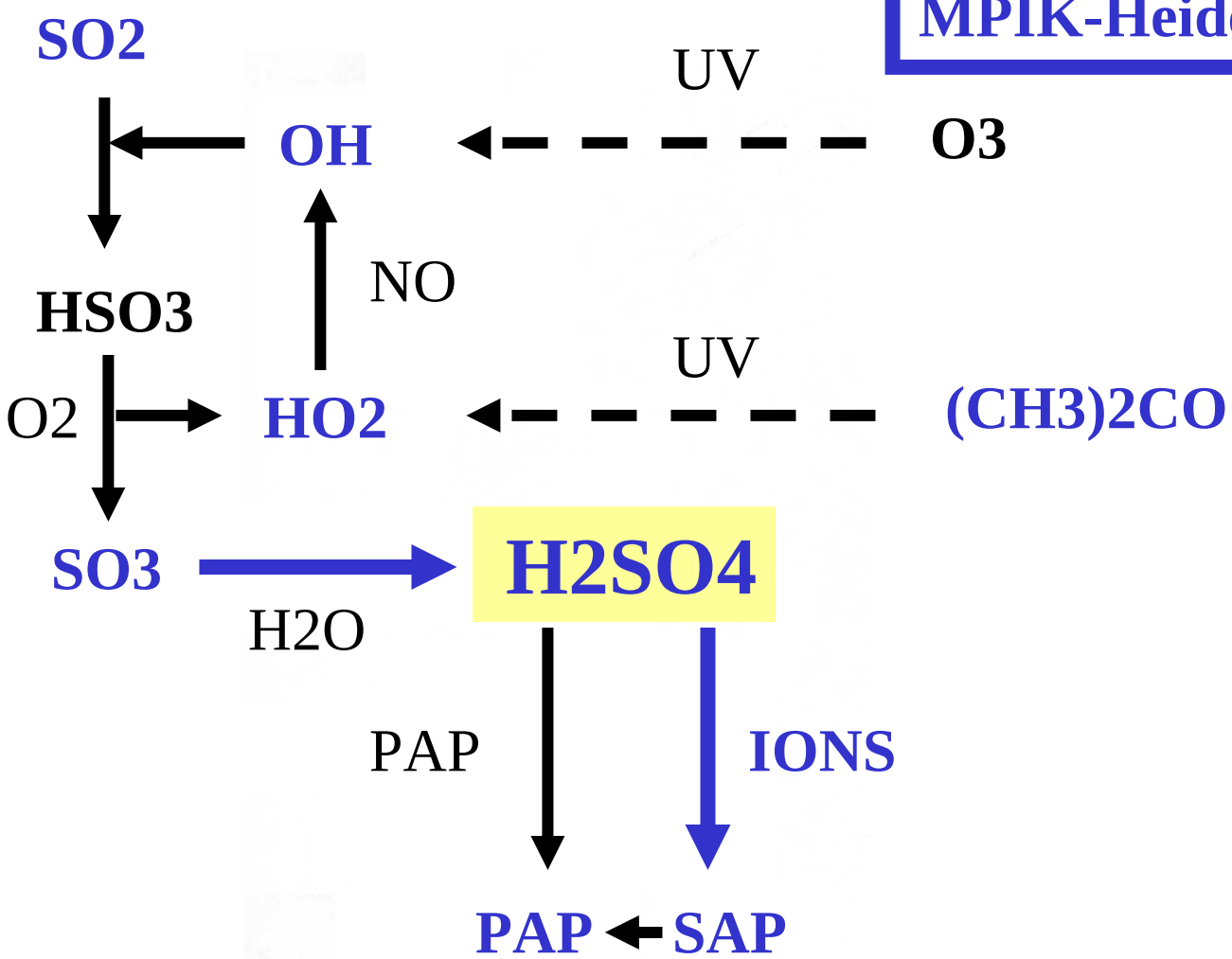
Atmospheric Gaseous Sulfuric Acid Sources and Sinks



Atmospheric Gaseous Sulfuric Acid Sources and Sinks

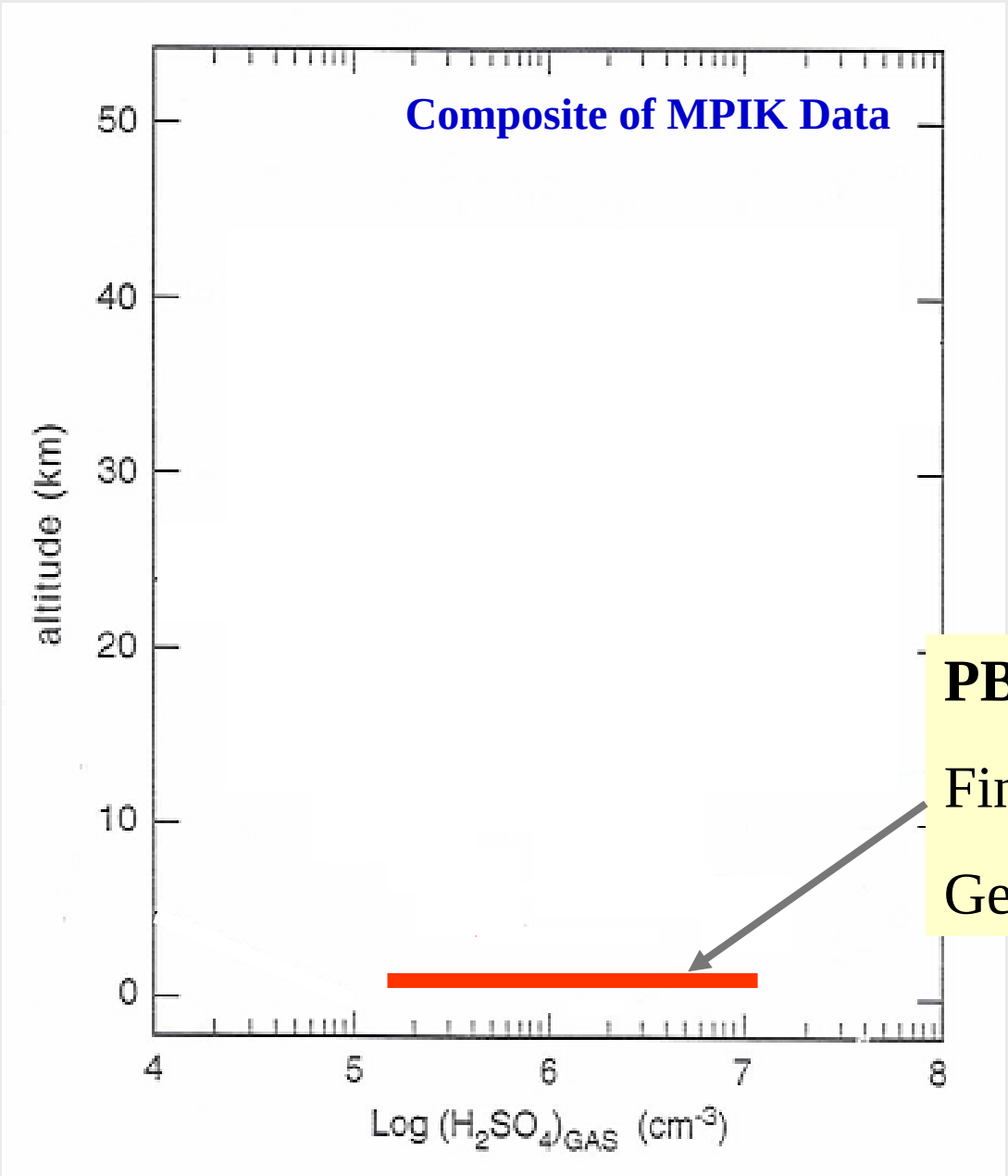
Measured by

MPIK-Heidelberg



Measurements of Atmospheric Gaseous Sulfuric Acid by MPIK Heidelberg

ATMOSPHERIC GASEOUS SULFURIC ACID

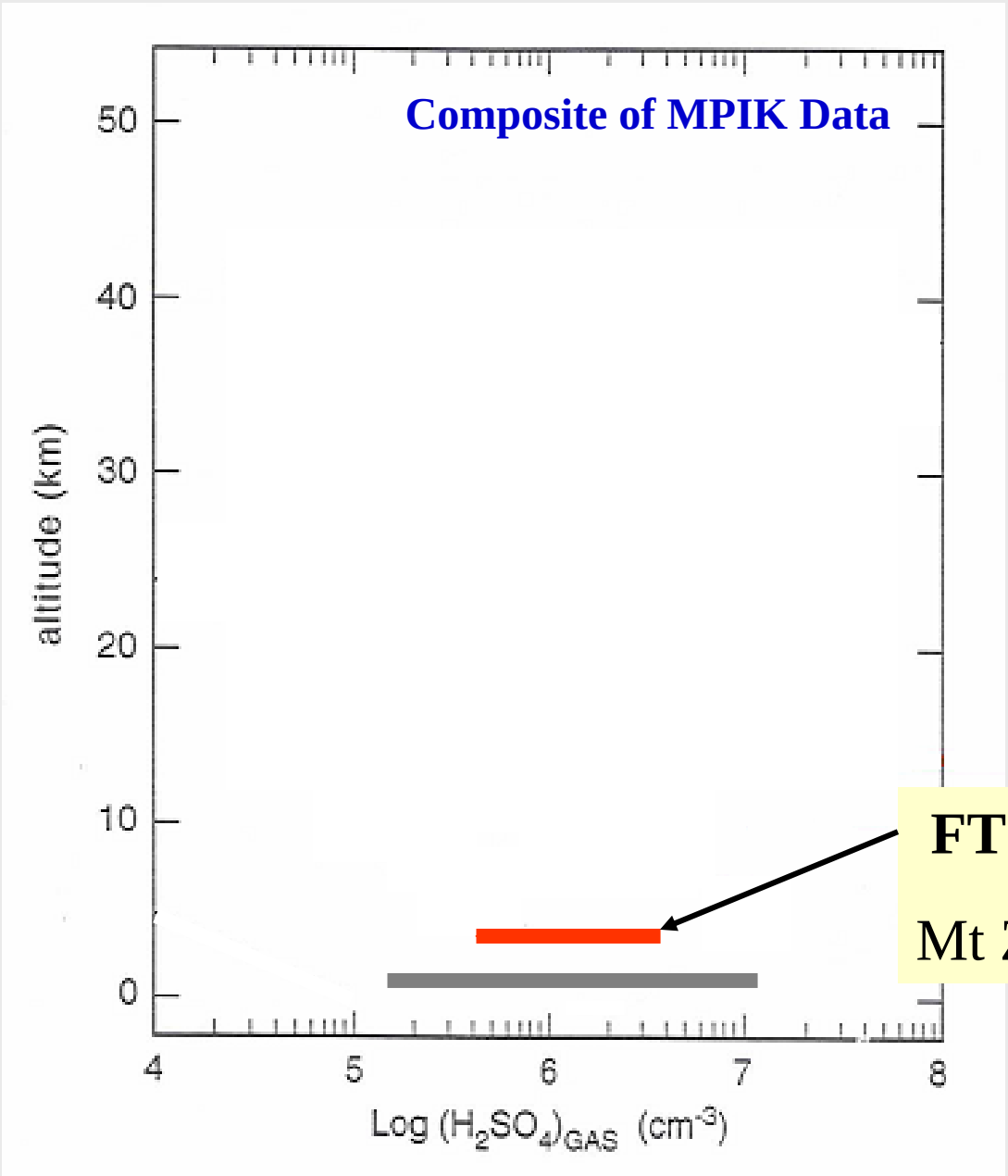


PBL:

