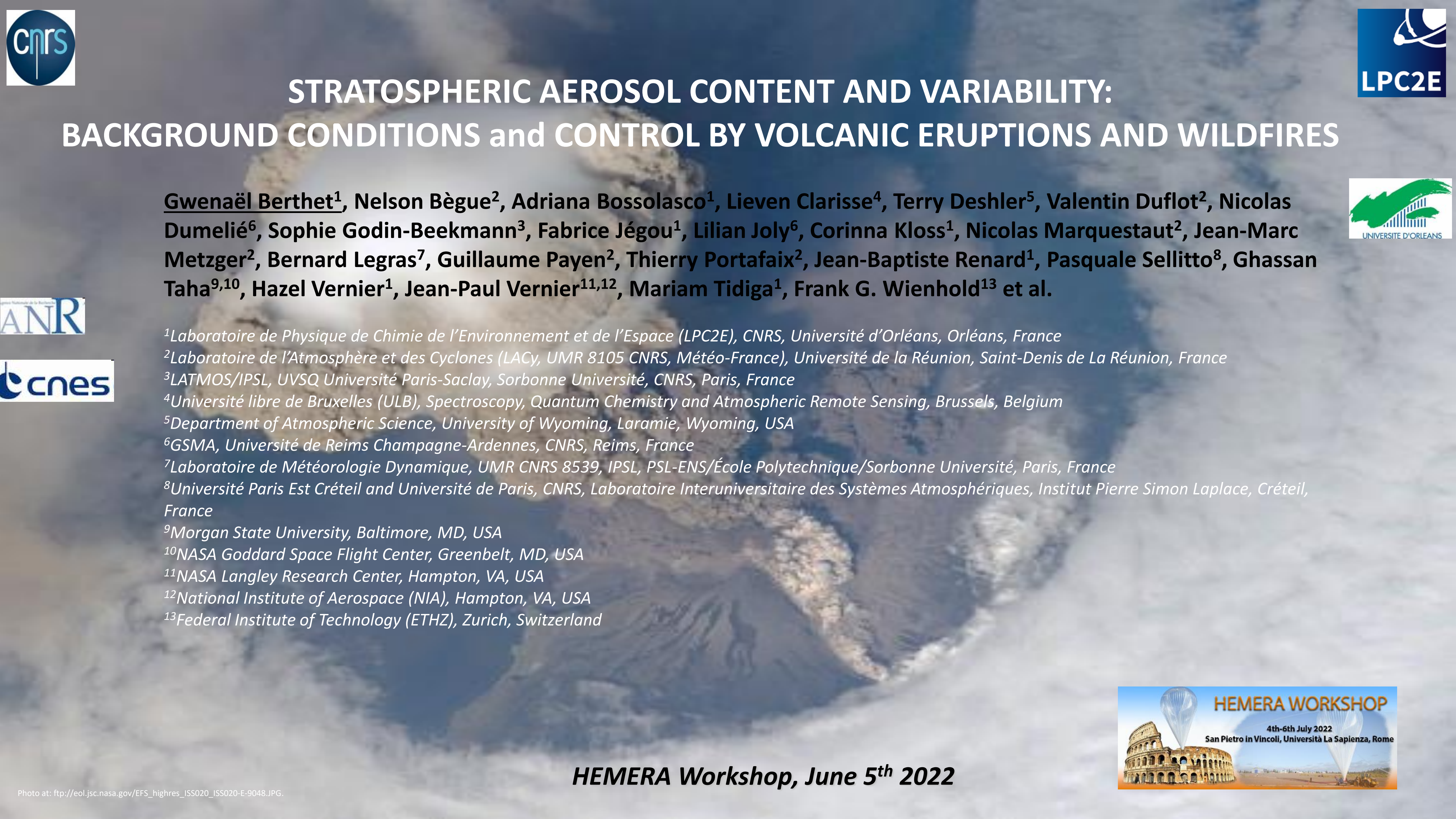


STRATOSPHERIC AEROSOL CONTENT AND VARIABILITY: BACKGROUND CONDITIONS and CONTROL BY VOLCANIC ERUPTIONS AND WILDFIRES

Gwenaël Berthet[1], Nelson Bègue[2], Adriana Bossolasco[1], Lieven Clarisse[4], Terry Deshler[5], Valentin Duflot[2], Nicolas Dumelié[6], Sophie Godin-Beekmann[3], Fabrice Jégou[1], Lilian Joly[6], Corinna Kloss[1], Nicolas Marquestaut[2], Jean-Marc Metzger[2], Bernard Legras[7], Guillaume Payen[2], Thierry Portafaix[2], Jean-Baptiste Renard[1], Pasquale Sellitto[8], Ghassan Taha[9,10], Hazel Vernier[1], Jean-Paul Vernier[11,12], Mariam Tidiga[1], Frank G. Wienhold[13] et al.

[1]Laboratoire de Physique de Chimie de l'Environnement et de l'Espace (LPC2E), CNRS, Université d'Orléans, Orléans, France
[2]Laboratoire de l'Atmosphère et des Cyclones (LACy, UMR 8105 CNRS, Météo-France), Université de la Réunion, Saint-Denis de La Réunion, France
[3]LATMOS/IPSL, UVSQ Université Paris-Saclay, Sorbonne Université, CNRS, Paris, France
[4]Université libre de Bruxelles (ULB), Spectroscopy, Quantum Chemistry and Atmospheric Remote Sensing, Brussels, Belgium
[5]Department of Atmospheric Science, University of Wyoming, Laramie, Wyoming, USA
[6]GSMA, Université de Reims Champagne-Ardennes, CNRS, Reims, France
[7]Laboratoire de Météorologie Dynamique, UMR CNRS 8539, IPSL, PSL-ENS/École Polytechnique/Sorbonne Université, Paris, France
[8]Université Paris Est Créteil and Université de Paris, CNRS, Laboratoire Interuniversitaire des Systèmes Atmosphériques, Institut Pierre Simon Laplace, Créteil, France
[9]Morgan State University, Baltimore, MD, USA
[10]NASA Goddard Space Flight Center, Greenbelt, MD, USA
[11]NASA Langley Research Center, Hampton, VA, USA
[12]National Institute of Aerospace (NIA), Hampton, VA, USA
[13]Federal Institute of Technology (ETHZ), Zurich, Switzerland

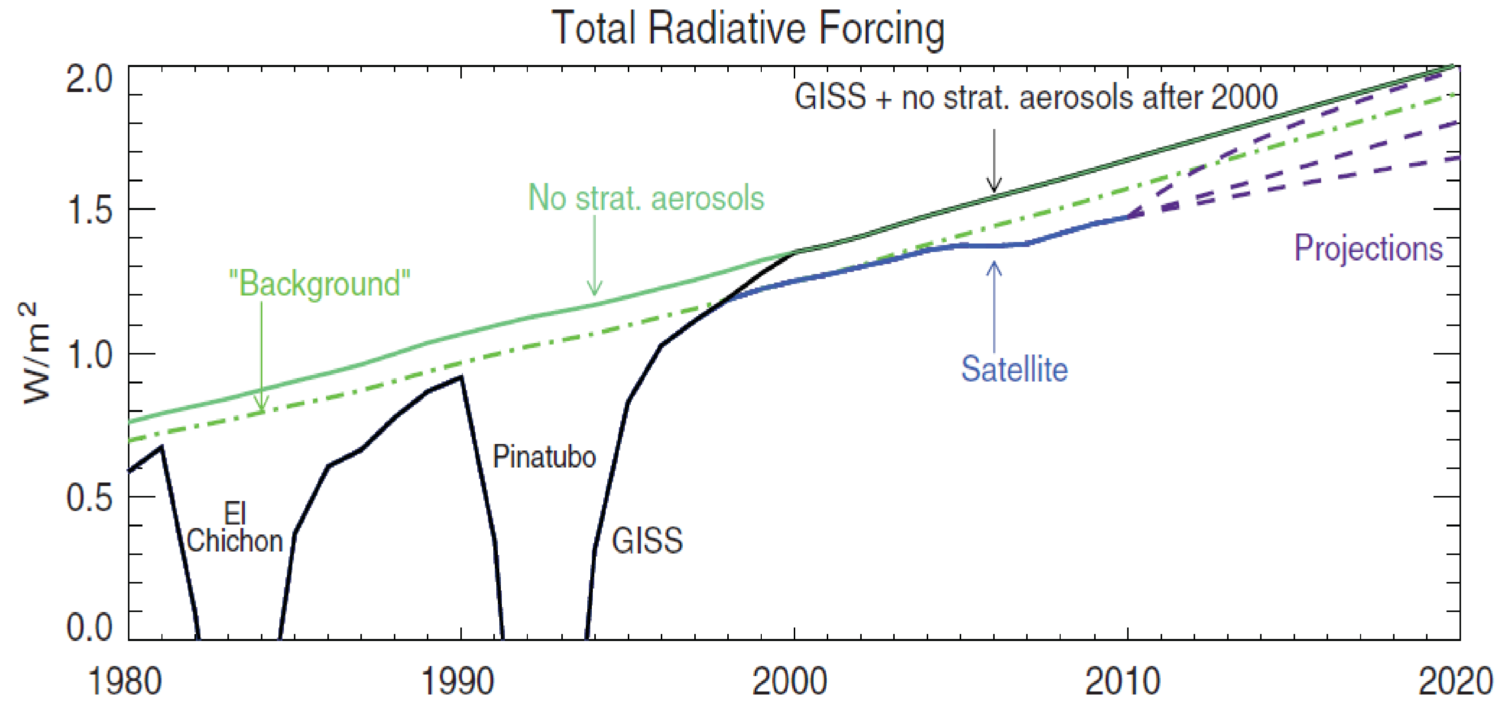
HEMERA WORKSHOP
4th-6th July 2022
San Pietro in Vincoli, Università La Sapienza, Rome

HEMERA Workshop, June 5th 2022

Photo at: ftp://eol.jsc.nasa.gov/EFS_highres_ISS020_ISS020-E-9048.JPG.

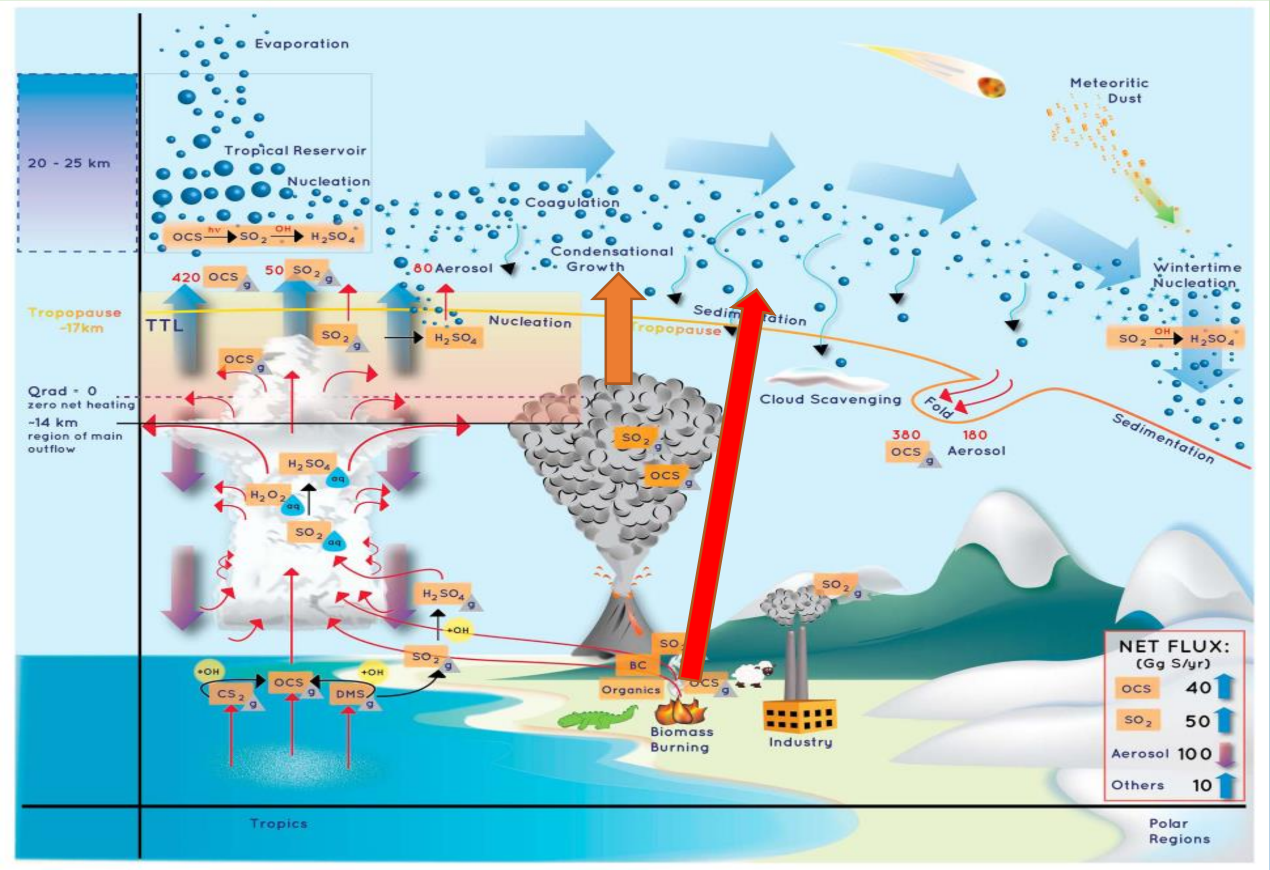
Radiative impact of stratospheric aerosols

Solomon et al., Science, 2011



-0.07°C due to stratospheric aerosols

Cycle of stratospheric aerosols



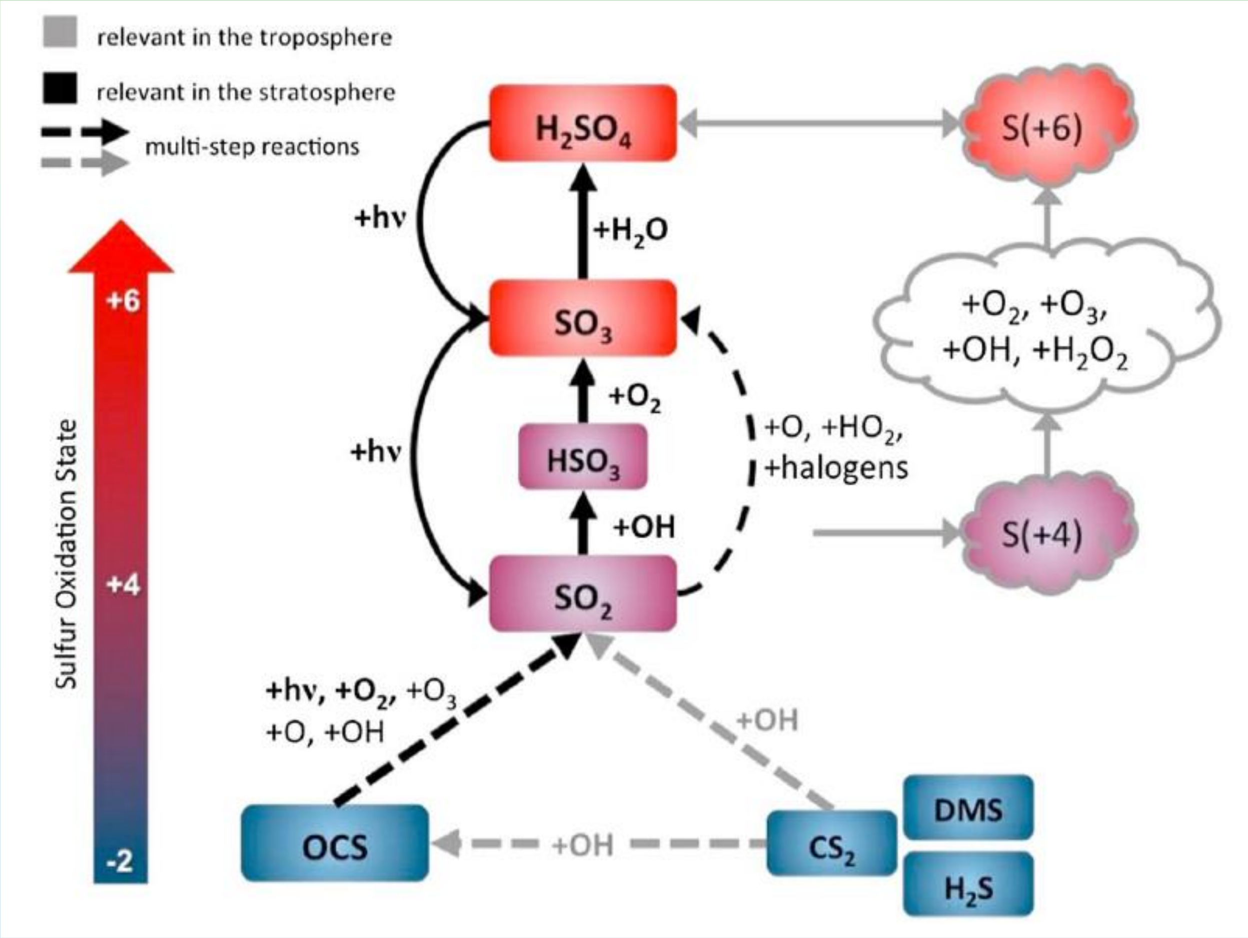
Kremser et al., Rev. of Geophys., 2016

Photolysis + oxydation of OCS in the stratosphere (contribution=56% in *Sheng et al. JGR (2015)* → emitted in the troposphere mainly from oceans, biomass burning (10-20%), wet areas, and anthropogenic activities; Bruhl et al. ACP