Finland (Hy)

Germany (HD)

ATMOSPHERIC GASEOUS SULFURIC ACID



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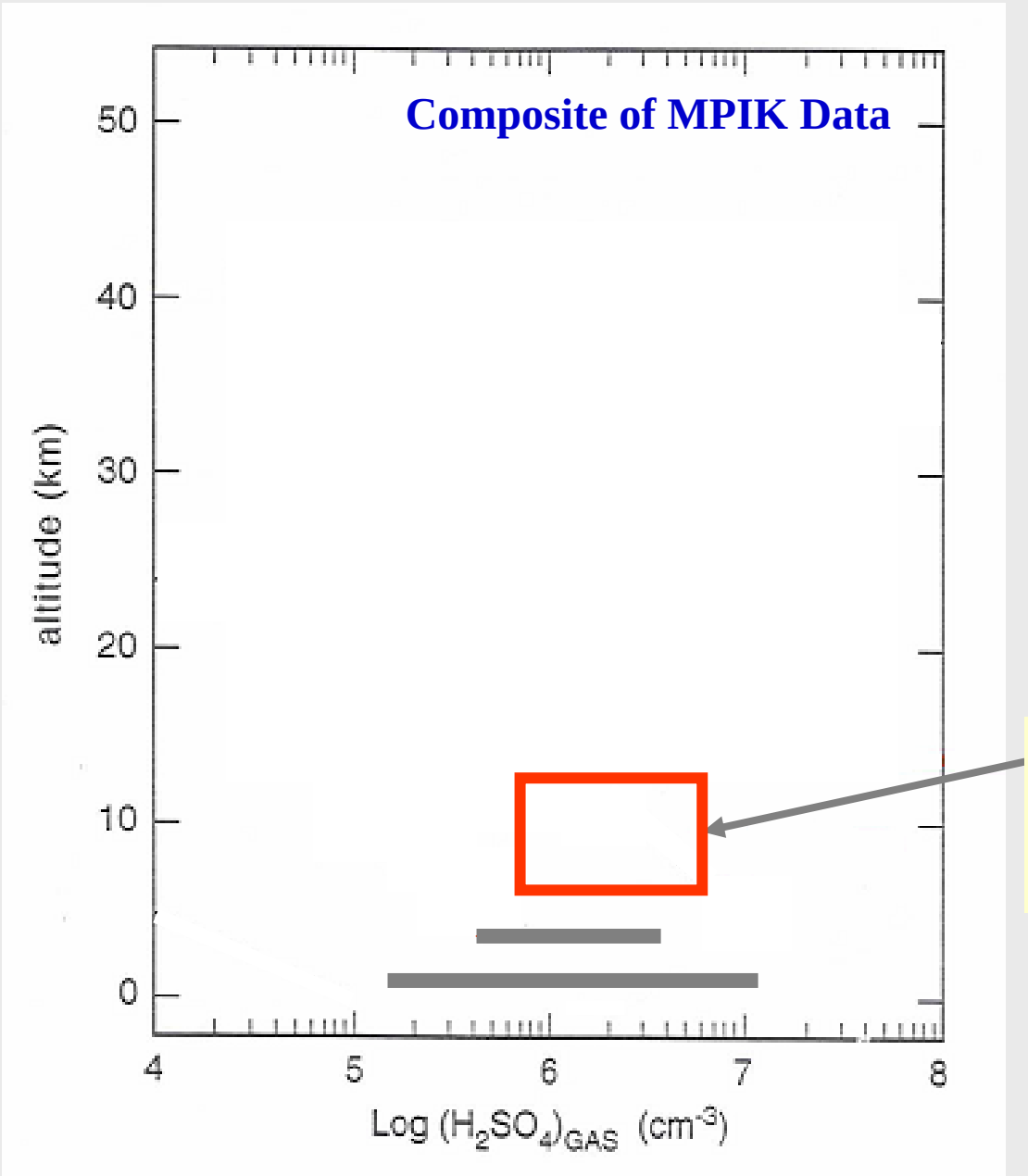

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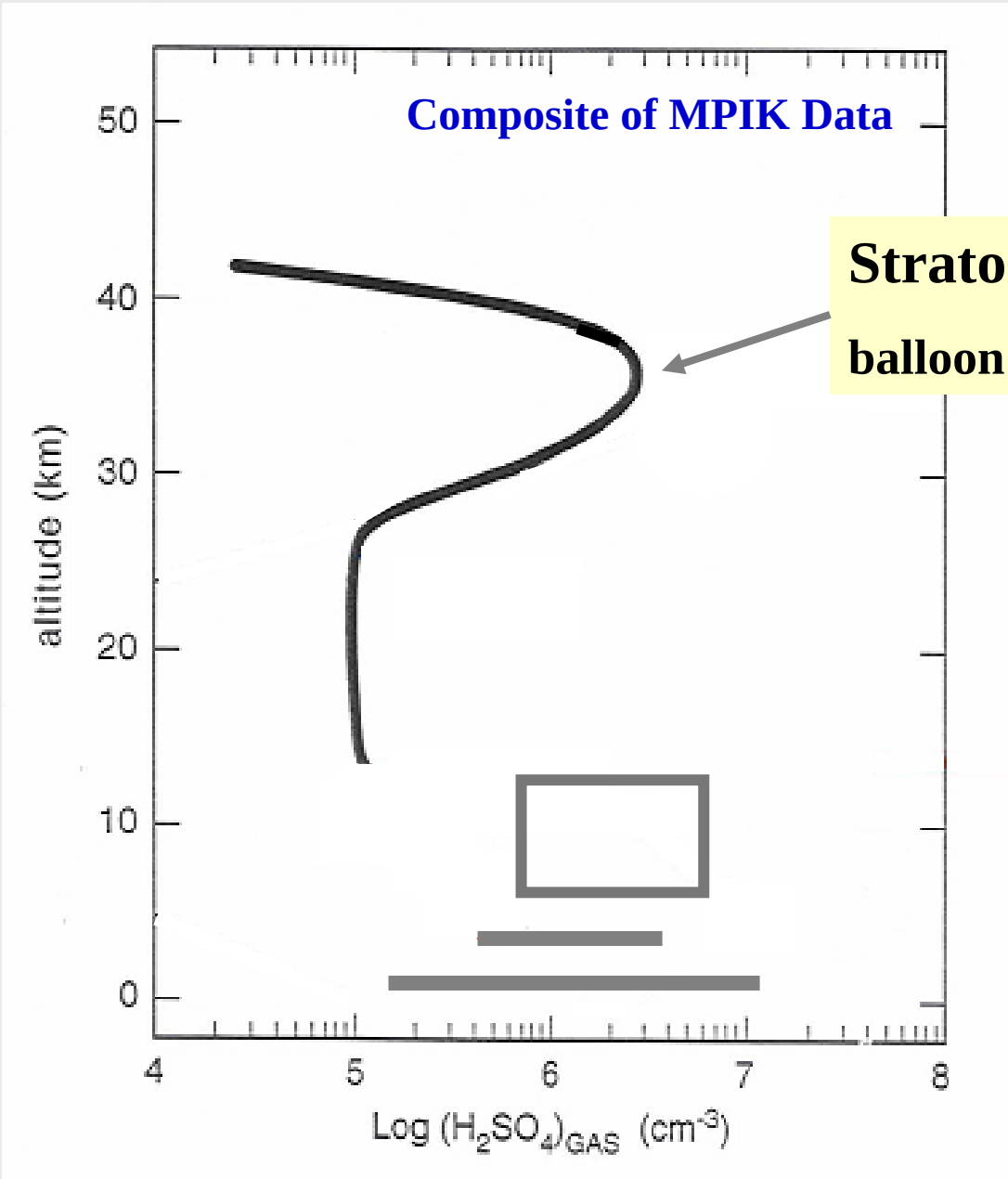

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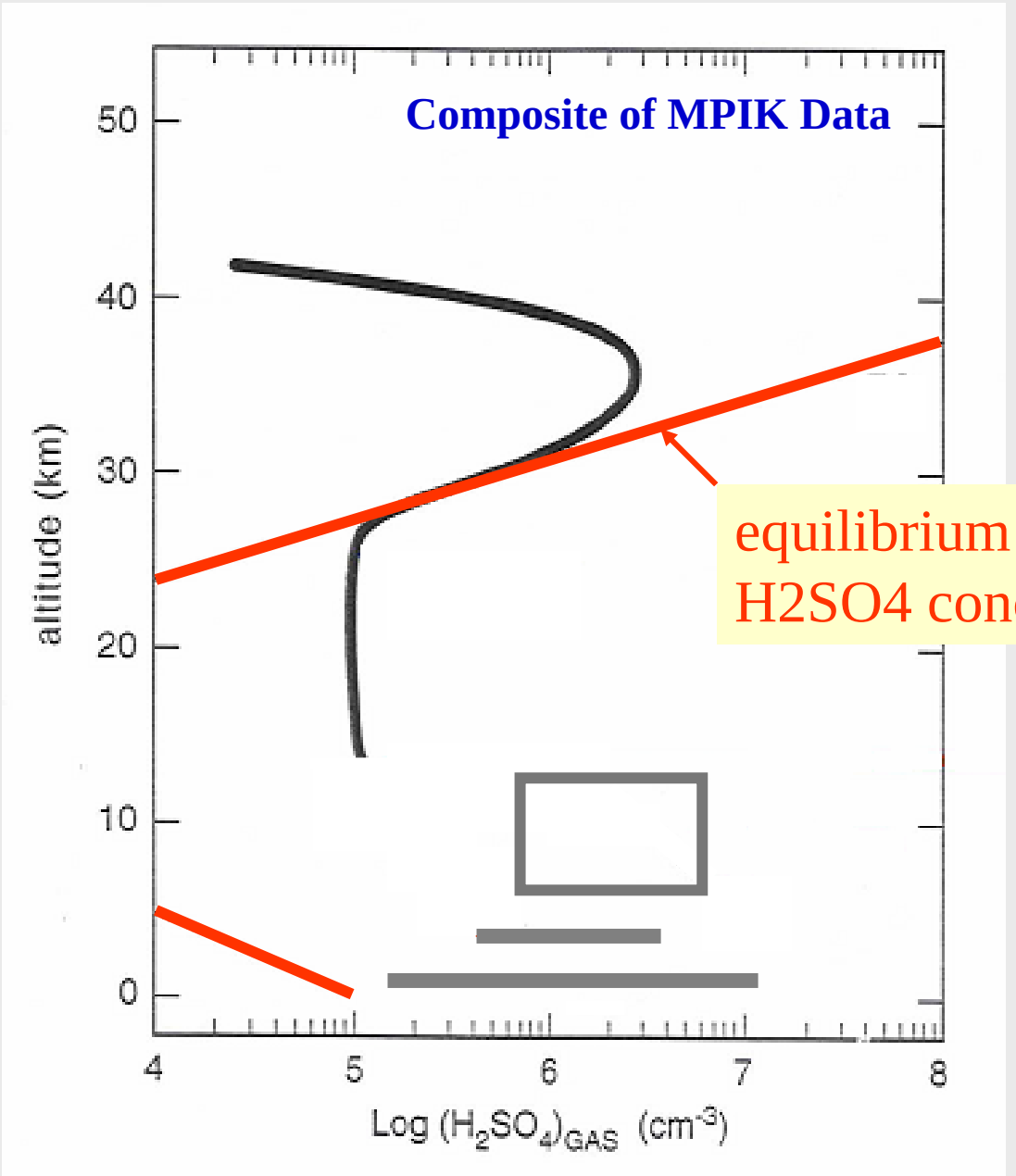