Oxydation of SO_2 in the stratosphere (contribution=44% in *Sheng et al. JGR (2015)* → anthropogenic activities and natural emissions (DMS, H_2S, volcanic degassing) with a similar contribution

Contribution of SO_2 from Chinese coal not significant (*Neely et al. GRL, 2013; Sheng et al. JGR, 2015*)

Sulfur oxidation



Kremser et al., Rev. of Geophys., 2016

OCS + hν → CO + S (λ ≤ 300 nm)

OCS + OH → CO2 + HS
OCS + O → CO + SO

Very important role of sulfate aerosols on stratospheric ozone chemistry

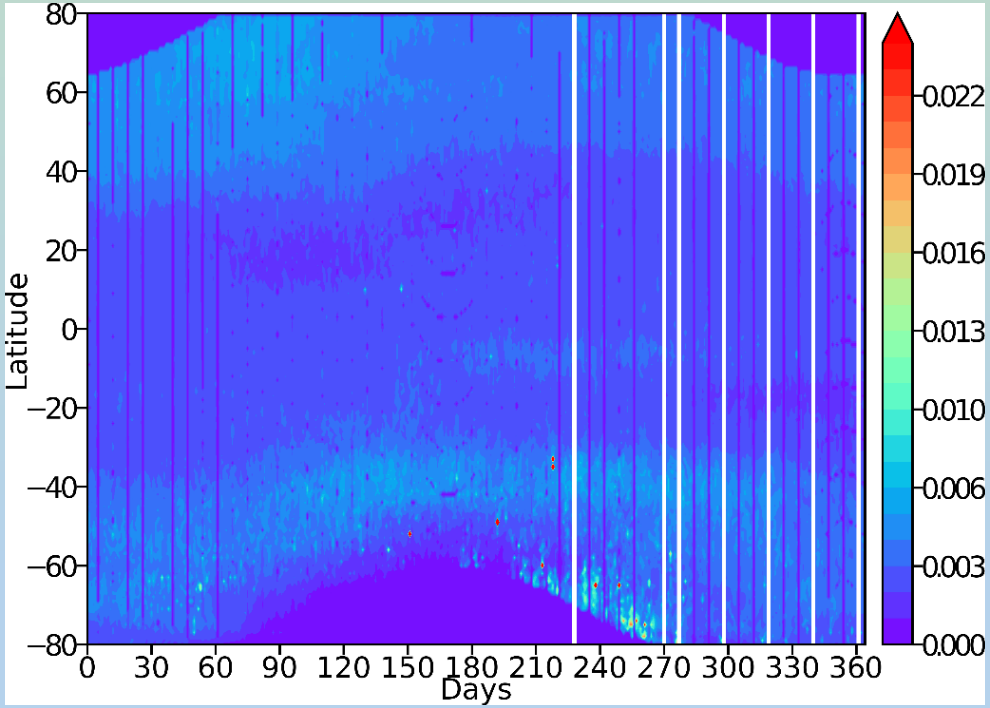
$\mathbf{N_2O_5 + H_2O \rightarrow 2HNO_3}$
$\mathbf{ClONO_2 + H_2O \rightarrow HOCl + HNO_3}$ **(low T)** **…**

=> Reduction of NOx and of their ozone loss efficiency in the middle stratosphere
=> Increase of ozone loss by ClOx, BrOx et HOx in the lower stratosphere

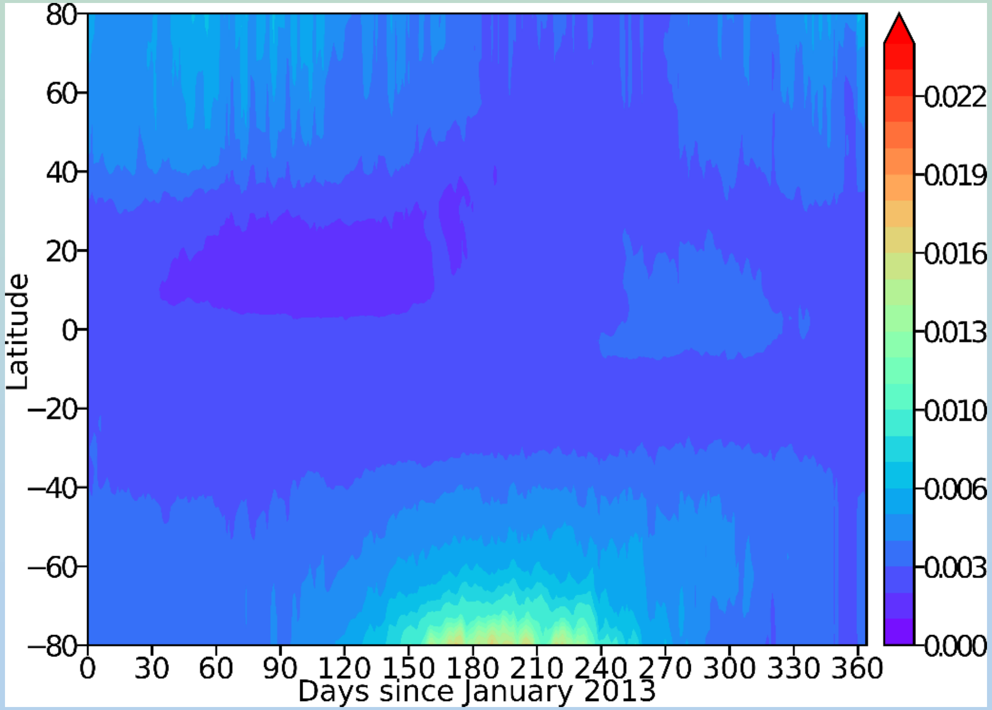
« Background » stratospheric aerosol

Latitudinal distribution of Aerosol Optical Depth (AOD)

OMPS, year 2013
675 nm

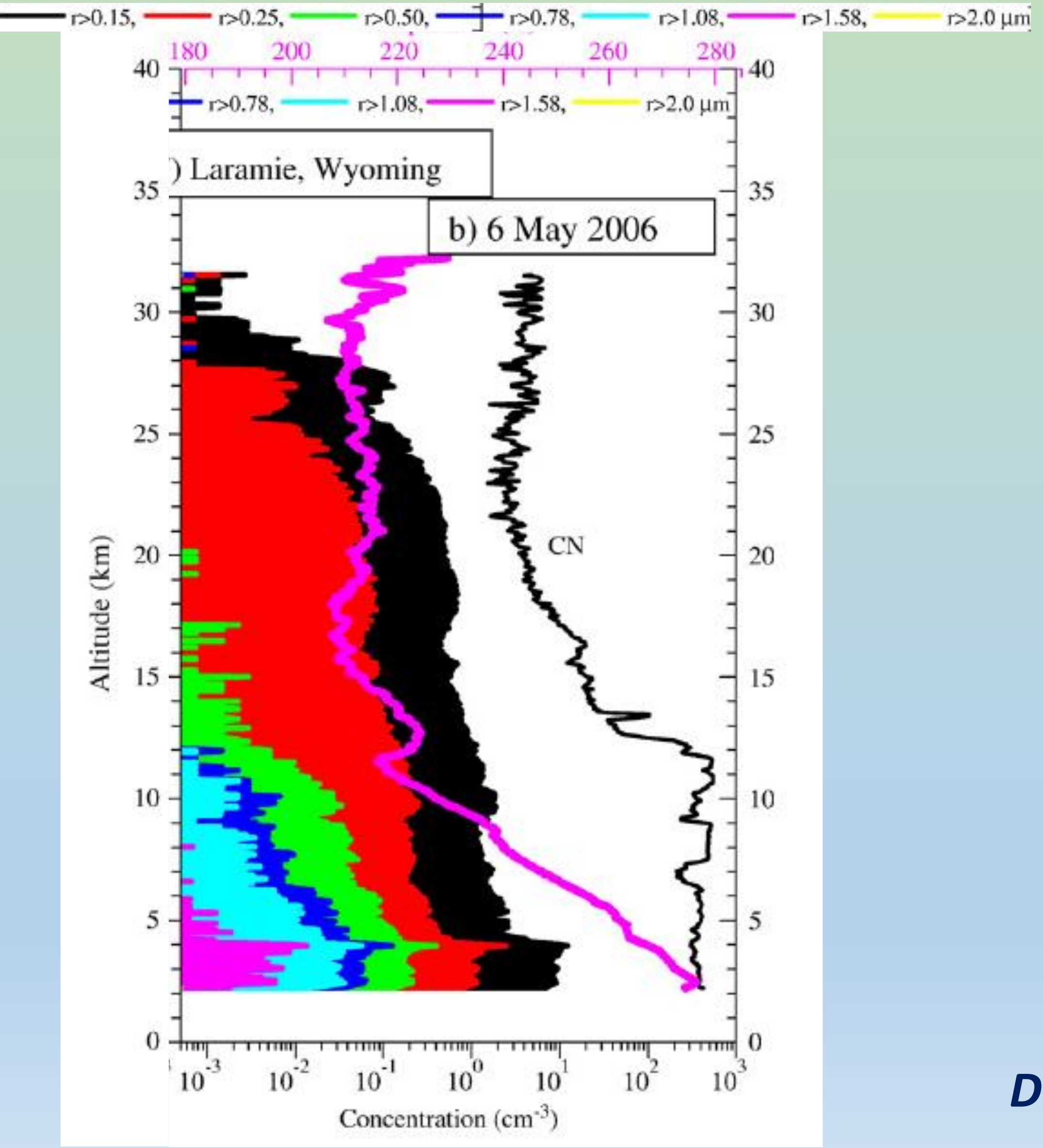


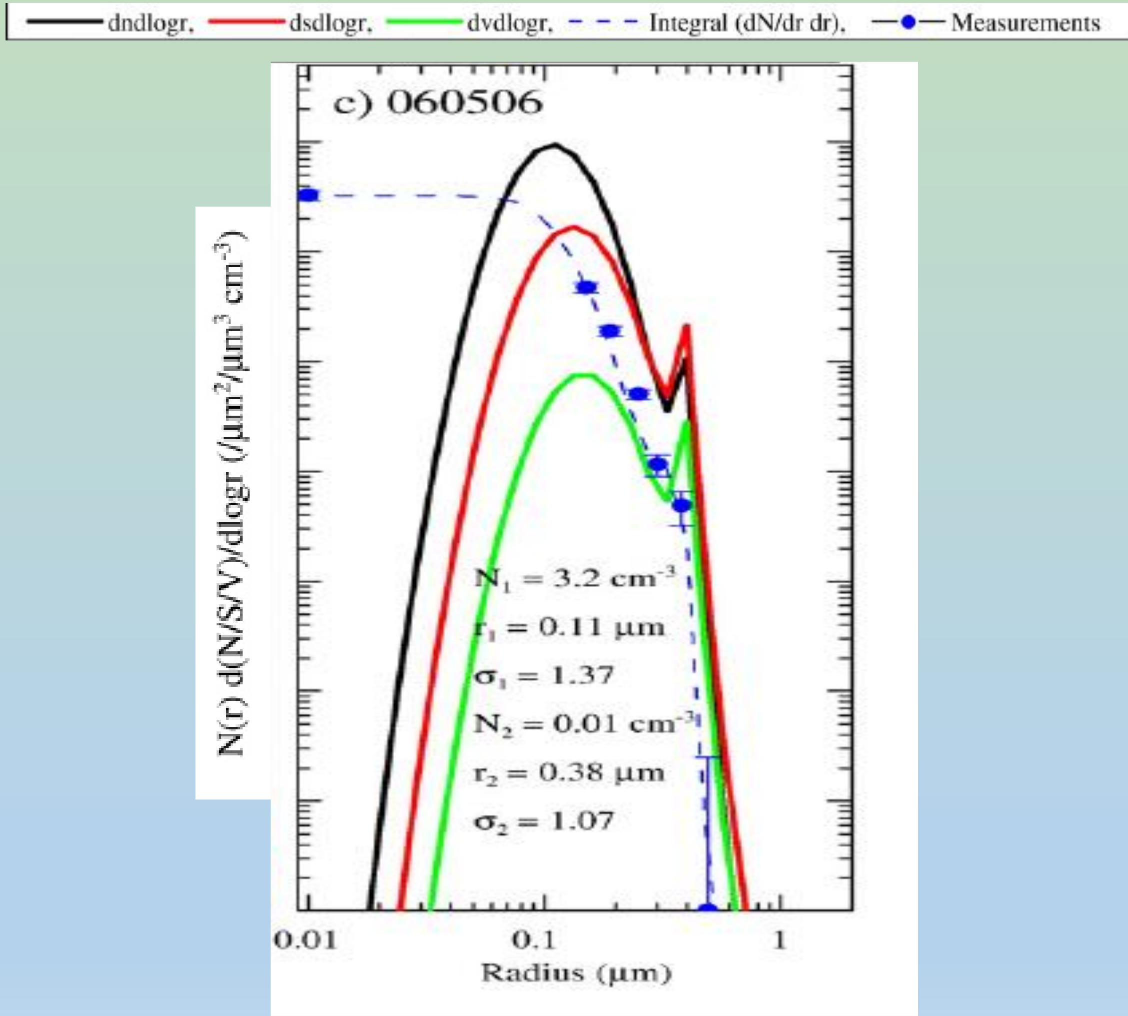
CESM1(WACCM)-CARMA, Year 2013



-> Slight seasonal cycle and latitudinal variations (tropopause temperatures and height, gas precursors, polar vortex)