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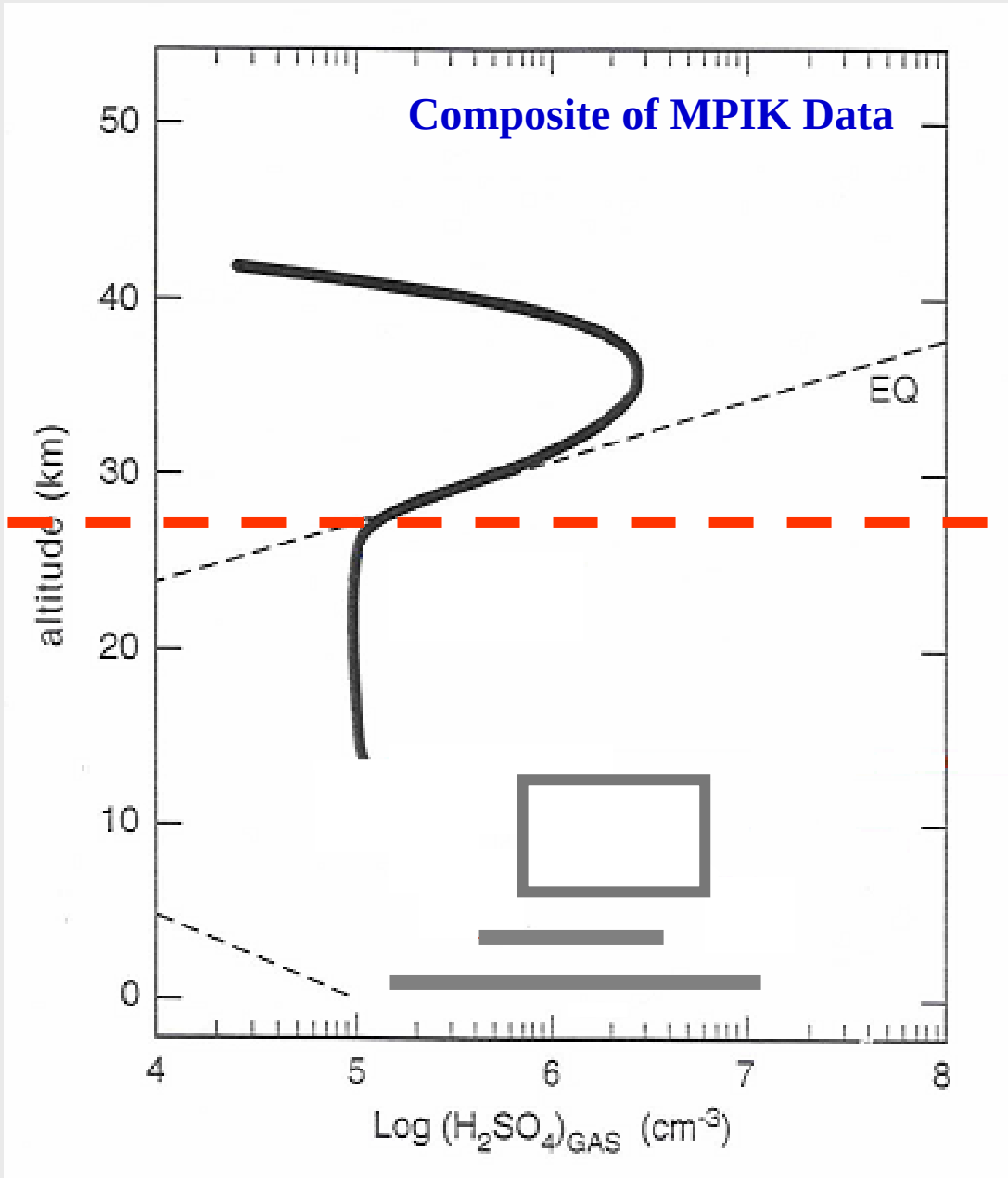

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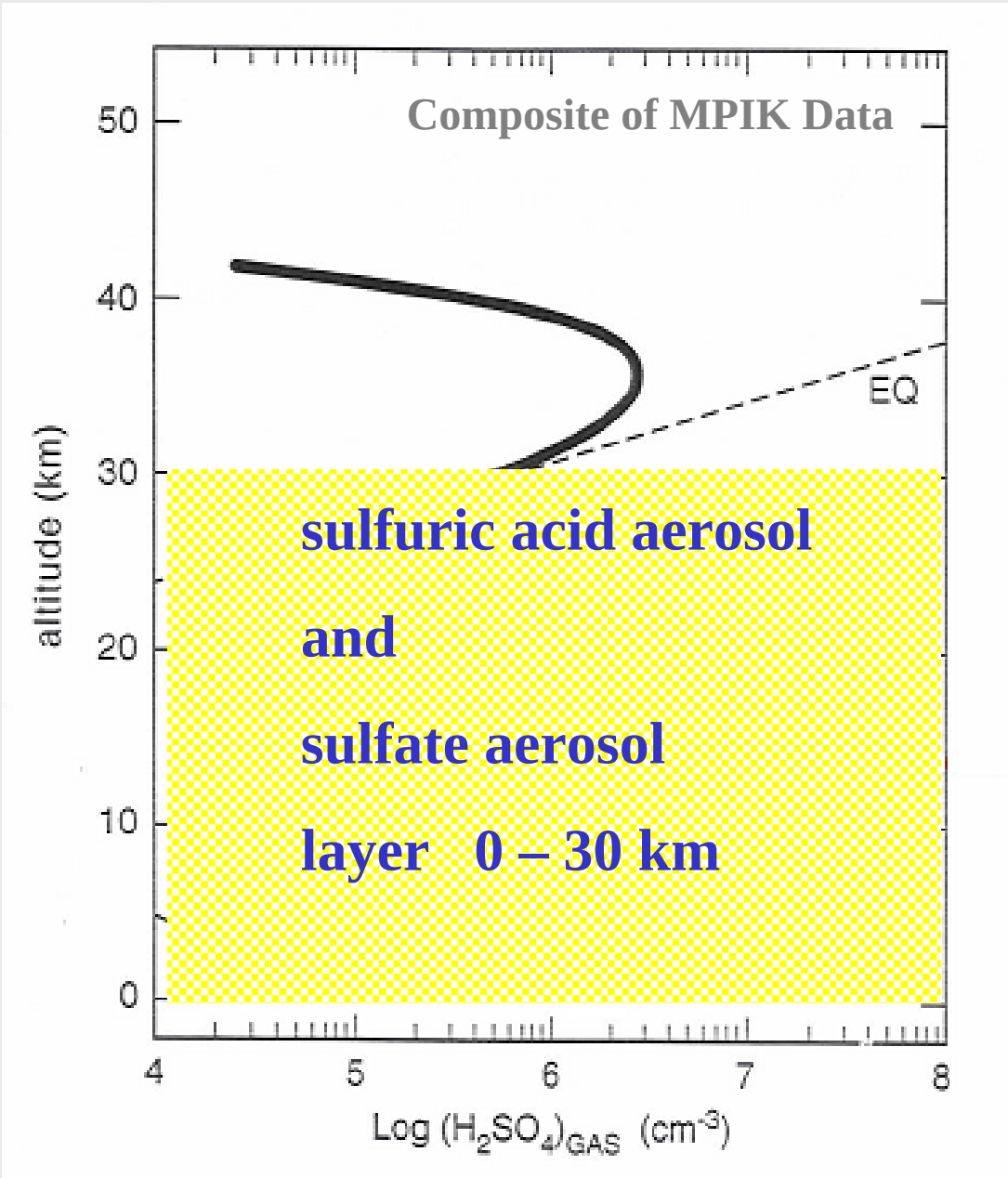

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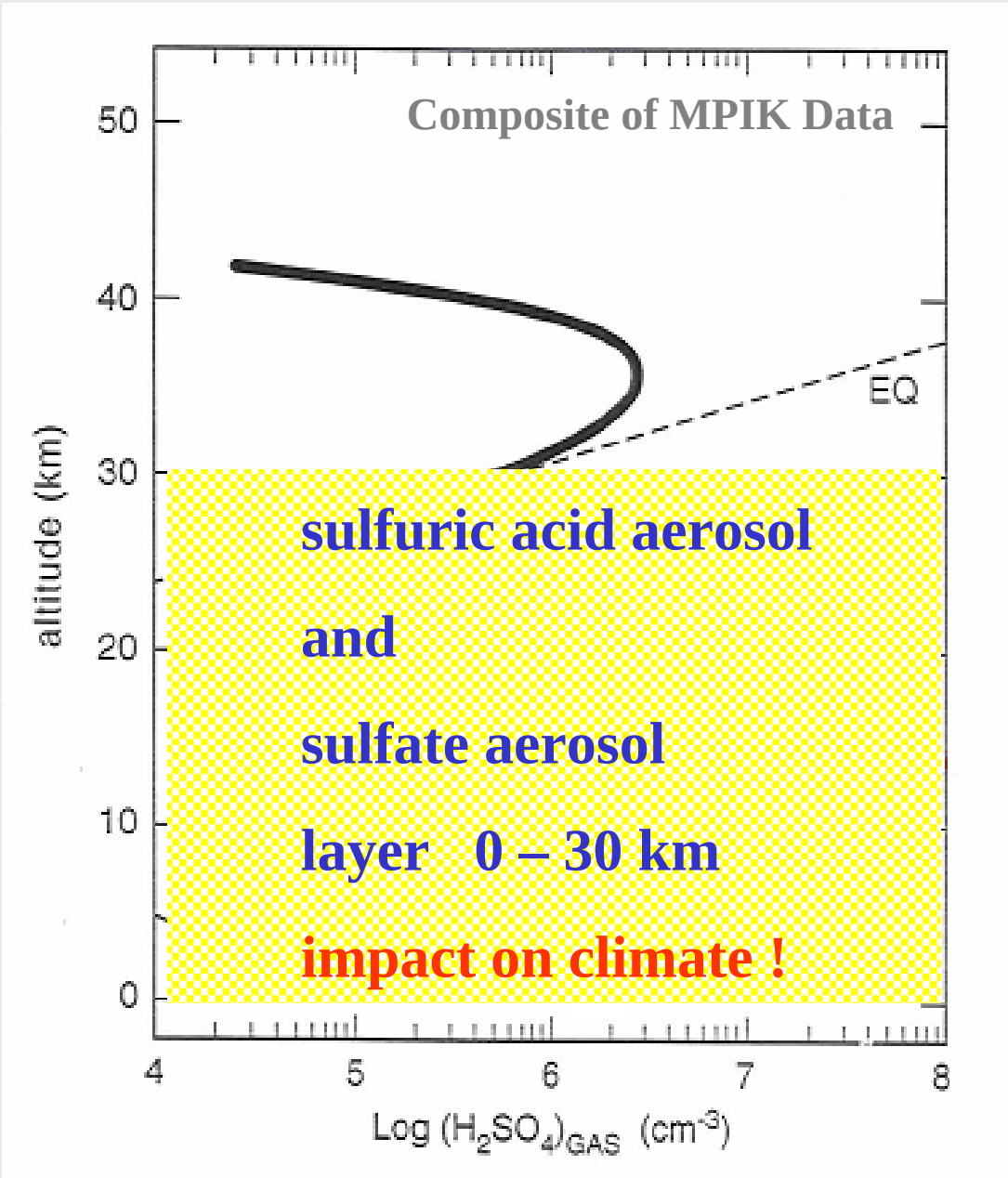