« Background » stratospheric aerosol population from in situ observations





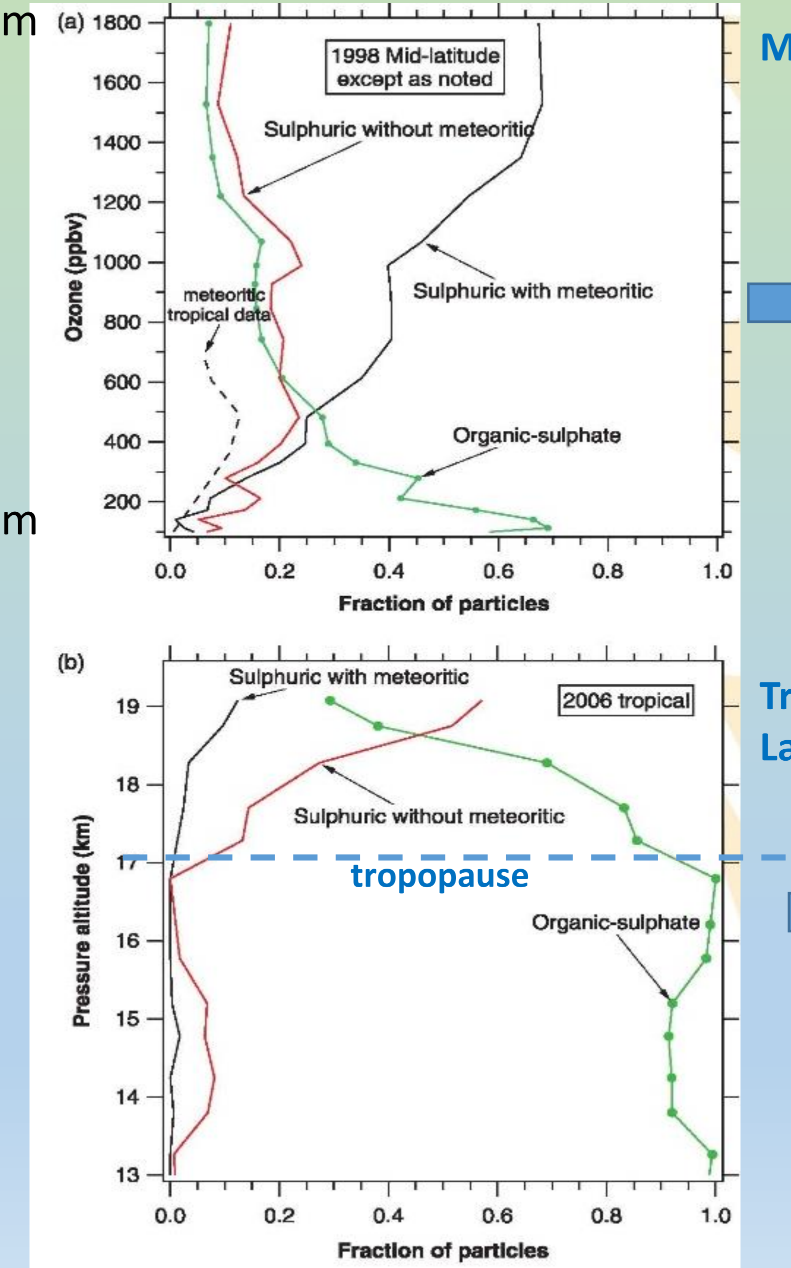
Deshler, Atmos. Res., 2008

Particle composition in the lower stratosphere: mass spectrometry observations from aircraft

Murphy et al., QJRMS, 2014

19 km

12 km



Mid-latitudes

Sulfuric acid particles with meteoritical material dominate above ~15 km

Tropical-Latitudes

Mixture between sulfate and organic particles largely dominate in the tropical UTLS

Black carbon in the lower stratosphere

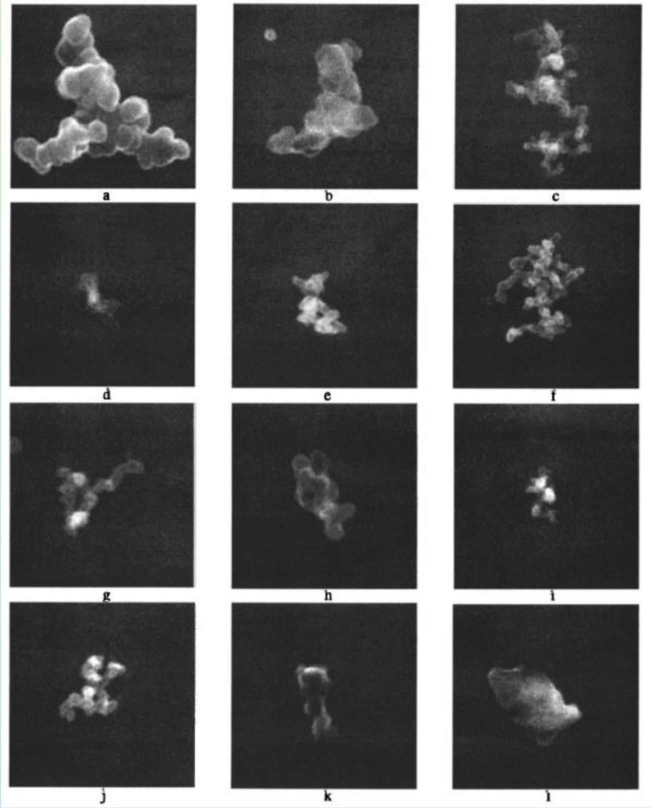
In fire plume quiescent period (or unreported events)

- ***Strawa et al., JGR, 1999***

Wire impactors

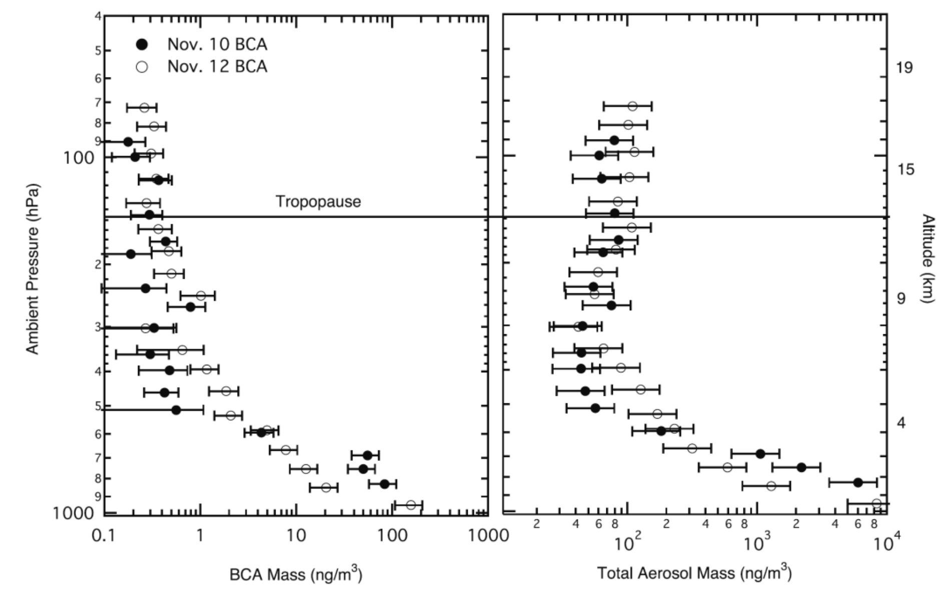
Black Carbon aerosol number densities ~1% of the total number

19 km 50°N



95% in the shape of aggregates

28-38°N



- ***Schwarz et al., JGR, 2006***

- **Soot photometer: laser-induced incandescence to detect refractory particles 0,15 - < 1µm**
- **Black carbon aerosols: <1% of total aerosol mass in the stratosphere**
- **at least 40% have "coating" (= internally mixed)**

Refractory particules in the lower stratosphere in the polar vortex (1/2)

Impactor for aerosol collection (M55 Geophysica)
+ laboratory transmission and scanning electron spectroscopy (TEM and SEM) for size/morphology and by EDX (Energy-dispersive X-ray) for composition.

Ebert et al., ACP, 2016

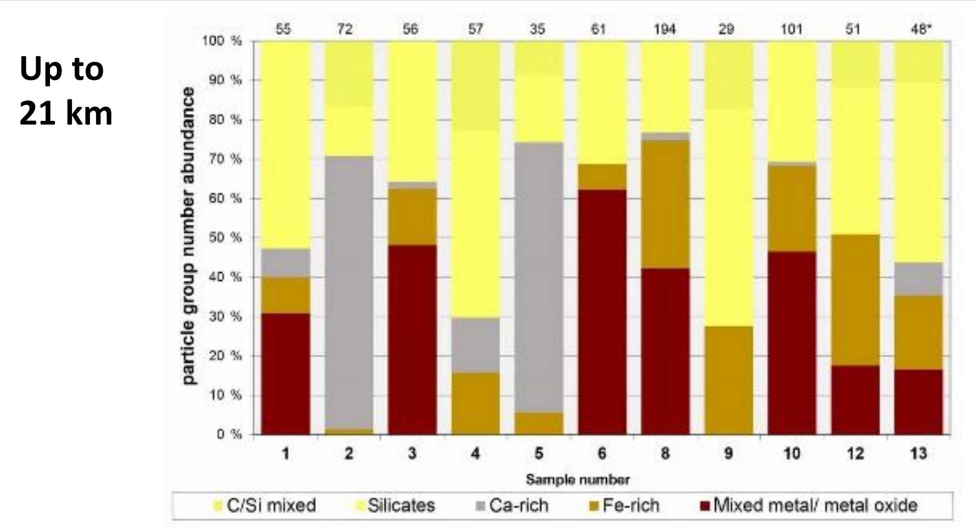
End of winter 2010: more refractory particles from the mesosphere
- sizes >0,5 µm : **silicate** spheroids, **silicate/carbon mixture**, **iron-rich particles**, **calcium-rich particles**, **complex mixture of metals**
- sizes <0,5 µm : **Presence of soot**
- estimated concentrations : 10^{-2} cm^{-3} for sizes of 0.75 µm

Schutze et al., ACP, 2017

Aerosol collection *in the Artic stratosphere* ER2 aircraft, *Jan-Feb 2000*
Mainly carboncaceaous particles associated with metals

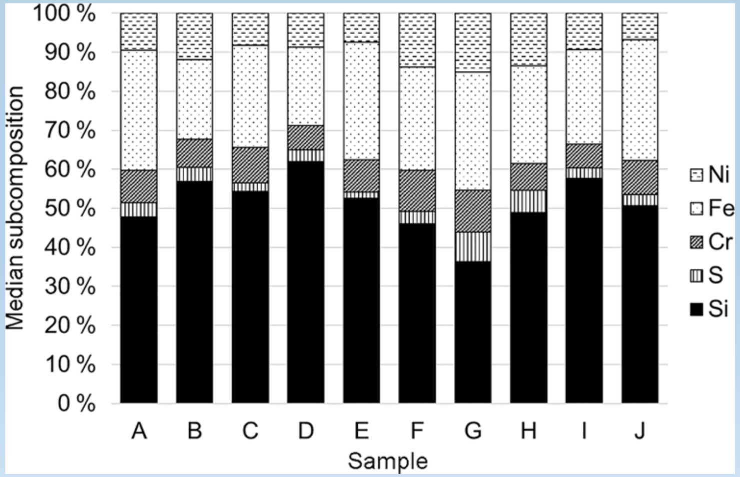
Composition:
- Particles with different morphologies than Ebert et al.'s results
- Only **amorphous refractory carbonaceous particles** collected (among the sulfate population) C+O (72-100%) + others **with metals**.
- Refractory carbonaceous particles not included in or coated by sulfate are surprisingly dominant
  -> in contradiction with results from Mass Spectrometry.
- Concentrations : a few percent of the total number of particles in the air (between 0.62 and 2.3 $(mg.air)^{-1}$

Arctic stratosphere, winter 2010 (Ebert et al., ACP, 2016)



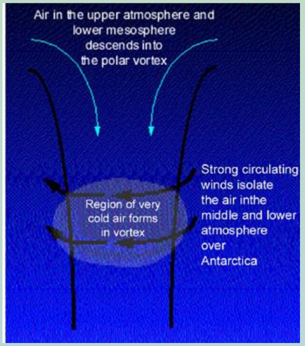
Up to 21 km

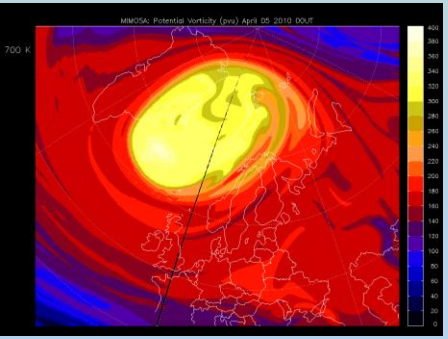
Arctic stratosphere, Jan-Feb 2000 (Schutze et al., ACP, 2017)

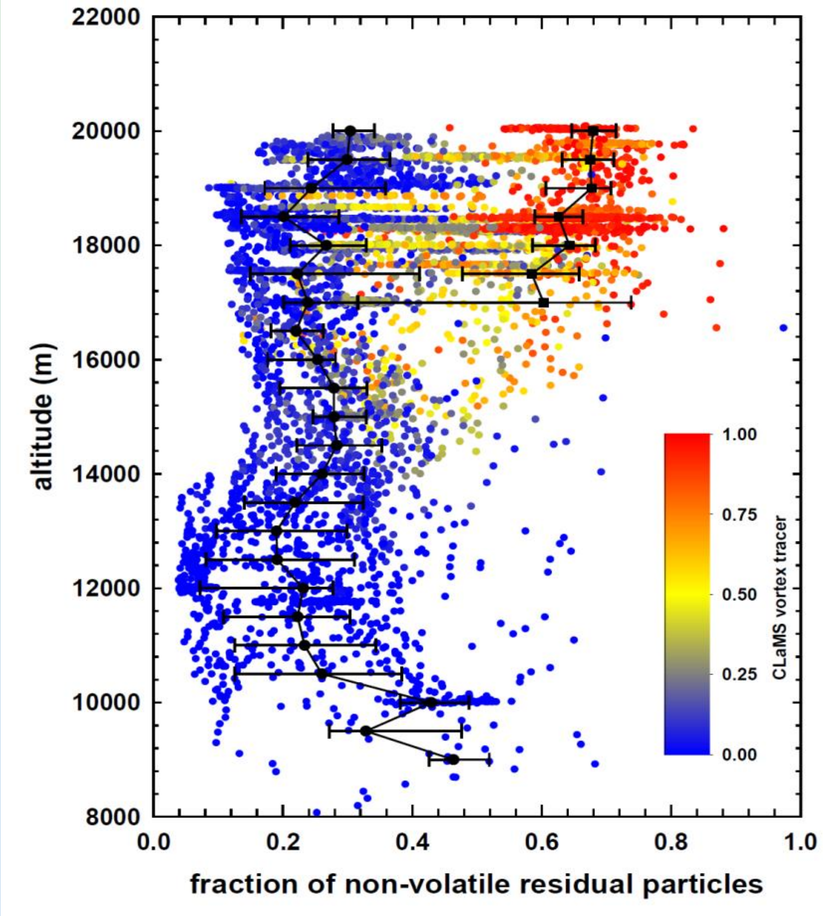


Refractory particules in the lower stratosphere in the polar vortex (2/2)

Disintegrated meteorites



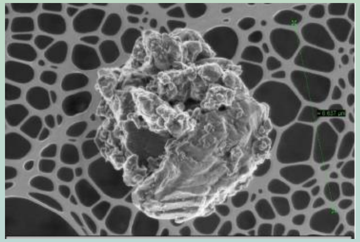




February-March 2003

Aerosol counter with particle vaporisation

Size range 0.4-23 µm



Interplanetary grains

In the polar vortex:

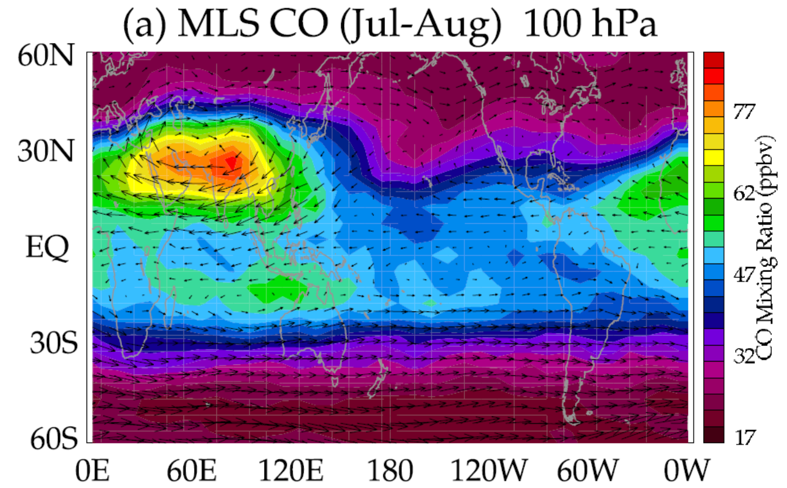
→ more than 60% contain non-volatile particles / Total concentration

→ Meteoric material transported from the mesosphere

Curtius et al., ACP, 2008

Asian monsoon anticyclone: presence of polluants and aerosols in the tropopause region (1/4)

~15-19 km altitude range

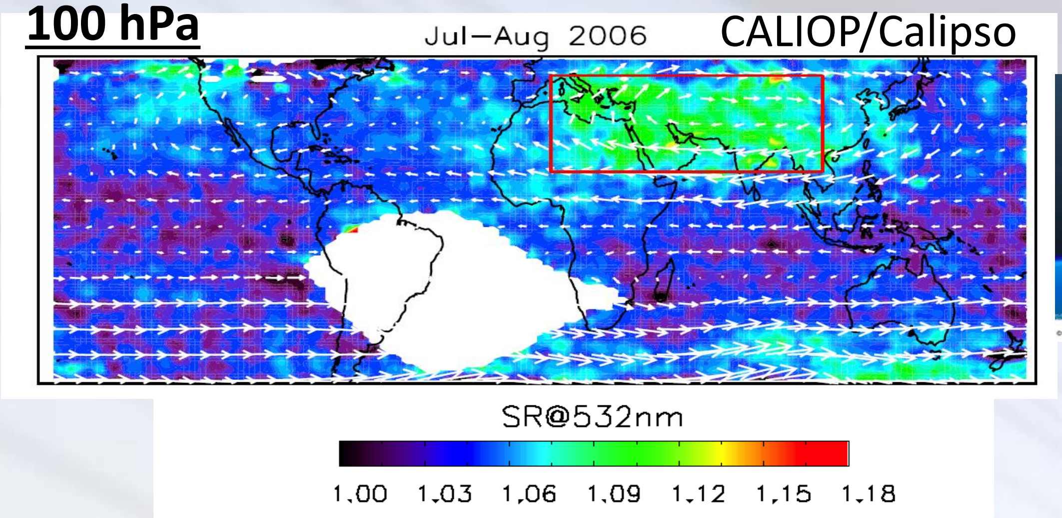


Park et al. (JGR, 2006)

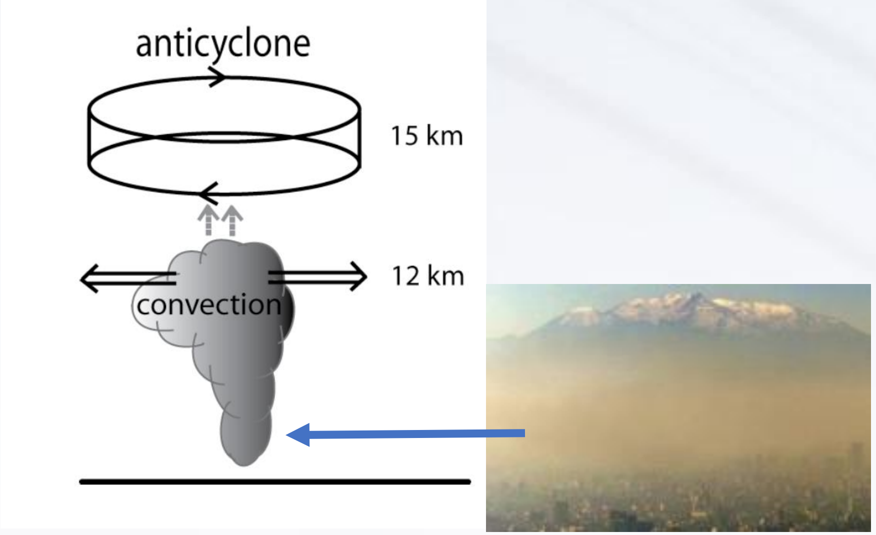
Asian Tropopause Aerosol Layer (ATAL):

100 hPa

CALIOP/Calipso

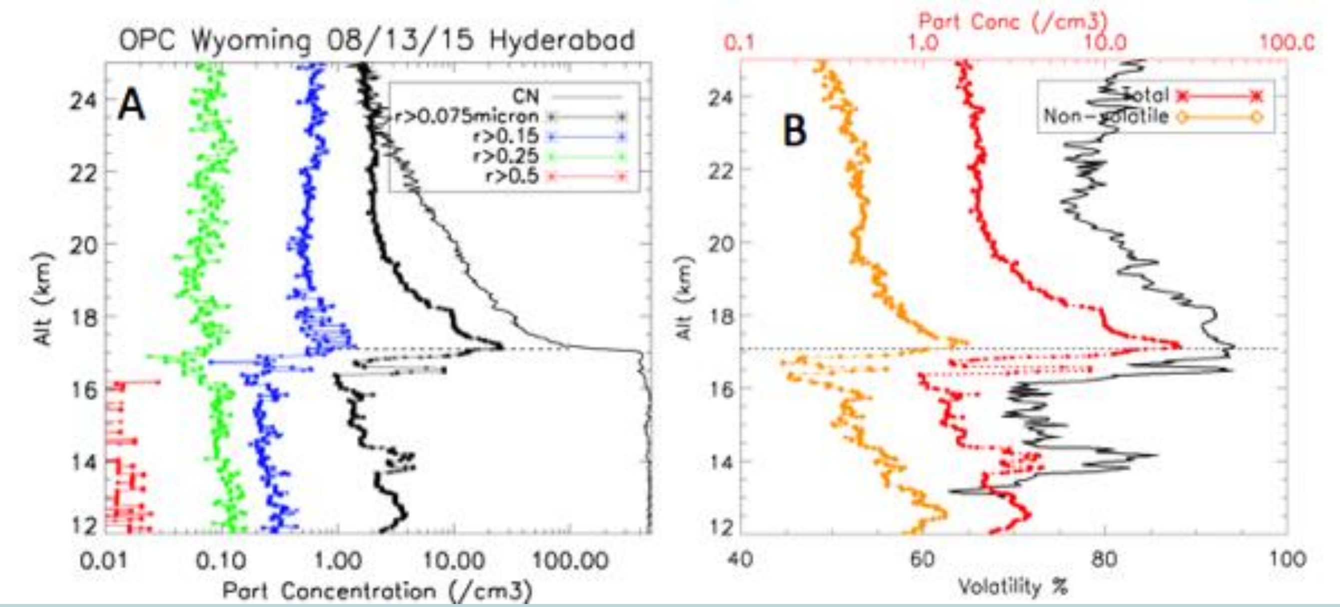


Vernier et al. (GRL, 2011)



Park et al., ACP, 2008

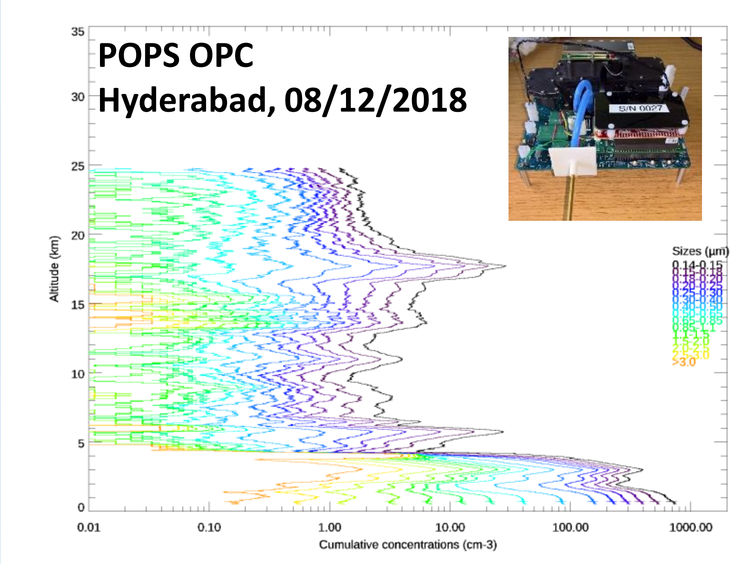
Aerosol counting measurements using balloons within the ATAL: BATAL campaigns (2/4)

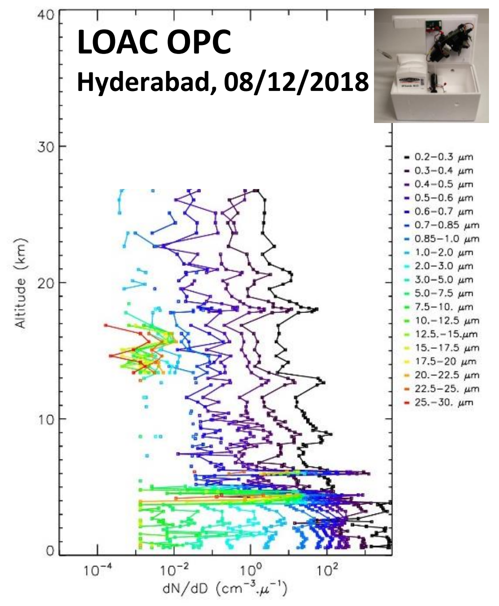


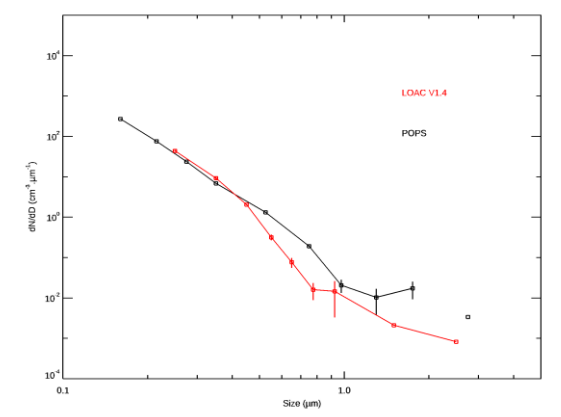
Vernier et al., BAMS, 2017

Campaigns coordinated by NASA, ISRO (India), NARL (India)

- Sharp increase of aerosol concentration near 16-18 km
- 90% of volatile aerosol
- small particles



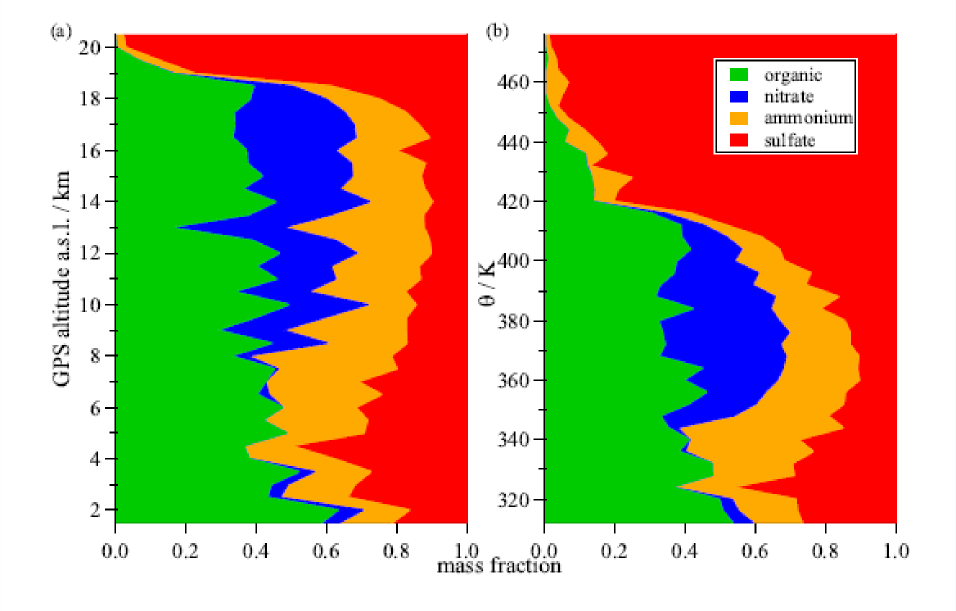




-> ATAL made of very small, volatile aerosol and thus newly formed particles ?

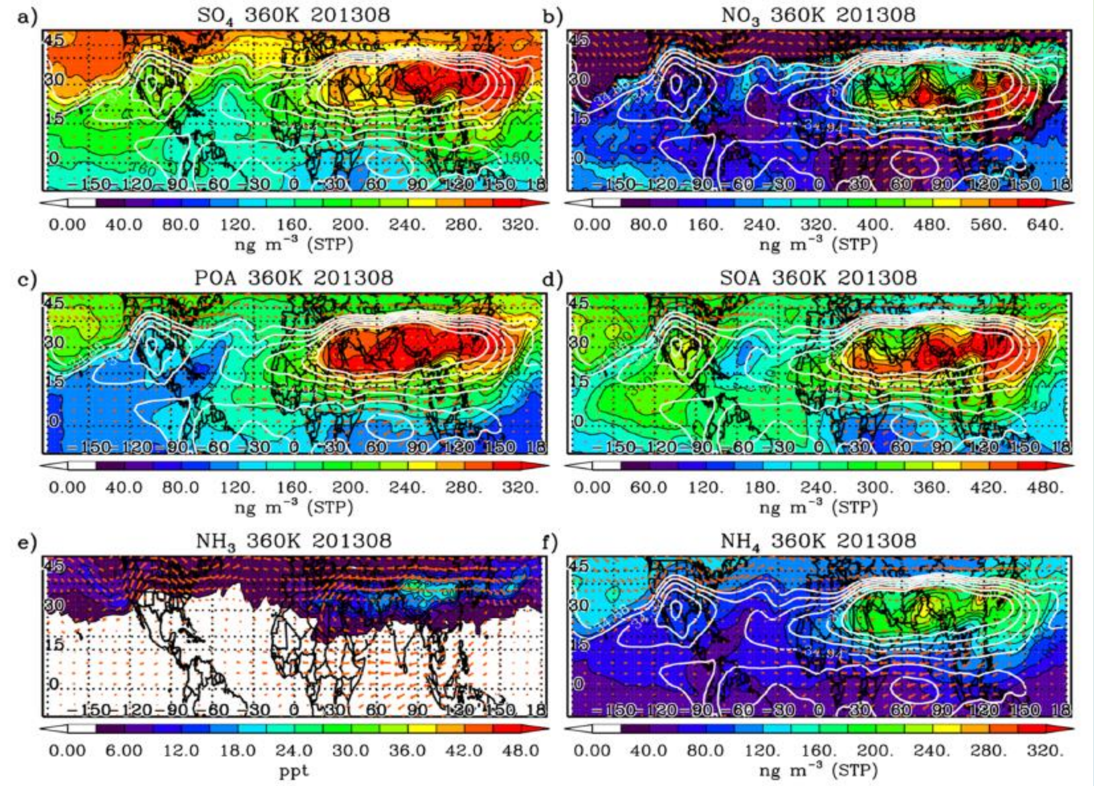
Aerosol composition within the ATAL (3/4)

StratoClim EU campaign, summer 2017
<u>Mass spectrometry </u>observations, M-55 Geophysica
Nepal, India, Bangladesh, Bay of Bengal