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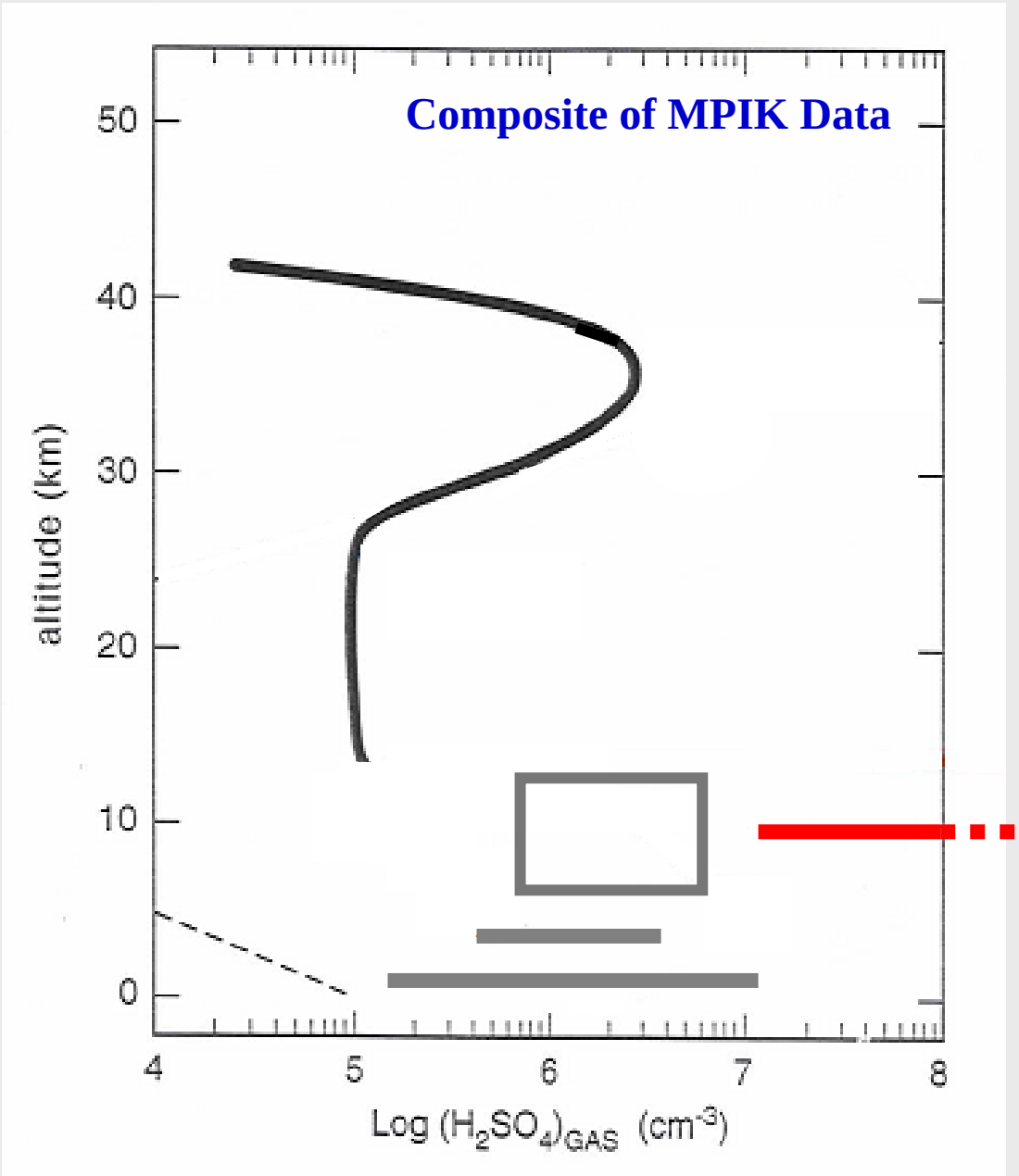

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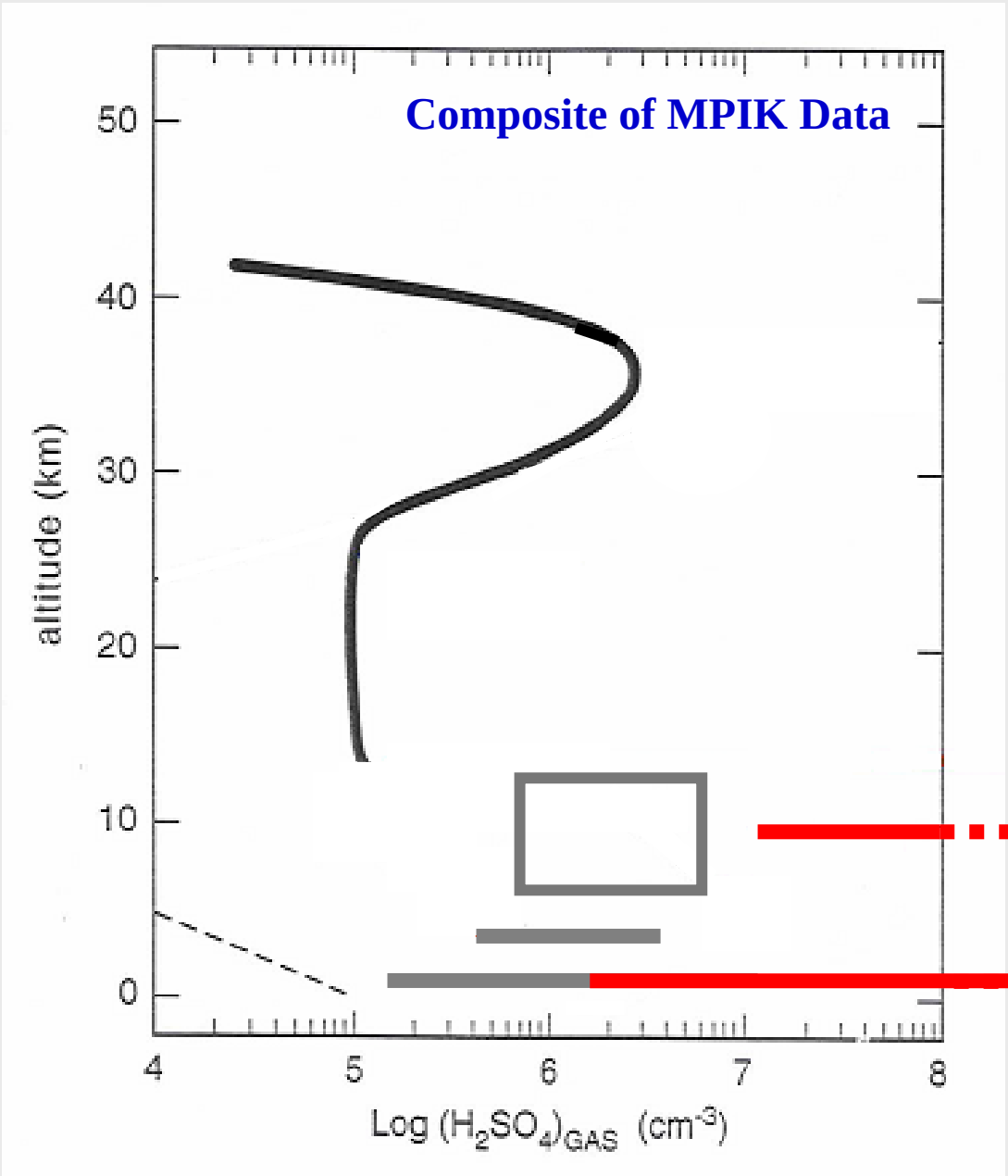


Atmospheric Gaseous Sulfuric Acid Measurements made by MPIK at ground level

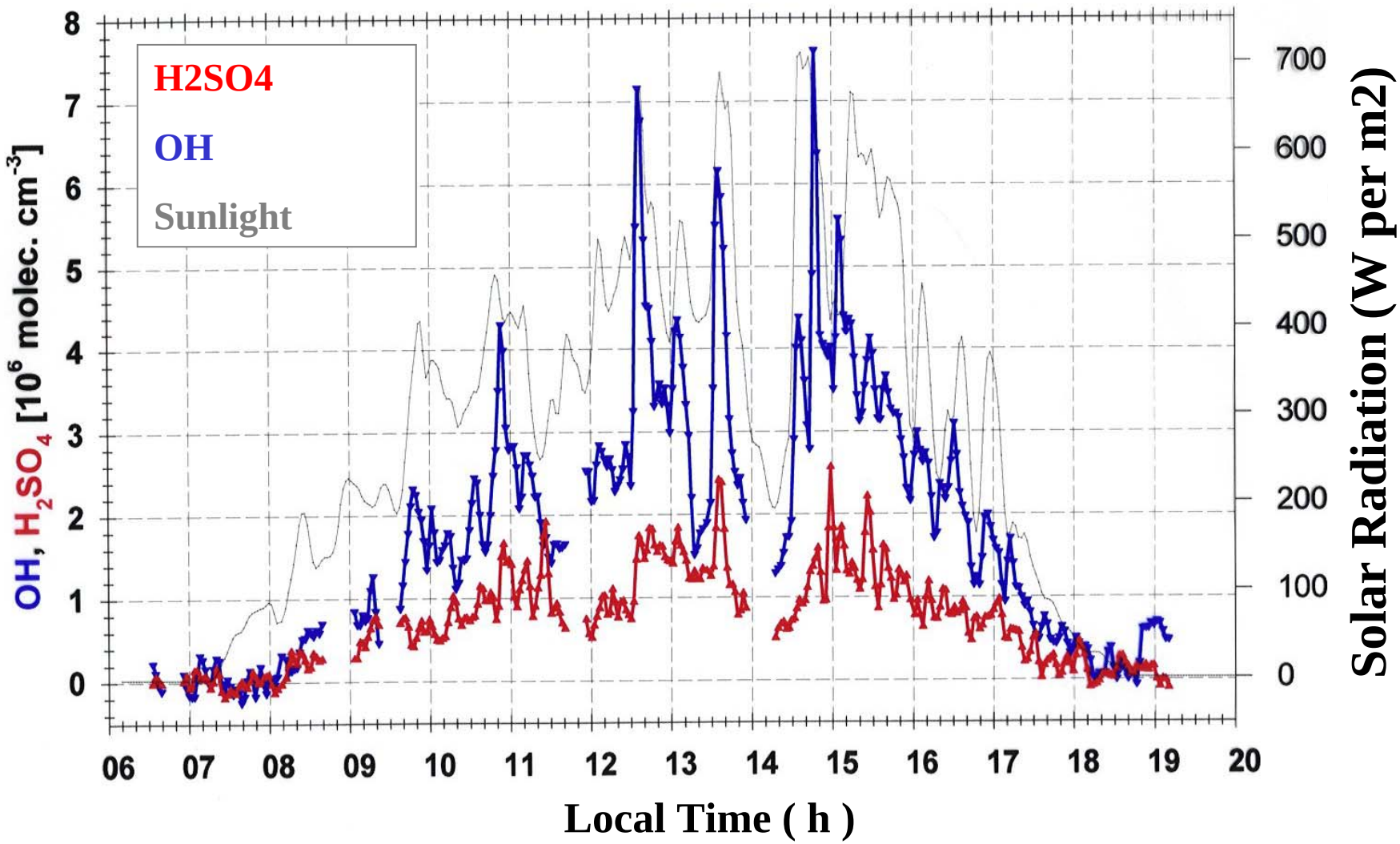
during projecs

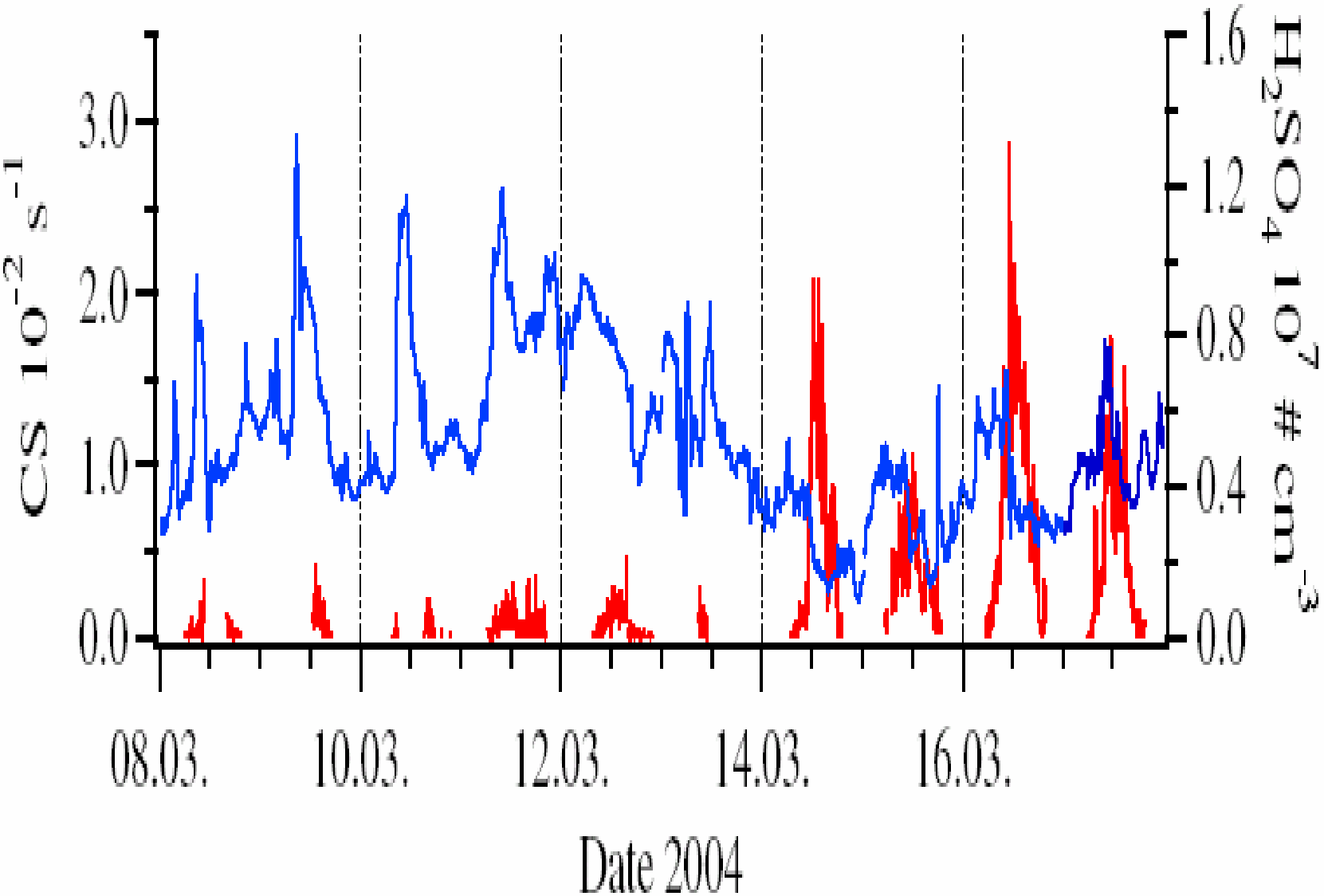
SCAVEX and QUEST

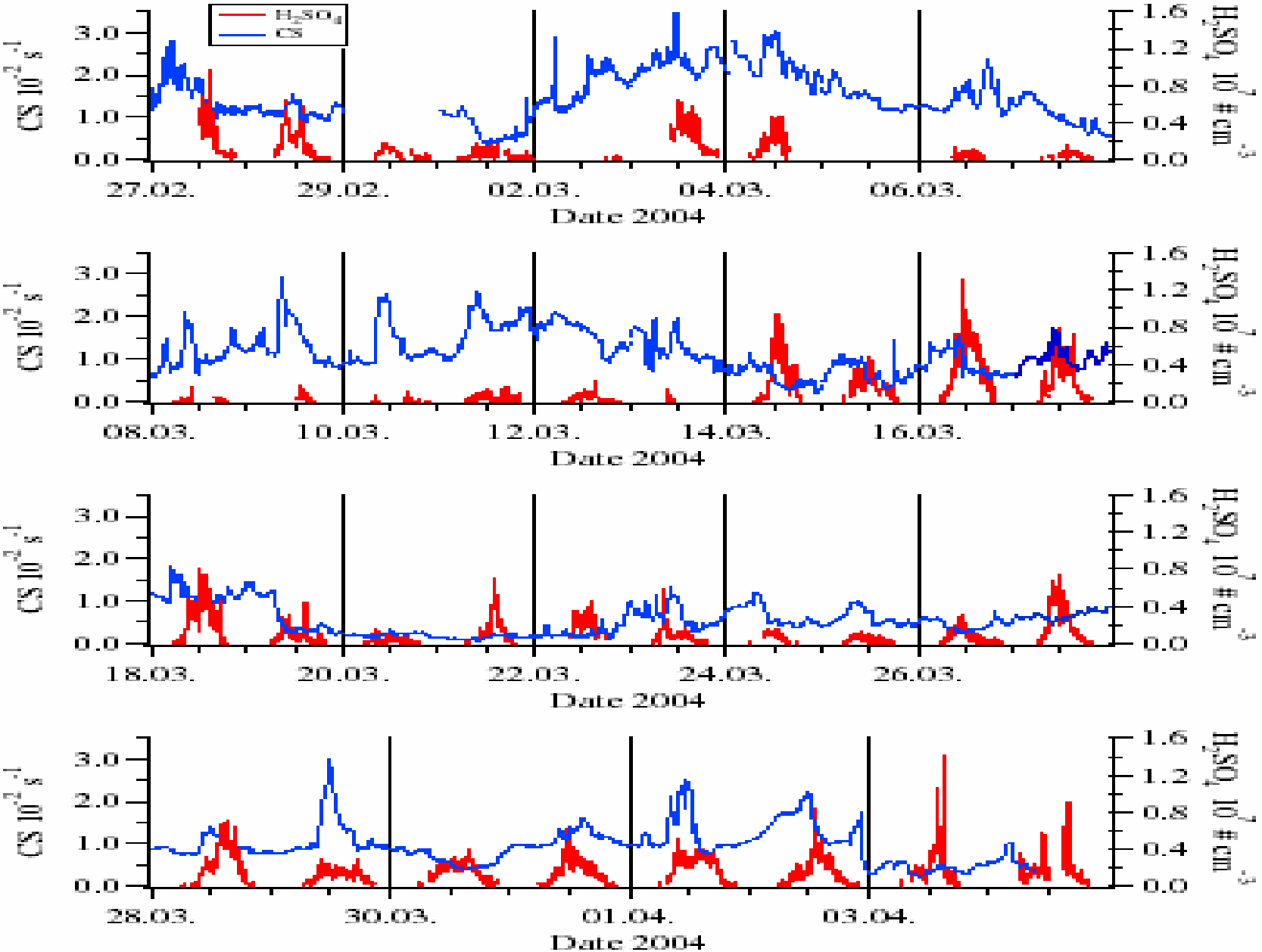
in close collaboration with

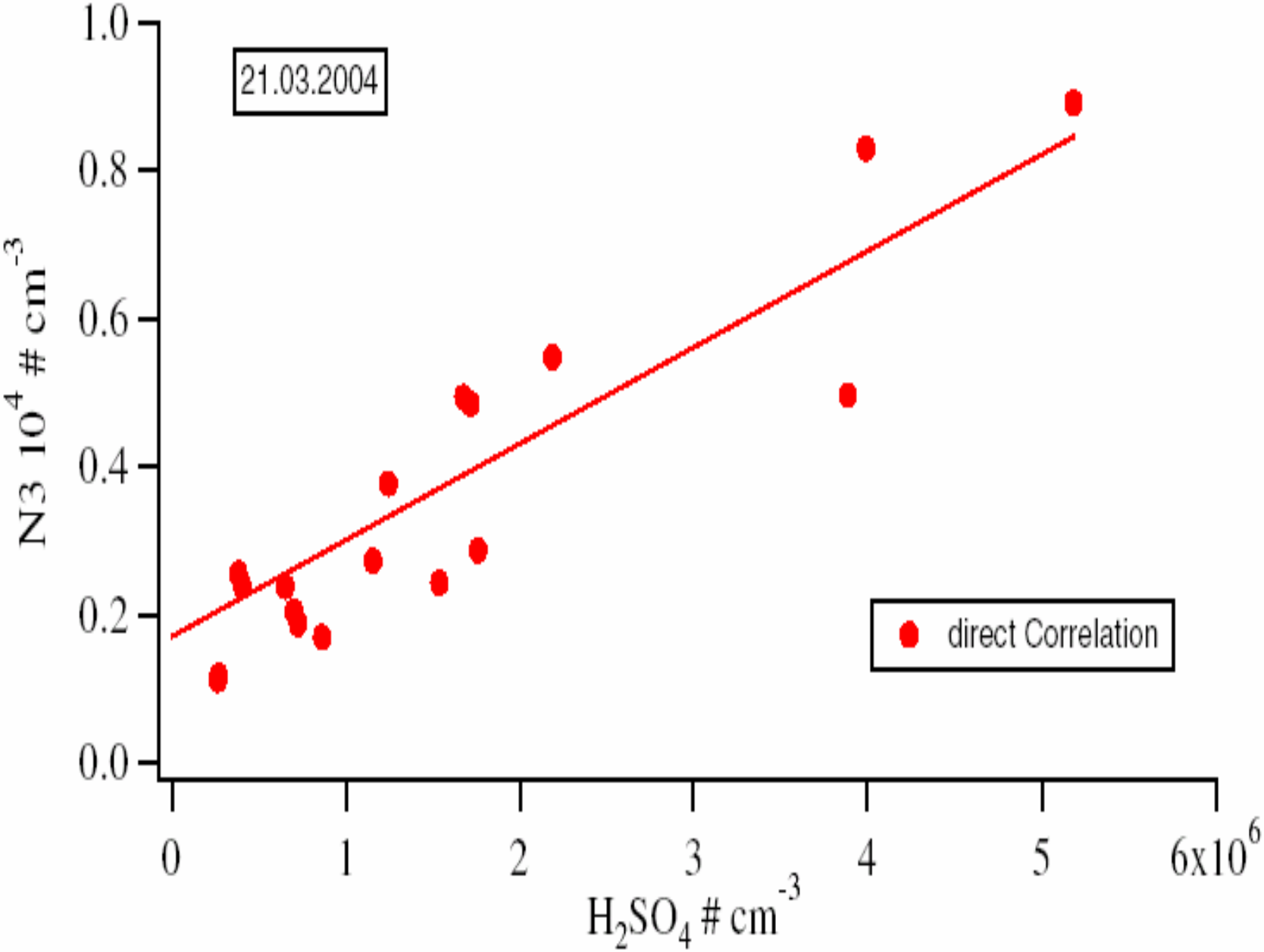
University of Helsinki and DLR

Mount Zugspitze SFH 2300 m altitude




CS 10^{-2} s^{-1}
3.0
2.0
1.0
0.0
1.6
1.2
0.8
0.4
0.0
H$_2$SO$_4$ 10^7 # cm^{-3}
08.03.
10.03.
12.03.
14.03.
16.03.
Date 2004


H2SO4
CS
CS 10-2 s-1
H2SO4 10^7 # cm-3
0.0
1.0
2.0
3.0
0.4
0.8
1.2
1.6
27.02.
29.02.
02.03.
04.03.
06.03.
Date 2004
08.03.