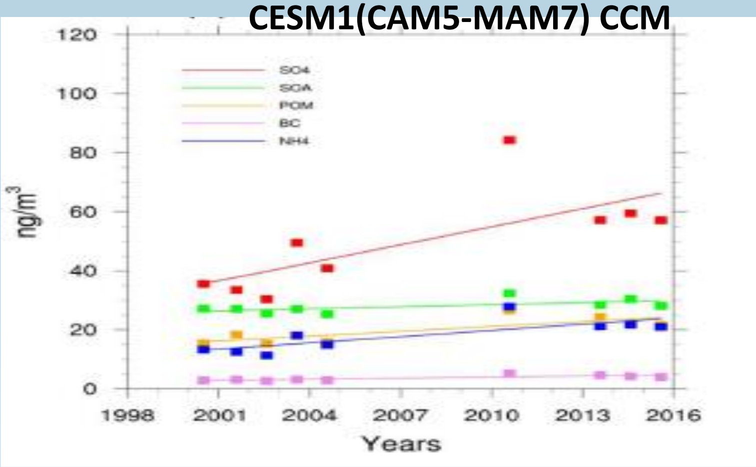
Appel et al., ACPD, 2022

GEOS-Chem CTM, summer 2013



Fairlie et al., JGR, 2019

CESM1(CAM5-MAM7) CCM



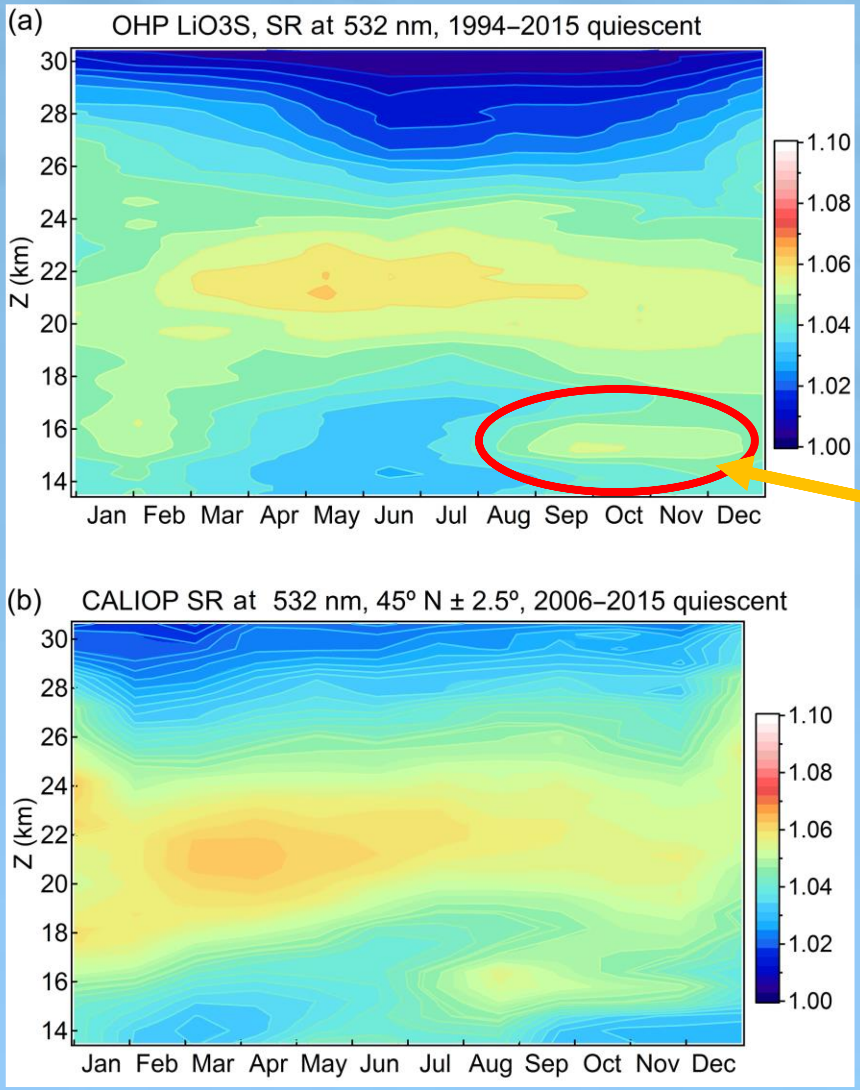
Bossolasco et al., ACP, 2021

-> ATAL particles mainly resulting from the conversion of inorganic and organic gas precursors rather than from the uplift of primary particles from below.

Signature at mid-latitudes (4/4)

Ground-based lidar
Observatory of Haute Provence (44°N; 6°E), France

CALIOP/Calipso
Zonal mean 45°N



SR = Scattering Ratio

Signature of ATAL aerosols above France after the breakdown of the monsoon anticyclone

Khaykin et al., ACP, 2017

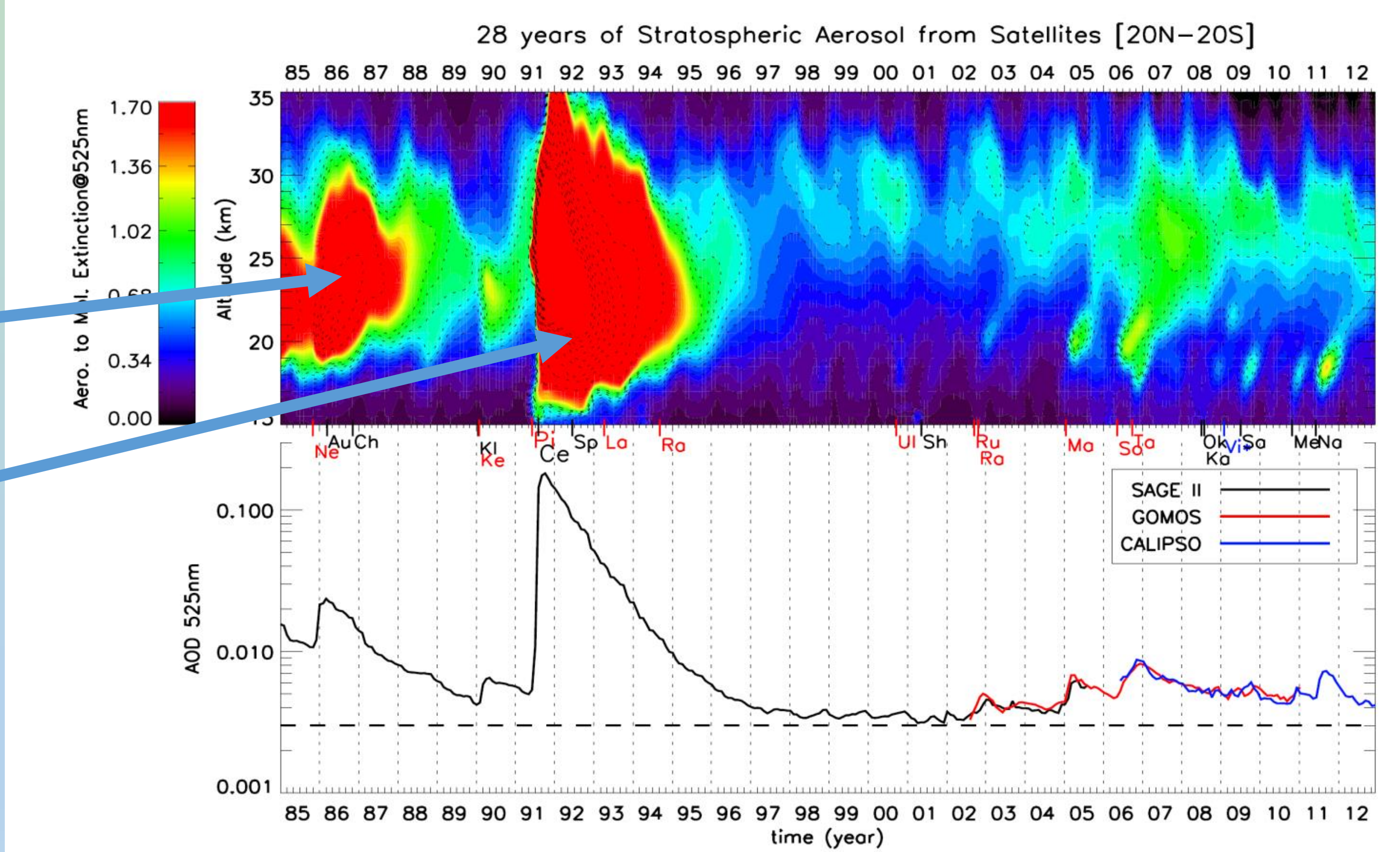
VOLCANIC PLUMES

Interannual variability of the stratospheric aerosol content

The 2 last major eruptions:

El Chichon

Pinatubo



Tropics 20°S-20°N

SAGEII UV-Visible spectrometer
CALIOP/Calipso space-borne lidar

Extinction ratio
(= Aerosol Extinction / Molecular Extinction)

Courtesy from

Jean-Paul Vernier

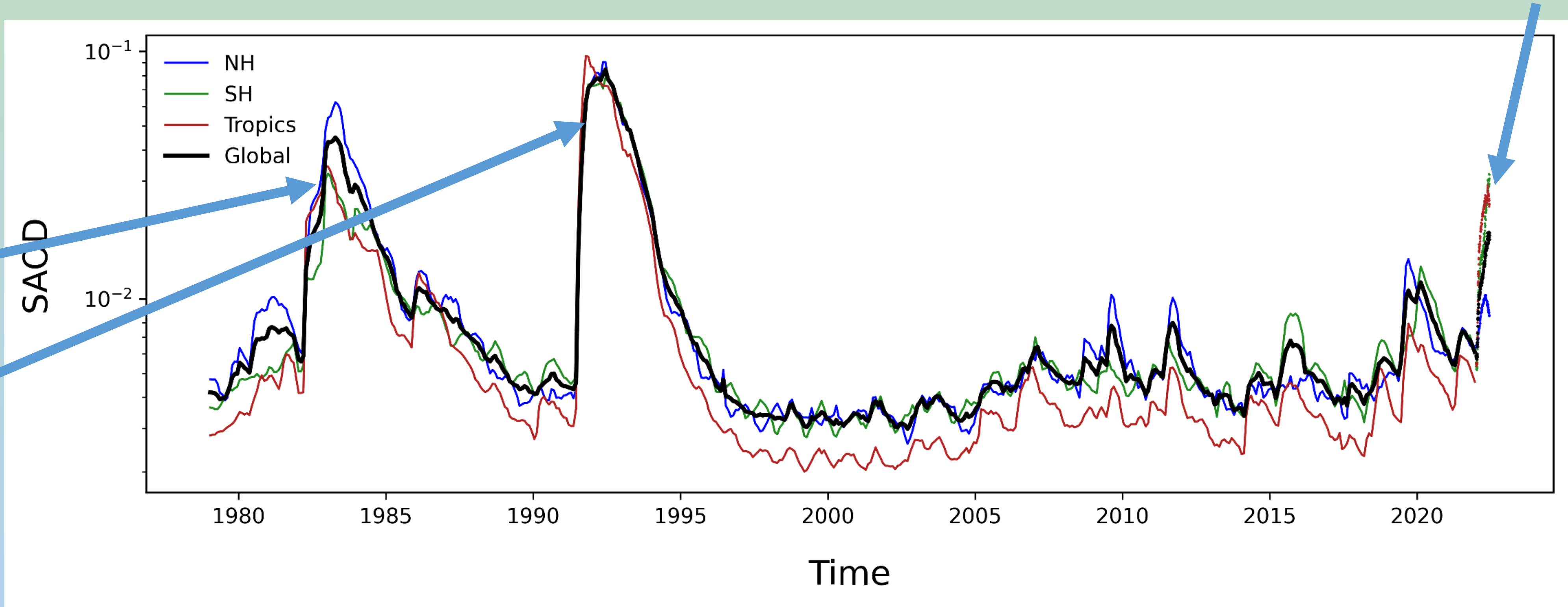
Interannual variability of the stratospheric aerosol content

Merging of satellite data: GloSSAC v2.2 + OMPS-LP

Hunga Tonga

The 2 last major eruptions:

El Chichon



Pinatubo

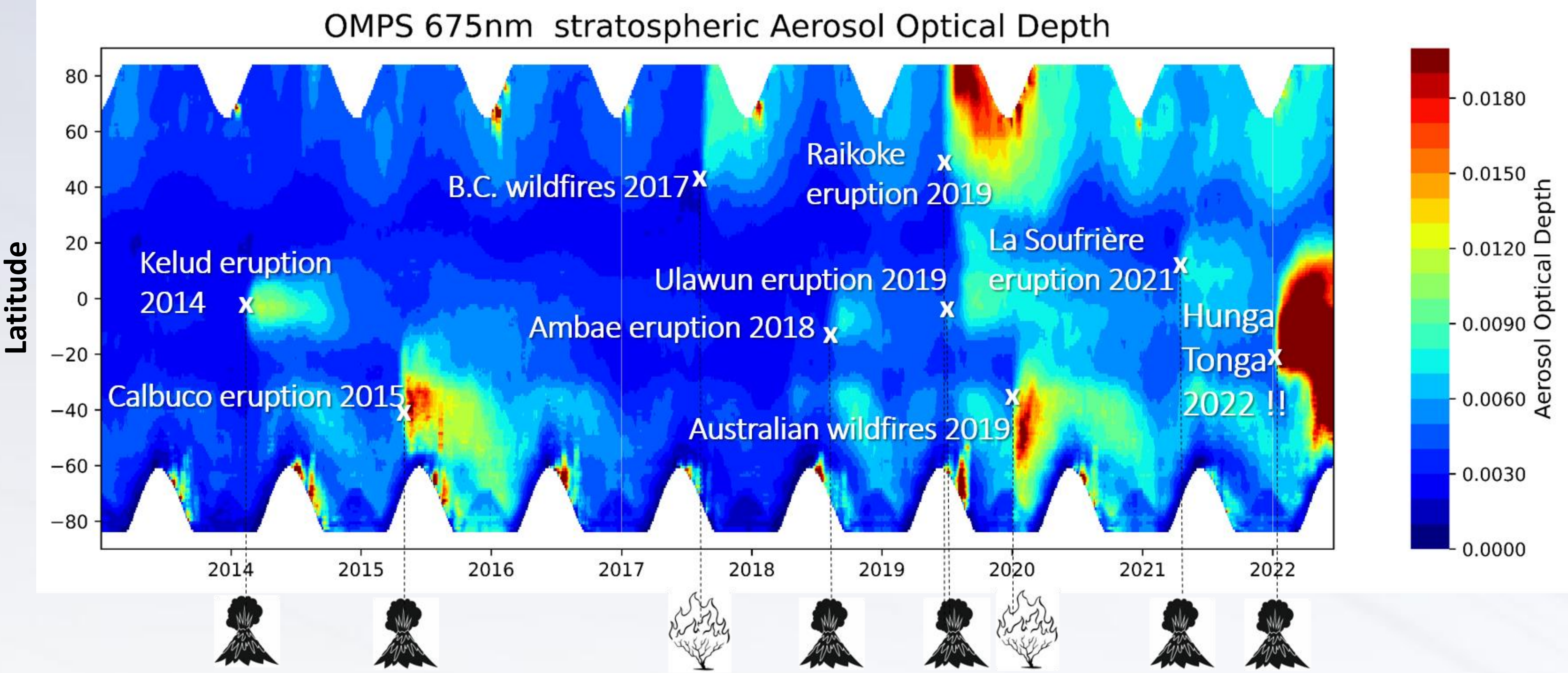
Courtesy from Pasquale Sellitto, Preliminary plot

Sellitto et al., under revision, Nature Comm., 2022, https://assets.researchsquare.com/files/rs-1562573/v1_covered.pdf?c=1650312853

doi: https://doi.org/10.21203/rs.3.rs-1562573/v1

Legras et al., ACPD, 2022, https://doi.org/10.5194/egusphere-2022-517

Over the more recent years



In situ observations of volcanic plumes (HEMERA support)

HEMERA frame and support for:

-**instrumental development** (e.g. LOAC V1.2 to V1.5 for **improved performances**: reduced stray light, powerful laser source)

-**rare regular comparisons** of in situ observations of stratospheric aerosols using small (latex) balloons: LOAC vs POPS + commercial instruments from NASA-LARC (J.-P. Vernier)

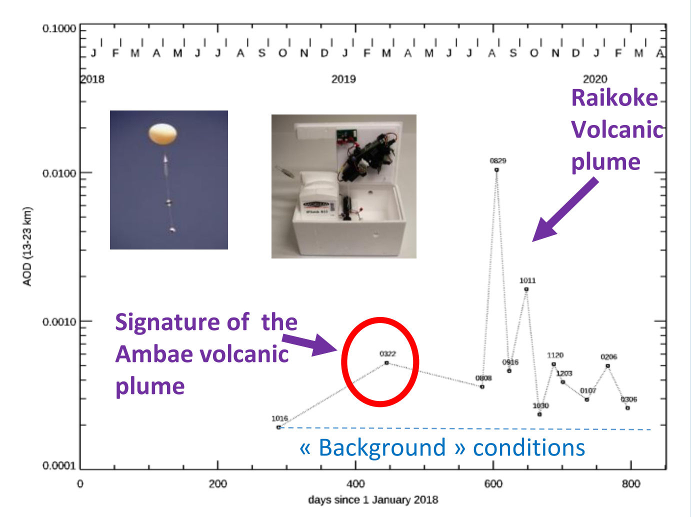
-Two **flights in Kiruna** in February and December 2020

Together with support from **University of Orleans** (VOLTAIRE project), **CNES**, **ANR** (French national Research Agency)

LOAC *(LPC2E* & MeteoModem company, France) *1 kg payload*

***POPS** (Handix Scientific company, USA) 1 kg payload*

Mid-latitudes



Raikoke Volcanic plume

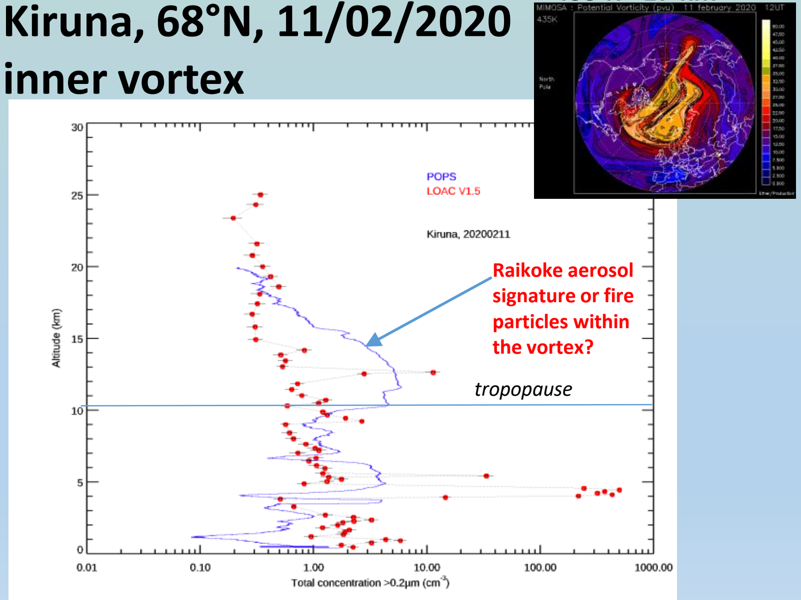
Signature of the Ambae volcanic plume

« Background » conditions

LOAC OPC flights from CNES base (AsA, 44°N) and MeteoModem company site (48°N), France

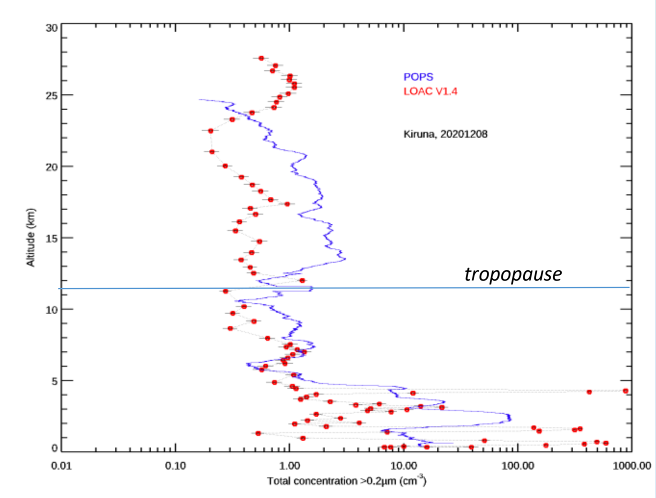
Kiruna, 68°N, 11/02/2020 inner vortex

435 K ~17 km



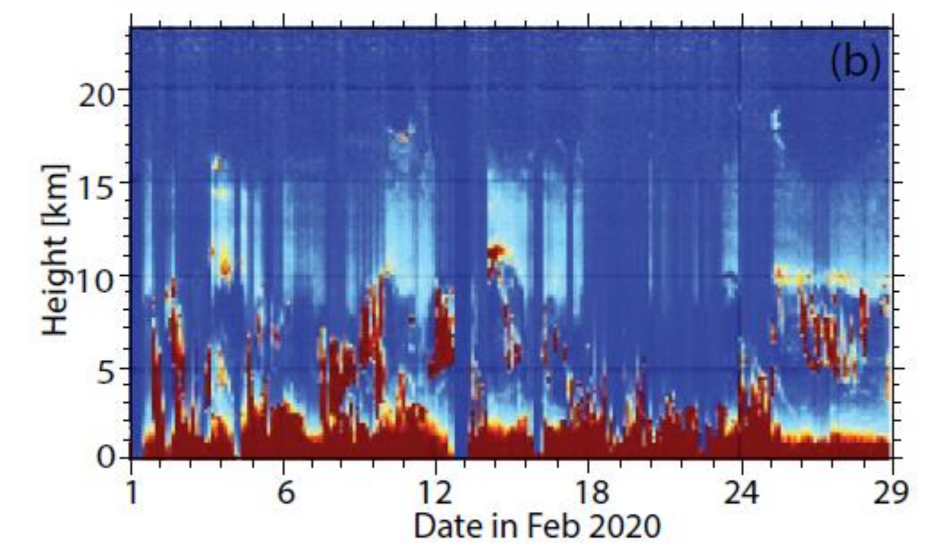
Raikoke aerosol signature or fire particles within the vortex?

Kiruna, 68°N, 08/12/2020 early vortex

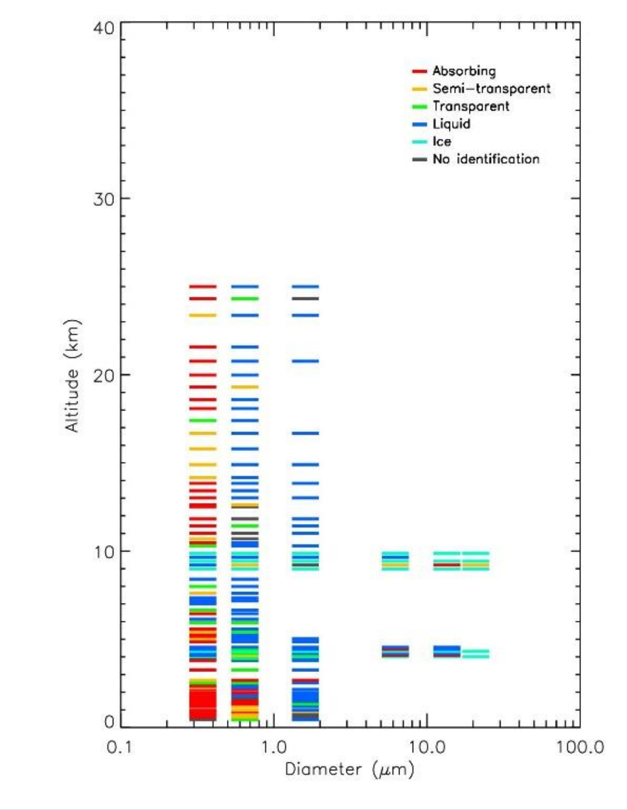


-> Impact of fire smoke on ozone depletion in winter 2019-2020 proposed by Ohneiser et al., ACP, 2021; Ansmann et al., ACPD, 2022)

Winter 2020



Lidar observations (of smoke haze?) over the Polarstern (85-88.5°N)
Ansmann, ACPD, 2022



LOAC v1.5 OPC
Kiruna, 68°N, 11/02/2020
inner vortex

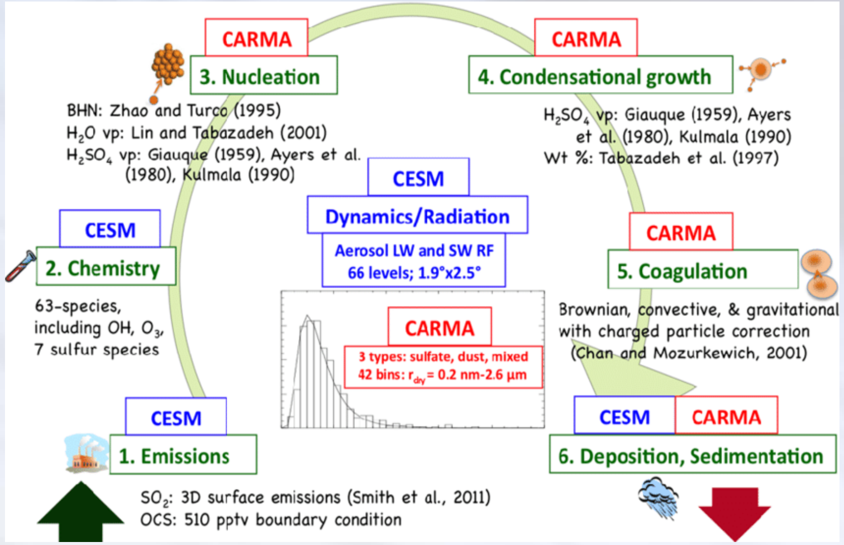
Need for model simulations including volcanic/smoke plume

Community Aerosol Model for Atmospheres (CARMA) Coupled with the CESM1(WACCM) Chemistry-Climate Model

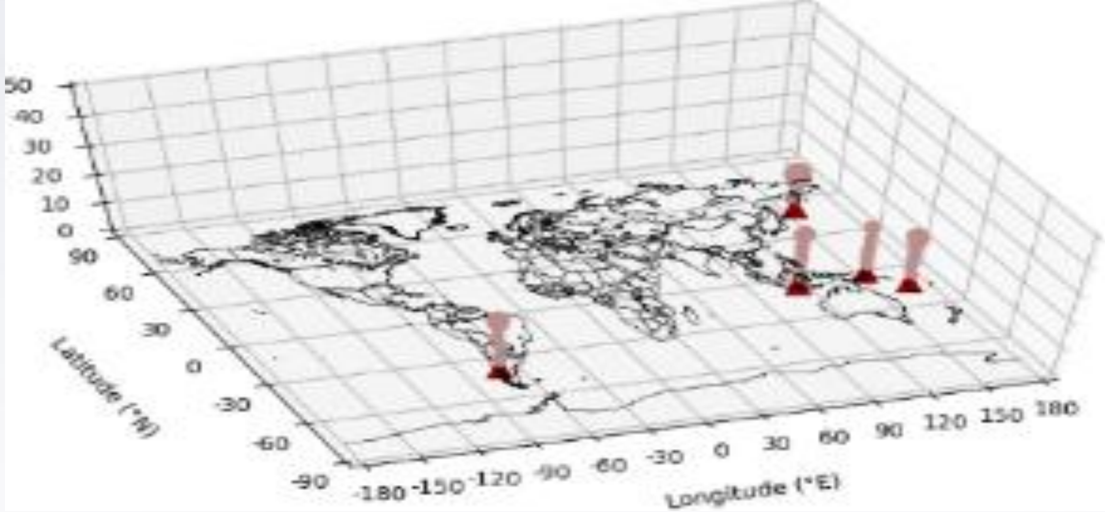
Simulations:
CESM1(WACCM) 1.9°x2.5° lon/lat, 88 vertical levels
Chemistry : waccm_mozart_sulfur
CARMA microphysical module : 30 aerosol size bins

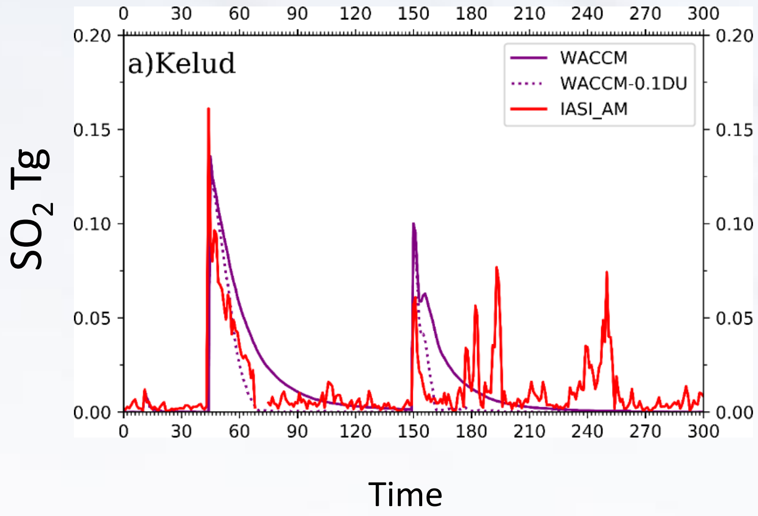
Dynamics: Nudging towards MERRA2 reanalysis

Volcanic emissions of SO_2 (injection timing, altitude, amount of sulfur): model driven by information from satellite data and/or literature



English et al., ACP, 2011



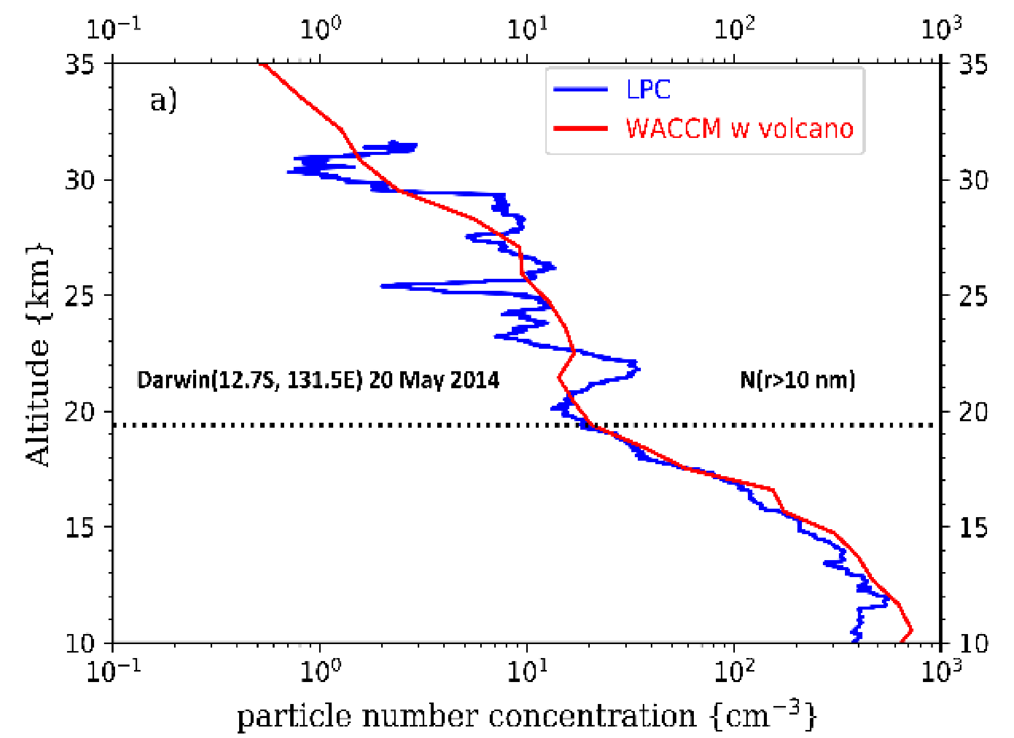


Tidiga et al., Atmosphere, 2022

Simulating the condensation nuclei profiles

LPC Optical particle counter (Univ. Wyoming; PI: Terry Deshler) vs WACCM-CARMA model

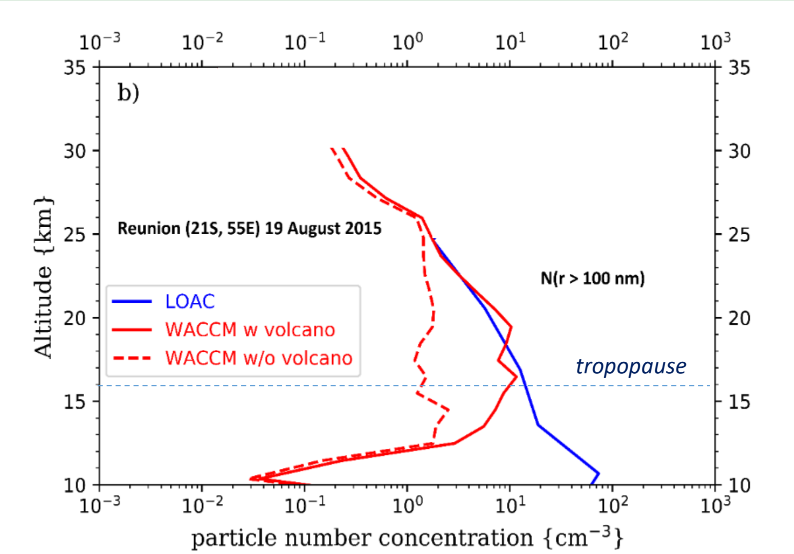
Darwin, Australia
KlAsh campaign
May 2014