10.03.
12.03.
14.03.
16.03.
Date 2004
18.03.
20.03.
22.03.
24.03.
26.03.
Date 2004
28.03.
30.03.
01.04.
03.04.
Date 2004


21.03.2004
direct Correlation
N3 10^4 # cm^-3
H2SO4 # cm^-3
1.0
0.8
0.6
0.4
0.2
0.0
0
1
2
3
4
5
6x10^6

Conclusions

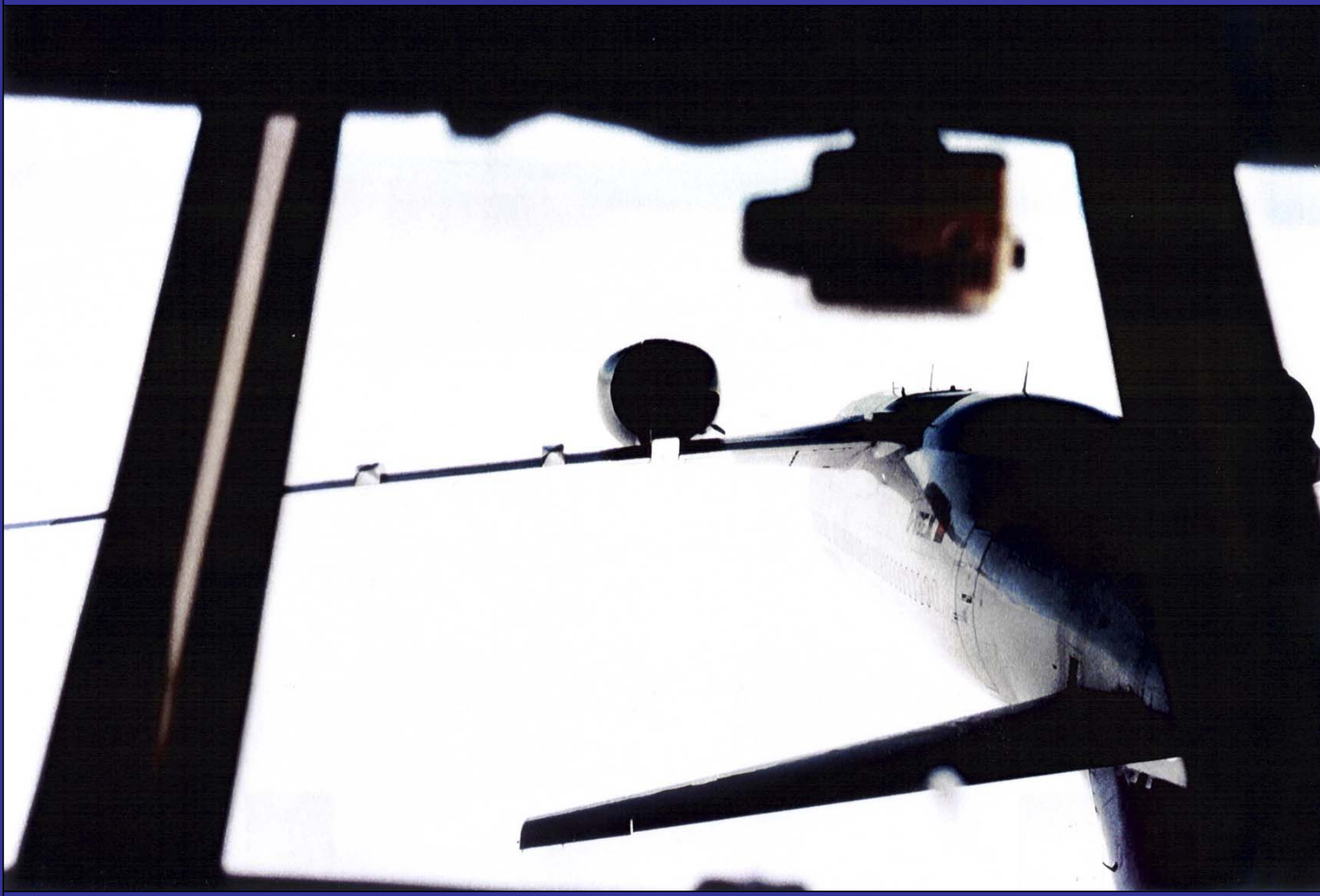
- particle formation triggered by H2SO4
- only about 5% of particle growth is due to H2SO4/H2O condensation
- particle growth probably due to condensable organics
- see our recent papers
  Fiedler et al (2005)
  Boy et al (2005)

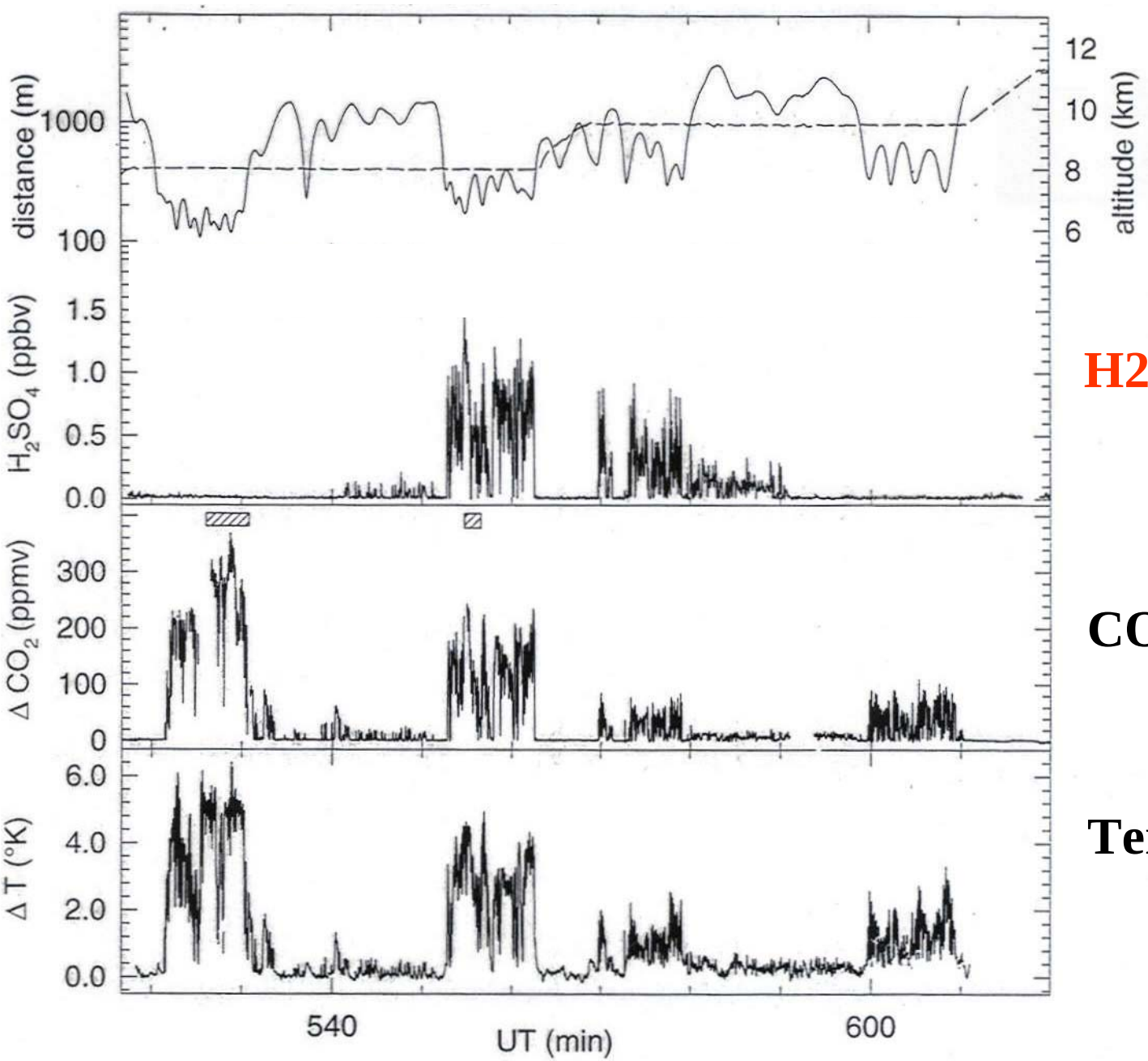
Atmospheric Sulfuric Acid Measurements made by MPIK in Aircraft Exhaust

measurements made in close collaboration with DLR


D-CMET


F-GMGB
F-GALP
GEHLMAX

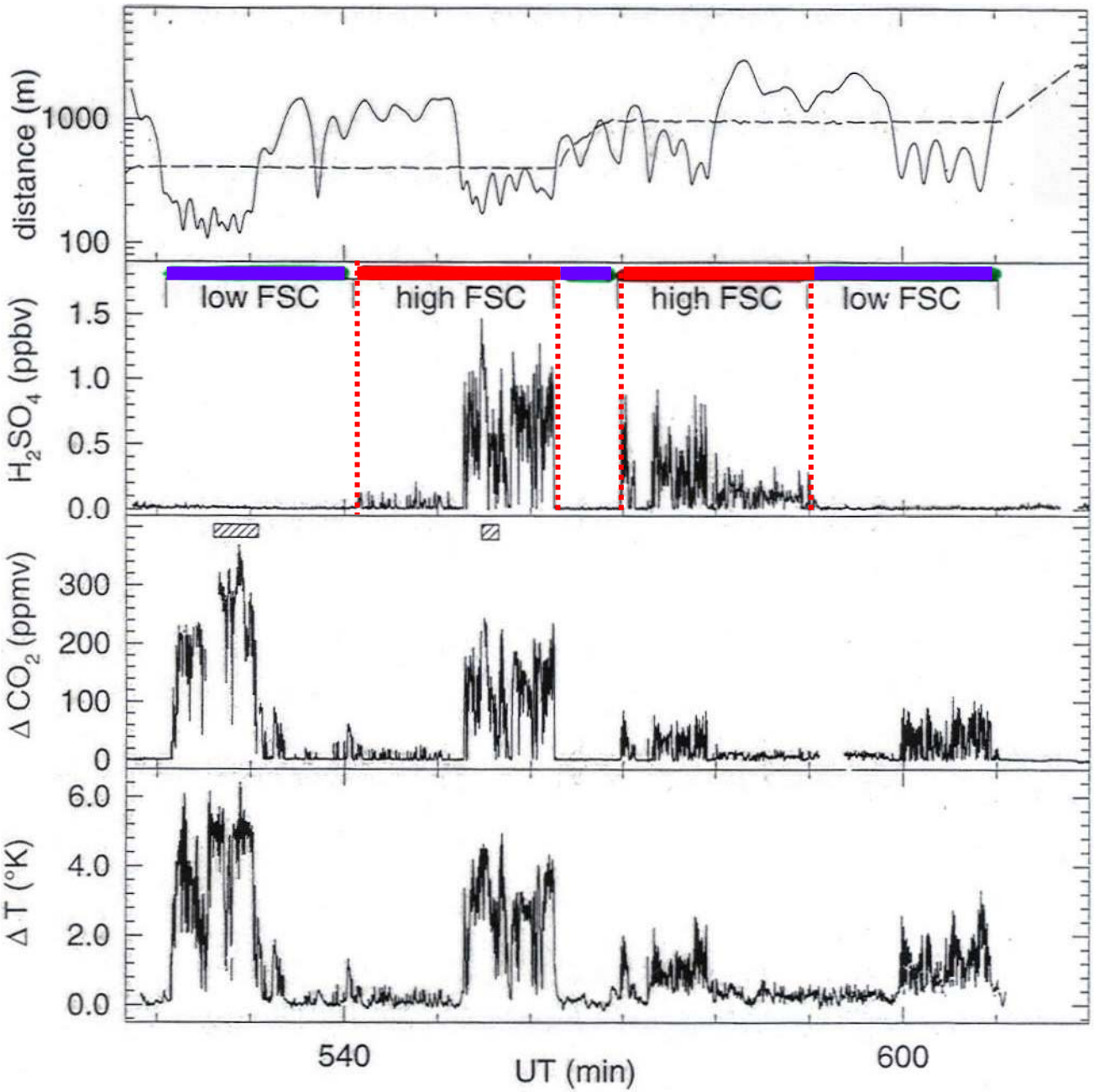




H2SO4

CO2

Temp



H2SO4

CO2

Temp

EMISSION INDEX (mg / kg)

for modern engine and FSC=400 (100-3000) ppmM

H2SO4 - CONDENSATE	73 (18 - 550)
SOOT	10
COND. HC.	10

Conclusions

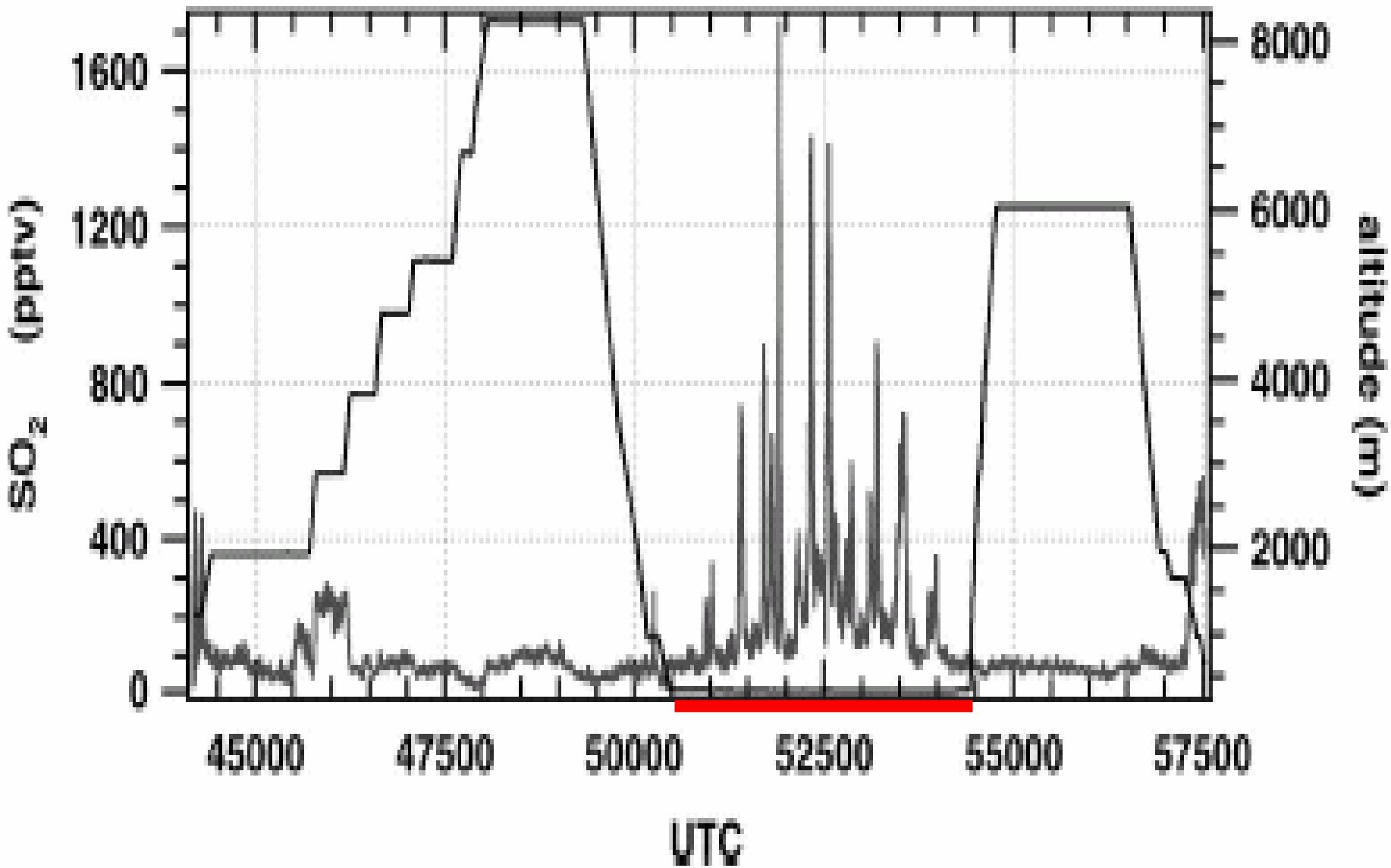
- about 2 - 4 % of fuel sulfur undergoes conversion to gaseous H2SO4
- most of the aerosol mass in an aircraft exhaust plume is due to H2SO4/H2O
- see our publications (following slide)

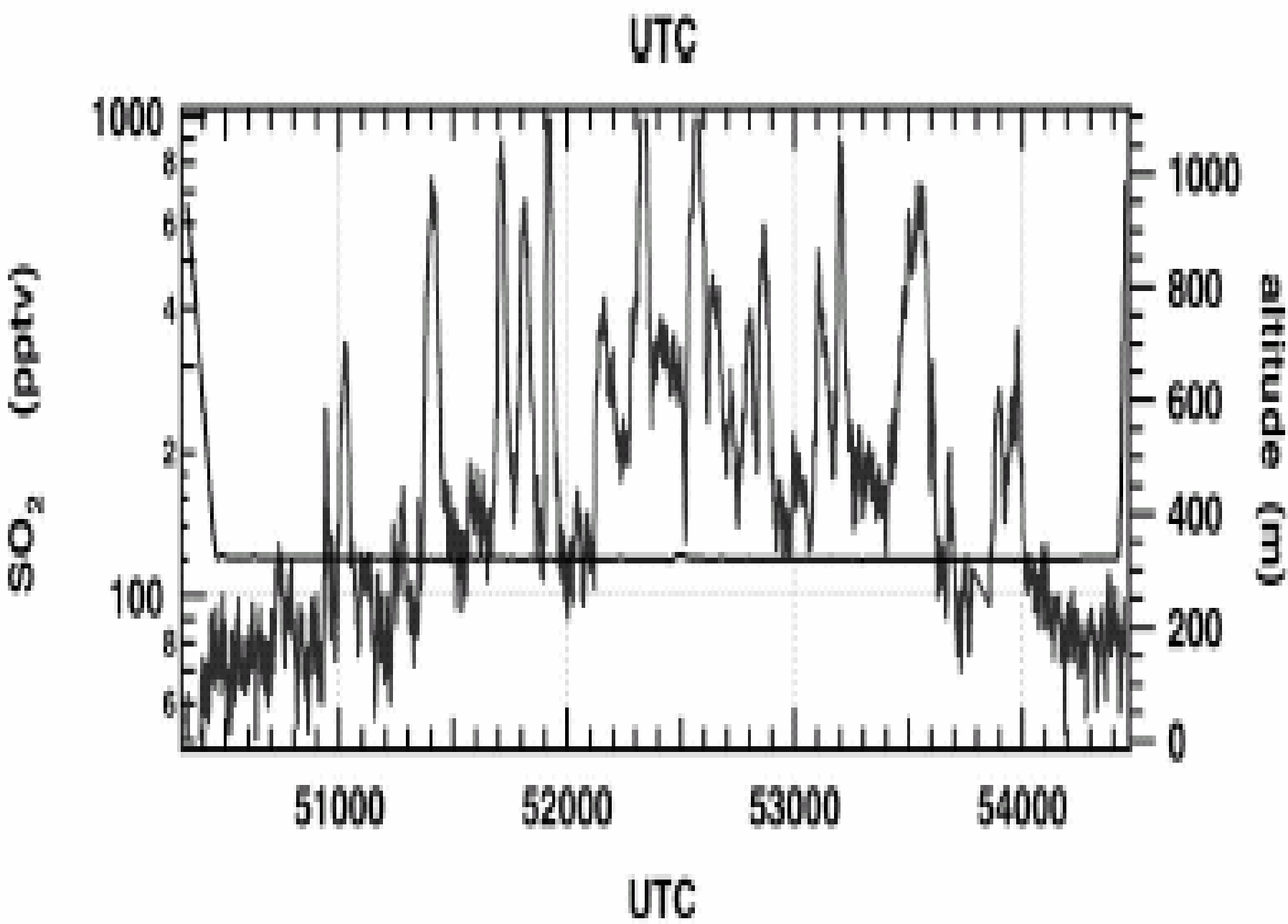
SO2 Measurements made by MPIK in Oceanship Exhaust