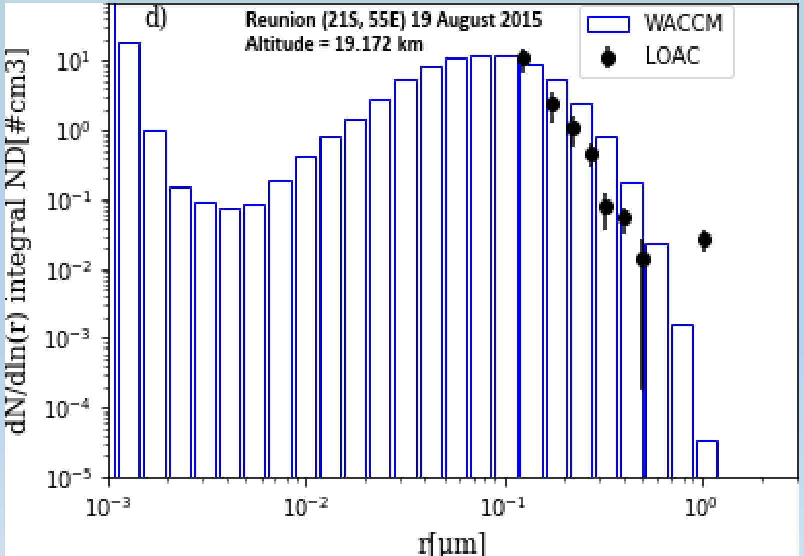


Tidiga et al., Atmosphere, 2022

Simulated vs observed size distributions of volcanic sulfuric acid particles

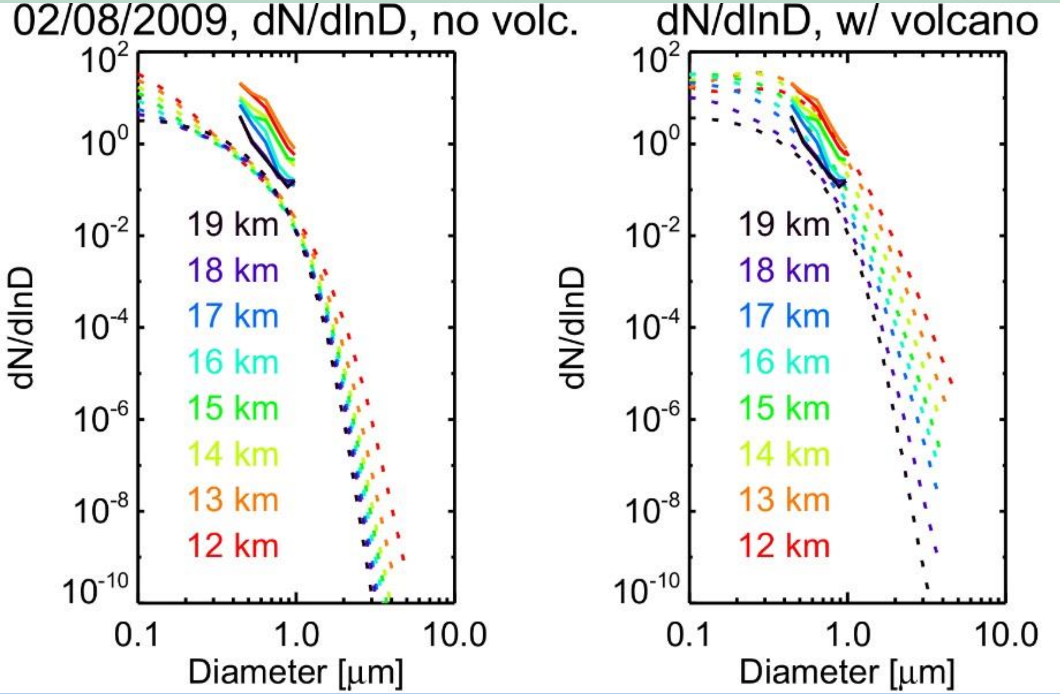
LOAC (v1.2) OPC vs WACCM-CARMA model
La Réunion island.
Calbuco volcanic plume (2015)





Adapted from
Tidiga et al., Atmosphere, 2022
and Zhu et al., JGR, 2018

STAC OPC vs WACCM-CARMA model, Kiruna, StraPolEté project, Northern Sweden. **Sarychev volcanic plume (2009)**



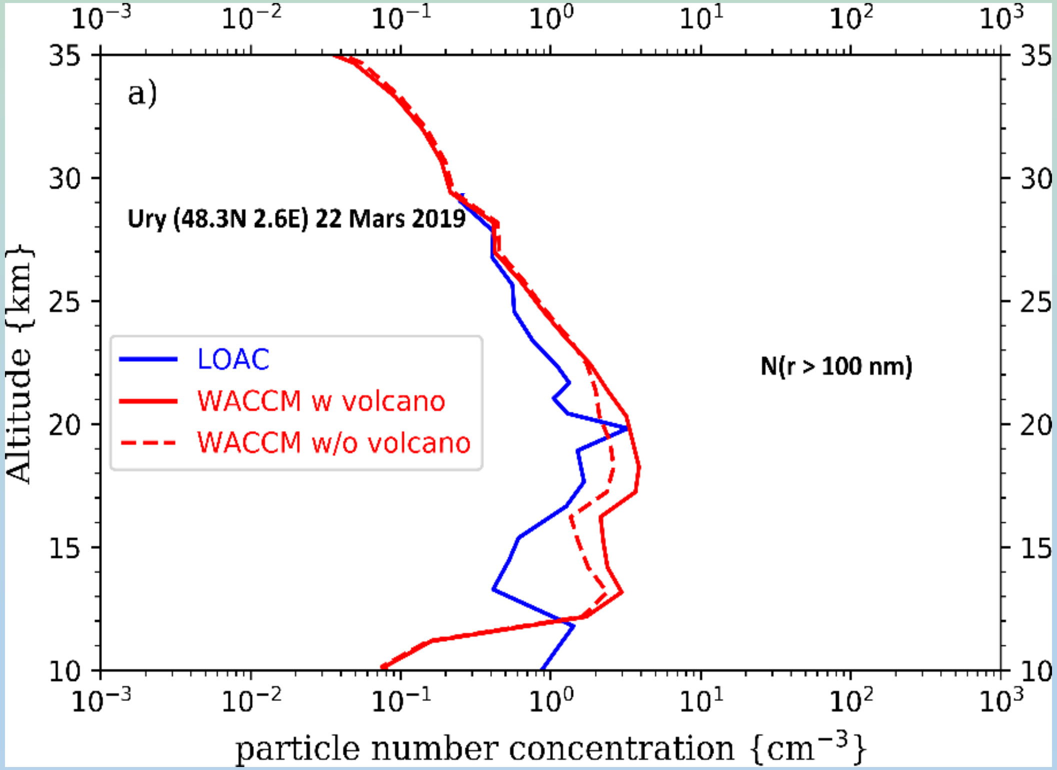
Dotted Lines= WACCM-CARMA model

Full lines = Optical Particle Counter observations

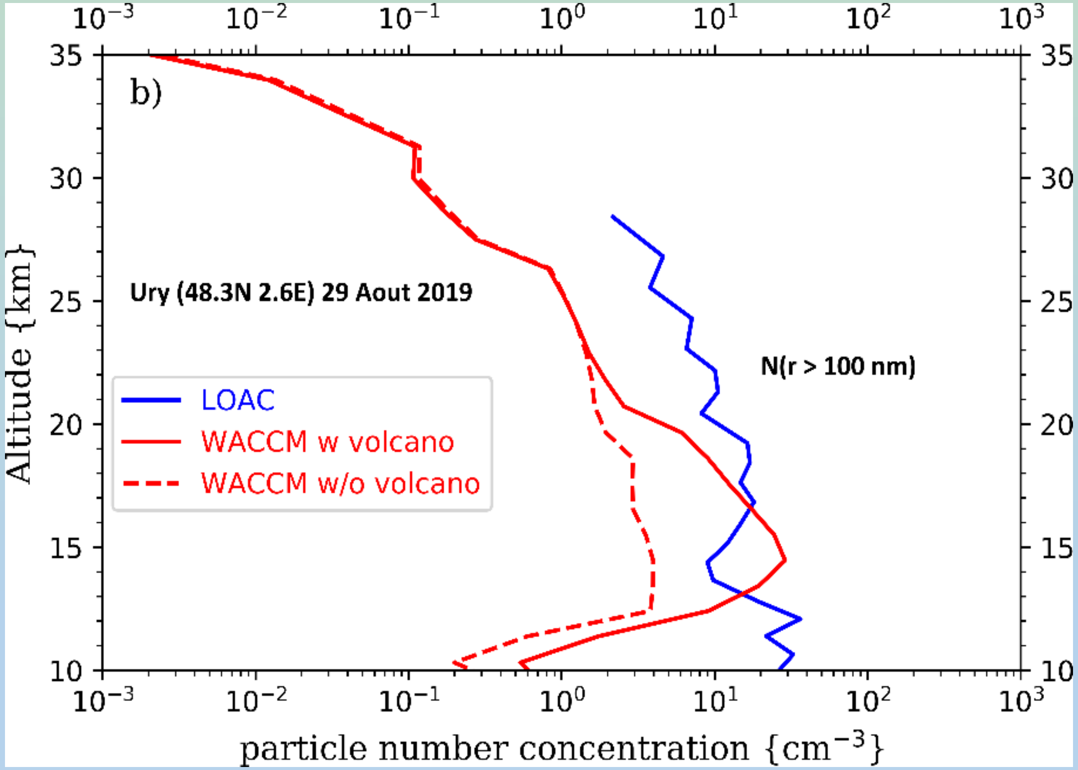
Lurton et al., ACP, 2018

Simulated vs observed size distributions of volcanic particles

« Aged » Ambae plume
(eruption 07/2018)
Decaying in 2019



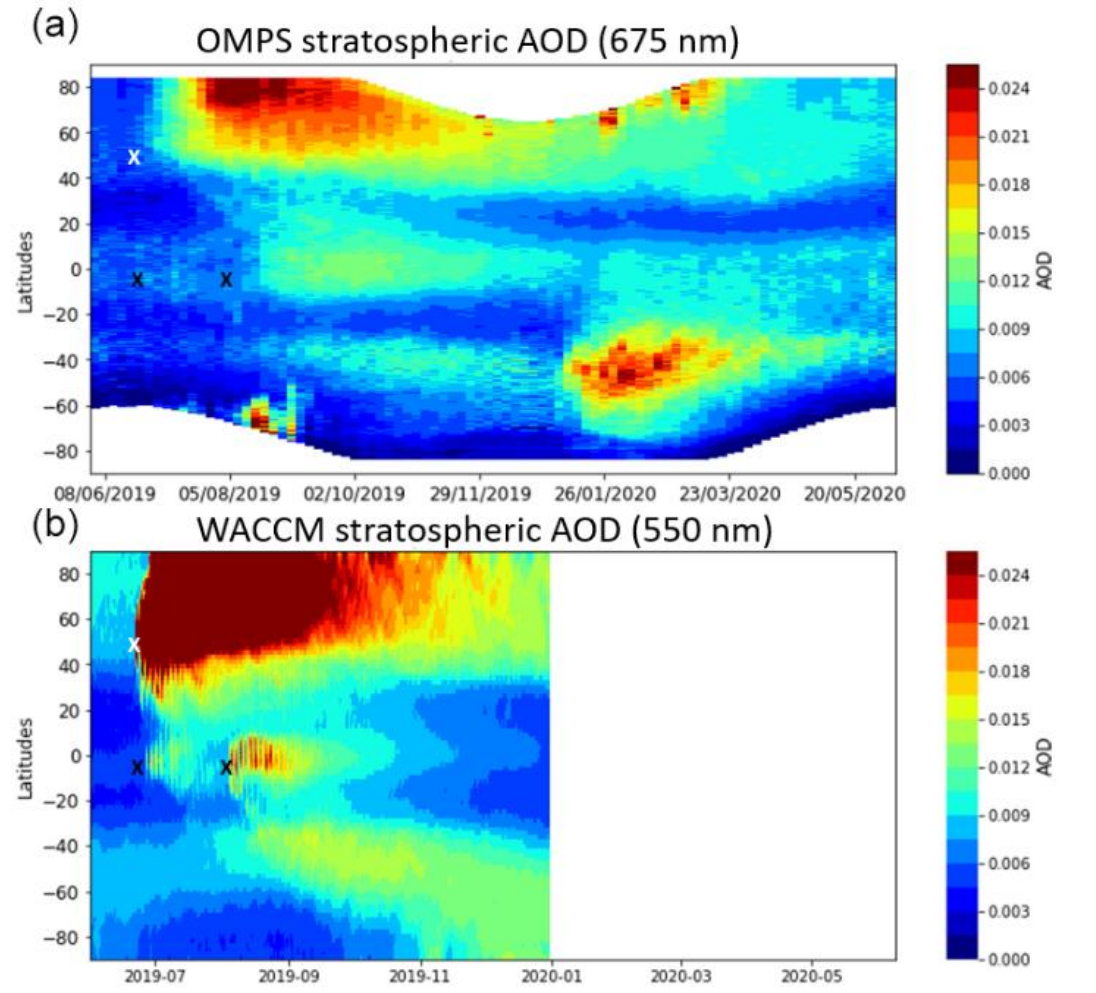
Early Raikoke plume
(eruption 06/2019)



Simulating the transport of the Raikoke plume

Aerosol Optical Depth

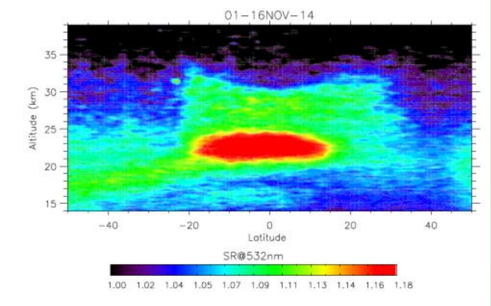
OMPS
vs
WACCM-CARMA model



Kloss et al., ACP, 2021

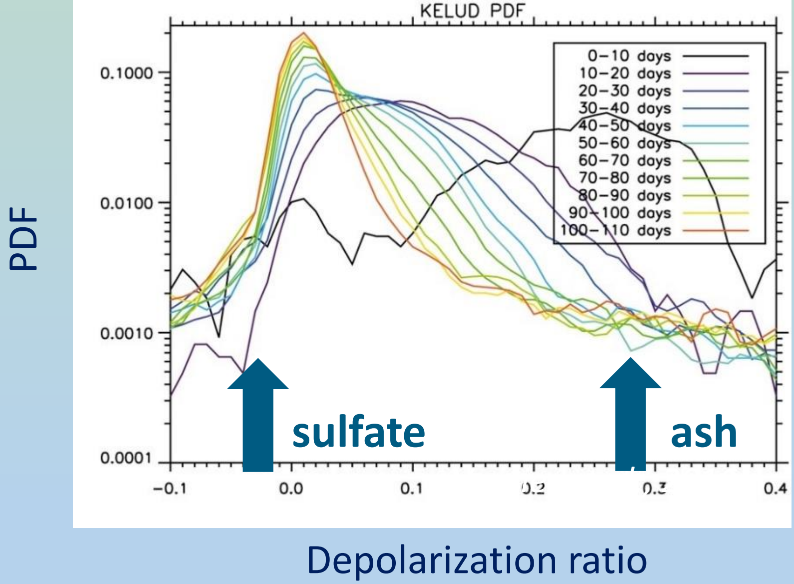
Injection parameters (SO_2 burden, altitude range, injection timing) not always derived robustly from satellite observations

Co-injection of ash



Ash detection by CALIOP/Calipso

Kelud volcano case (Feb. 2014)