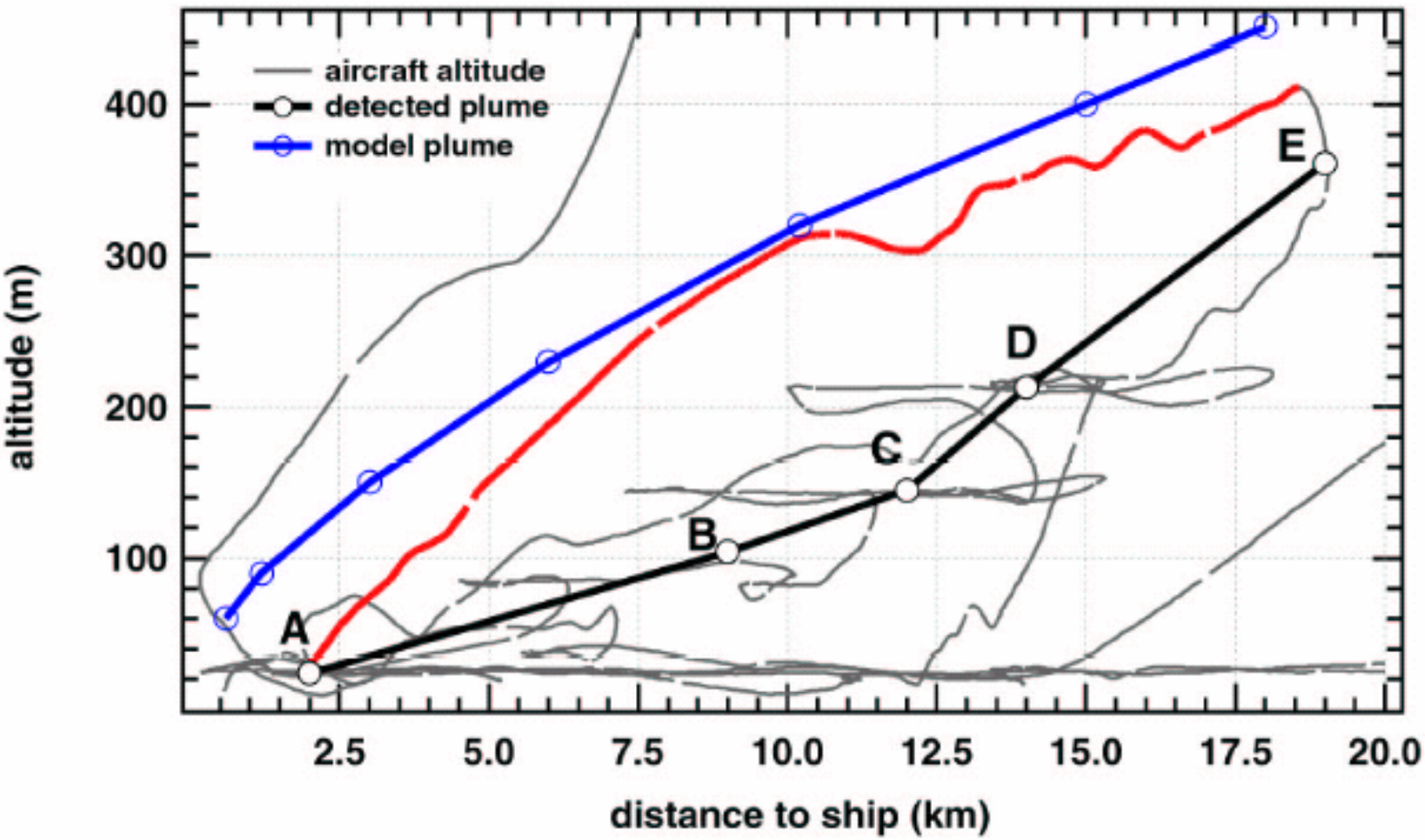
during project ITOP

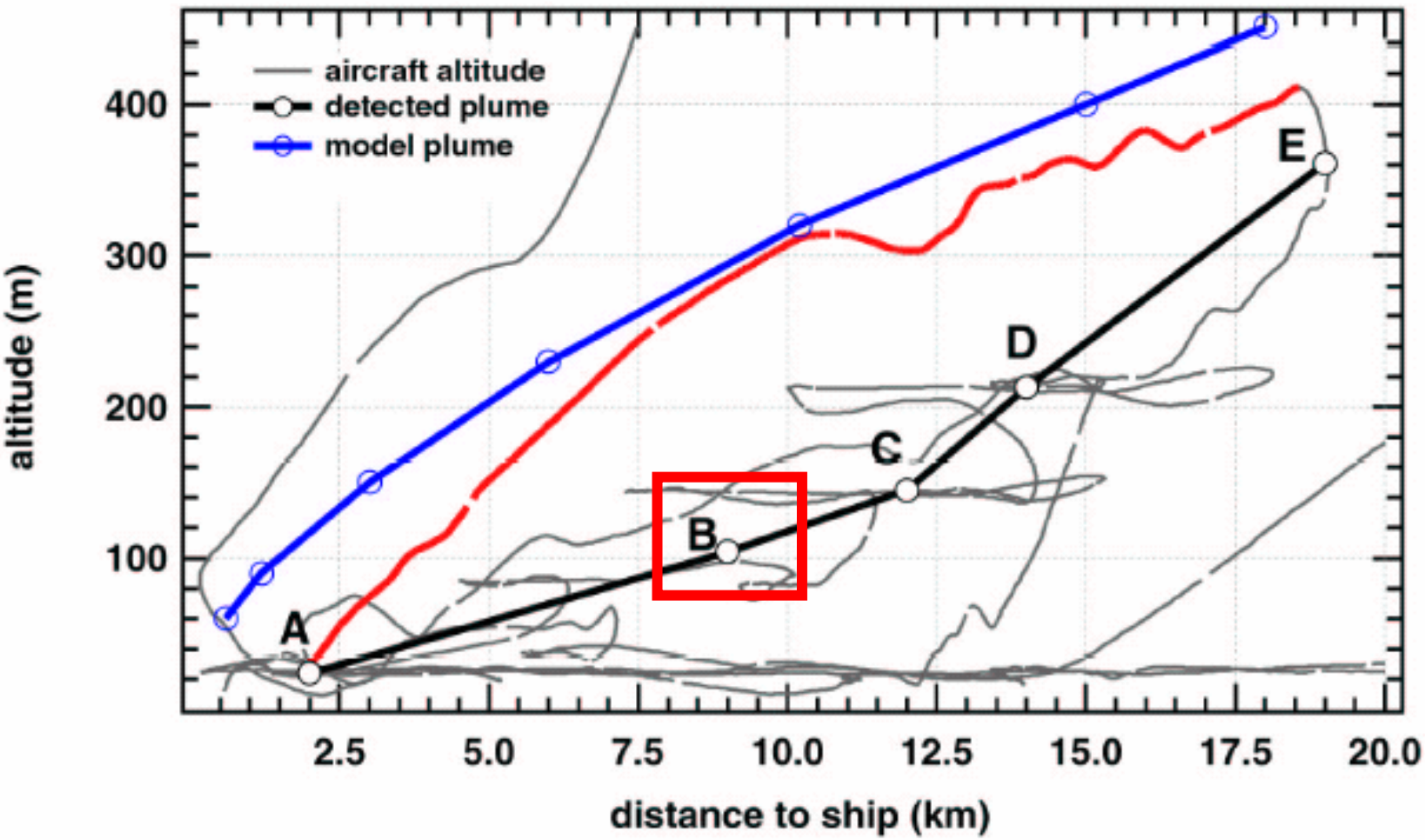
(in close collaboration with DLR)

Ship Tracks


1600
1200
800
400
0
SO_2 (pptv)
8000
6000
4000
2000
altitude (m)
45000
47500
50000
52500
55000
57500
UTC


UTC
1000
8
6
4
2
100
8
6
SO_2 (pptv)
1000
800
600
400
200
0
altitude (m)
51000
52000
53000
54000
UTC


aircraft altitude
detected plume
model plume
altitude (m)
400
300
200
100
A
B
C
D
E
2.5
5.0
7.5
10.0
12.5
15.0
17.5
20.0
distance to ship (km)


aircraft altitude
detected plume
model plume
altitude (m)
400
300
200
100
A
B
C
D
E
2.5
5.0
7.5
10.0
12.5
15.0
17.5
20.0
distance to ship (km)

Conclusions

- strong SO2 pollution in marine boundary layer near ship traffic ways
- sounding individual ship tracks is feasible
- next step H2SO4 and OH measurements in marine boundary layer and ship exhaust

Gaseous Sulfuric Acid Measurements made by MPIK in Automobile Exhaust

in close collaboration with

University of Helsinki

Conclusions from automobile measurements

- particle formation is induced by H_2SO_4/H_2O nucleation
- less than 10% of particle growth is due to H_2SO_4/H_2Ocondensation
- particle growth is probably mostly due to condensable organics

Acknowledgements

- DLR
- University of Helsinki
- IFT Leipzig
- Members of our MPIK-Heidelberg group

MEMBERS OF MPIK HEIDELBERG GROUP

(2000-2005 , **2004-2005 only**)

SCIENTISTS

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Dr.S.Eichkorn, Dr.J.Curtius

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H.Aufmhoff, B.Umann, E. Katragkou, S.Wilhelm,J.Ücker, S.Eichkorn,
M.Hanke, A.Kiendler,

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