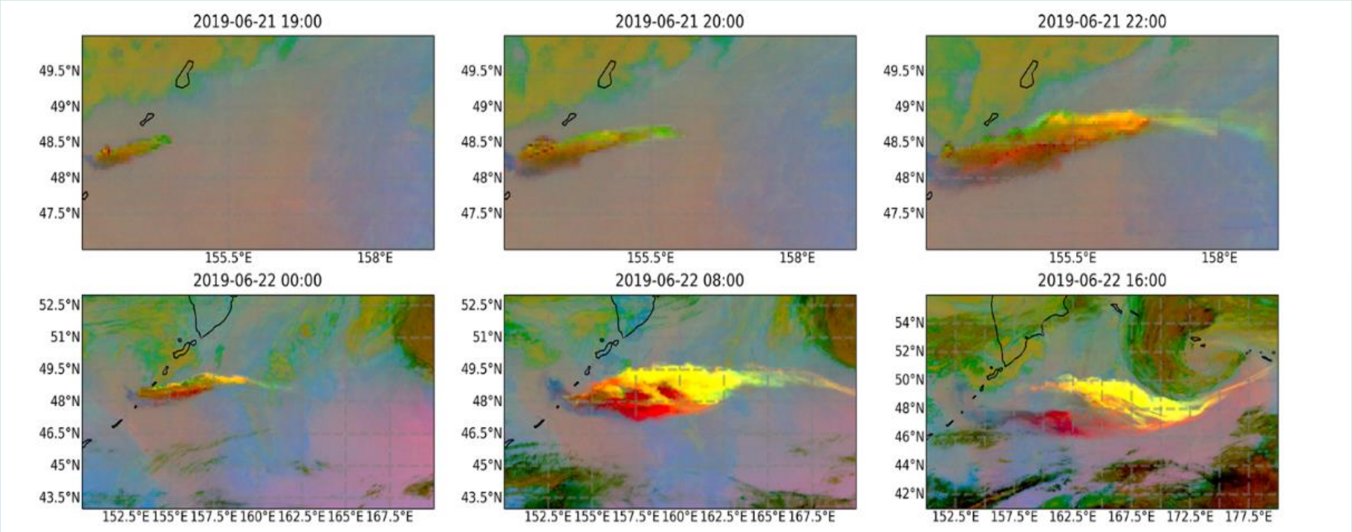


Vernier et al., JGR, 2016

Raikoke volcano, Kuril Islands (north of Japan)

Source - © 2019 NASA / Earth Observatory

Himawari RGB images
June 2019



Red: ash **Green**: sulfur **Yellow**: mixture

Kloss et al., ACP, 2021

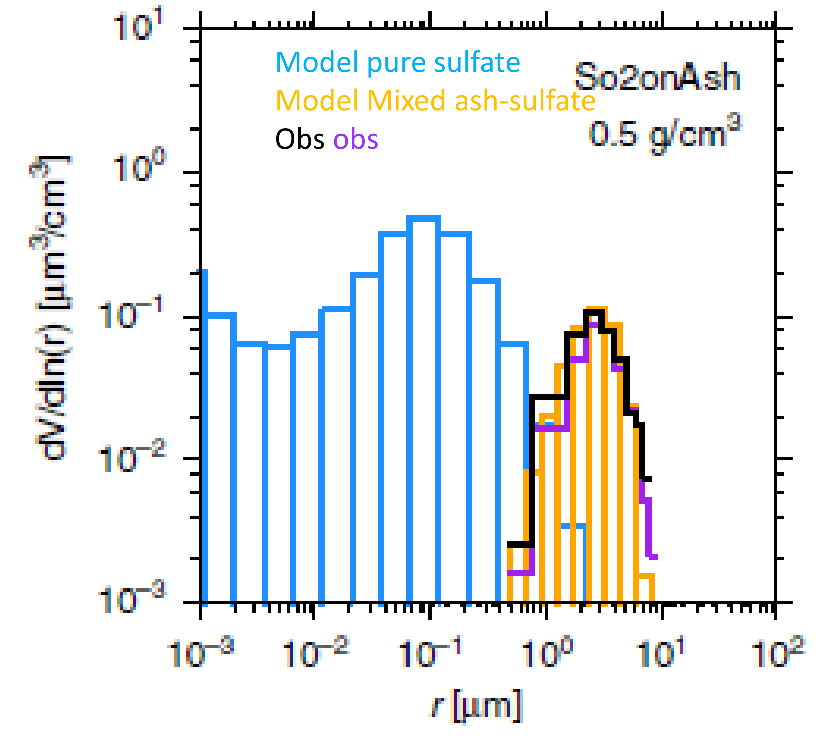
Radiative effects of ash on the plume transport (see ***Muser et al., ACP, 2020***)

+ injection of water vapour?
+ interaction with smoke plumes from north America?

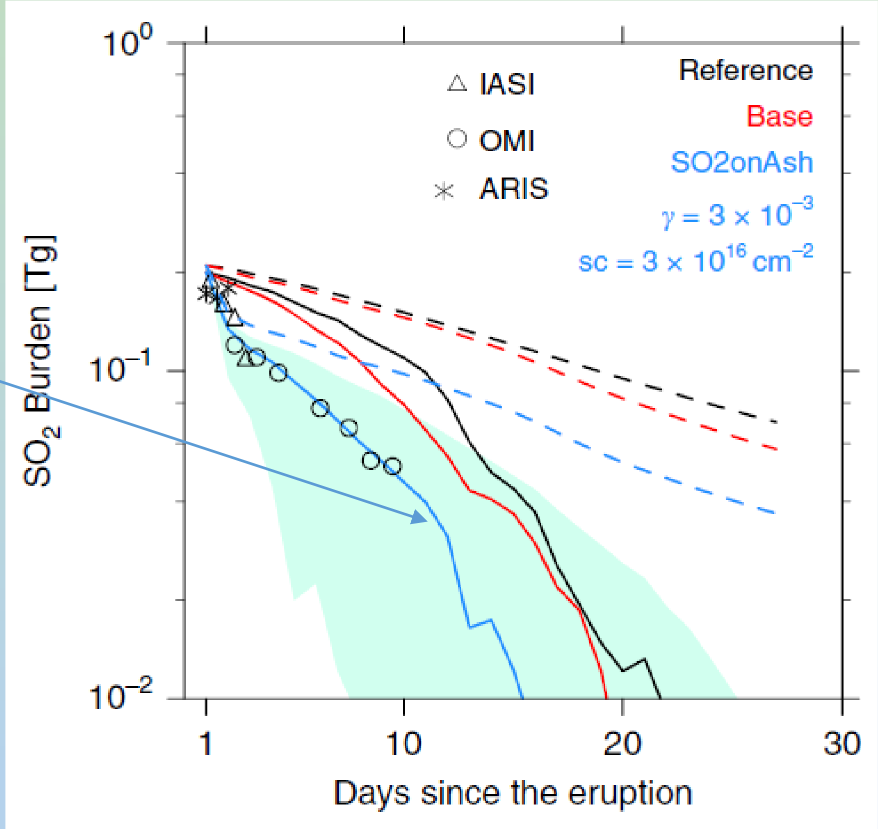
Chemical role of ash

Kelud volcanic plume (2014)

Above Guam Island
16.5-18.5 km, March 2014



With SO_2 uptake on ash



Zhu et al., Nature Comm., 2020

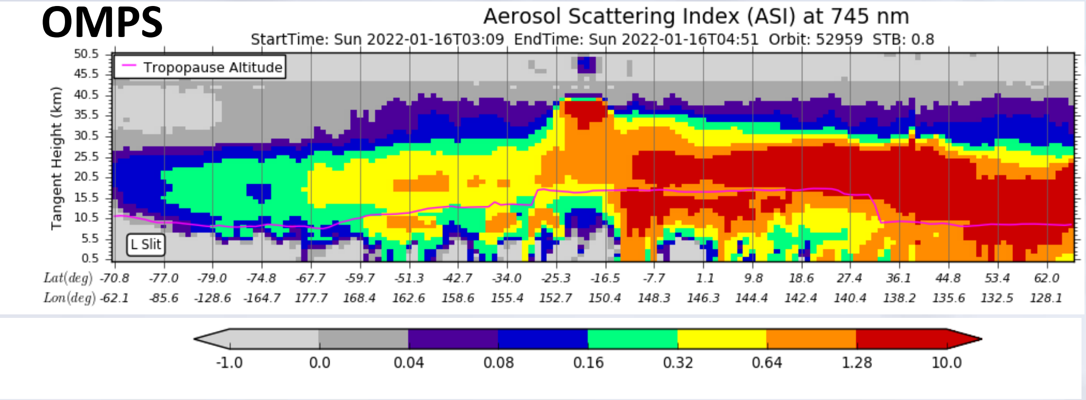
-> Ash tends to decrease SO_2 lifetime

Impact of Hunga Tonga eruption (January 2022)
La Réunion island

LOAC V1.5 flights from La Réunion island under alert

Support: HEMERA and VOLTAIRE project (Univ. of Orleans)

OMPS



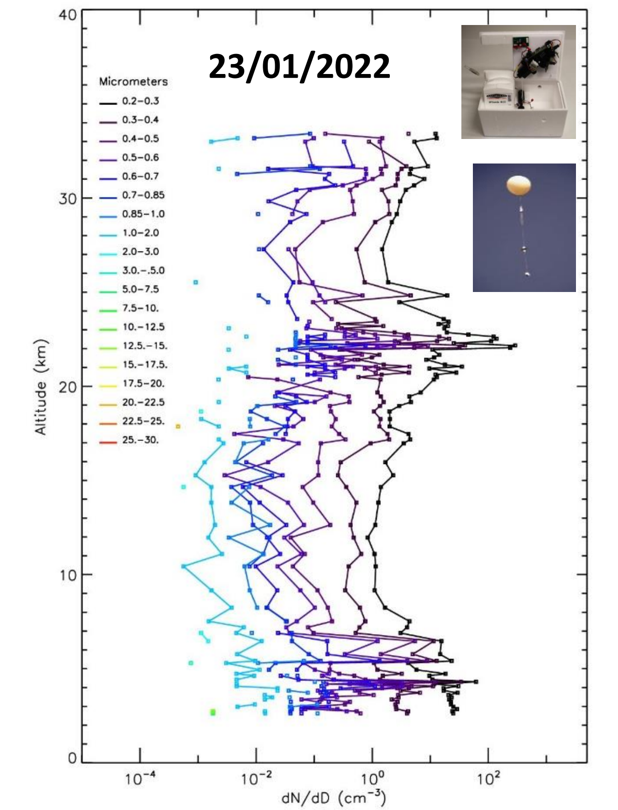
LOAC V1.5

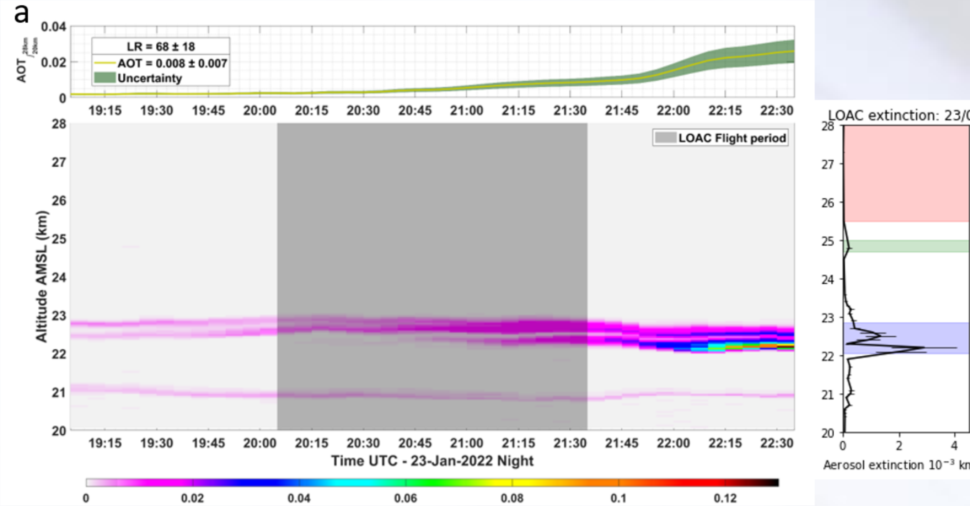
https://la1ere.francetvinfo.fr

© 2022 KMA/Simon Proud/NCEO

Eruption with unique characteristics:

-Injection **above 40 km** (signal at **50 km**!)
-Injection of **ash** , **halogens**, huge quantities of **water vapour** with very strong impact on sulfur chemistry, production and evolution of the aerosols, and stratospheric ozone chemistry



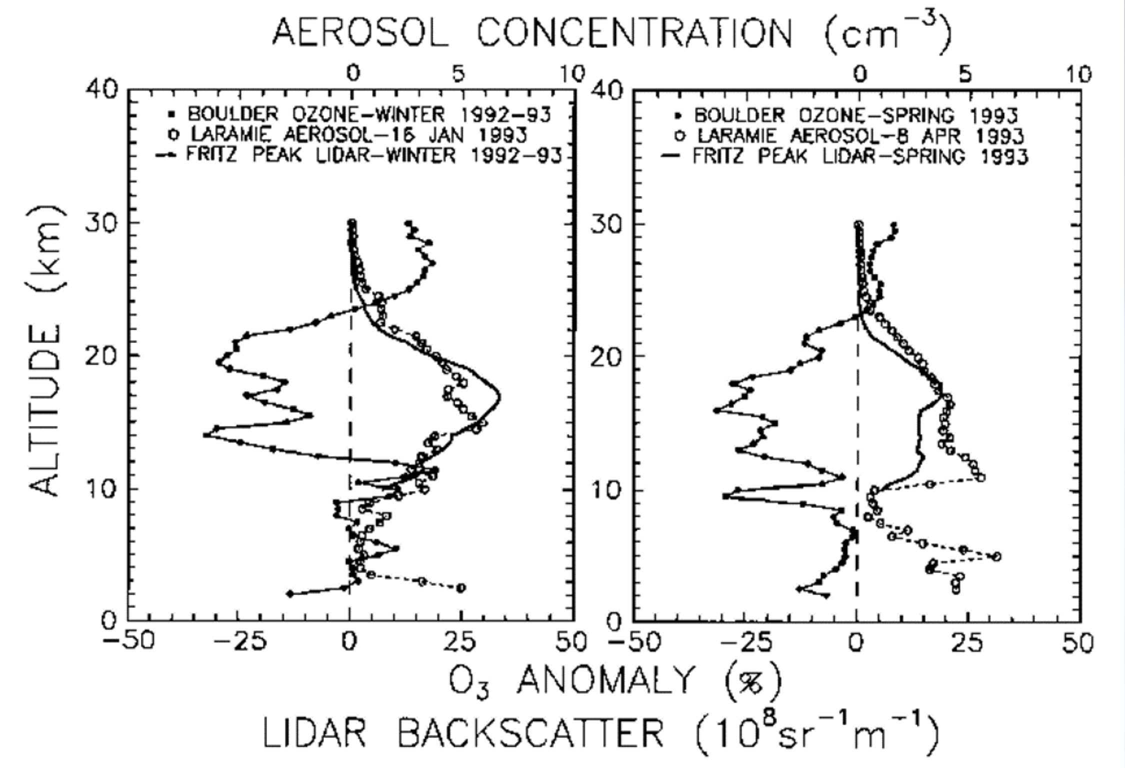


Optically-absorbing features of the small particles

Kloss et al., under revision, 2022

Ozone loss at mid-latitude after the Pinatubo 1991 eruption

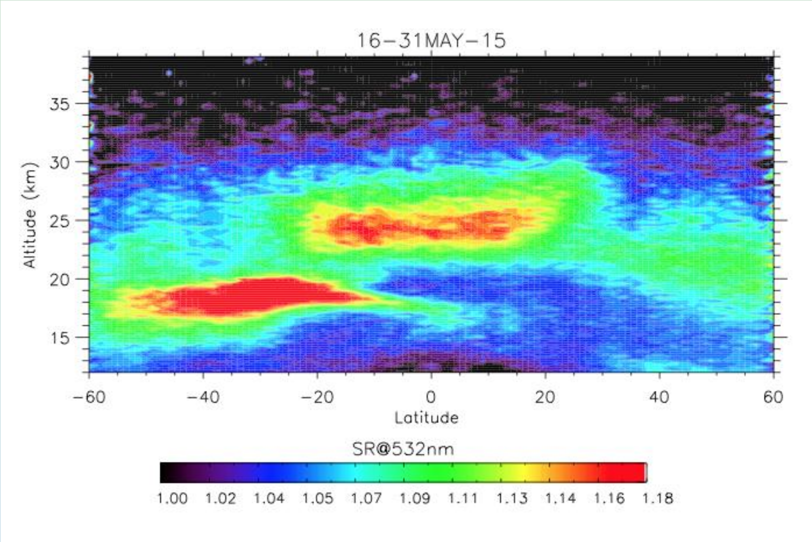
~20 Tg of SO_2 injected



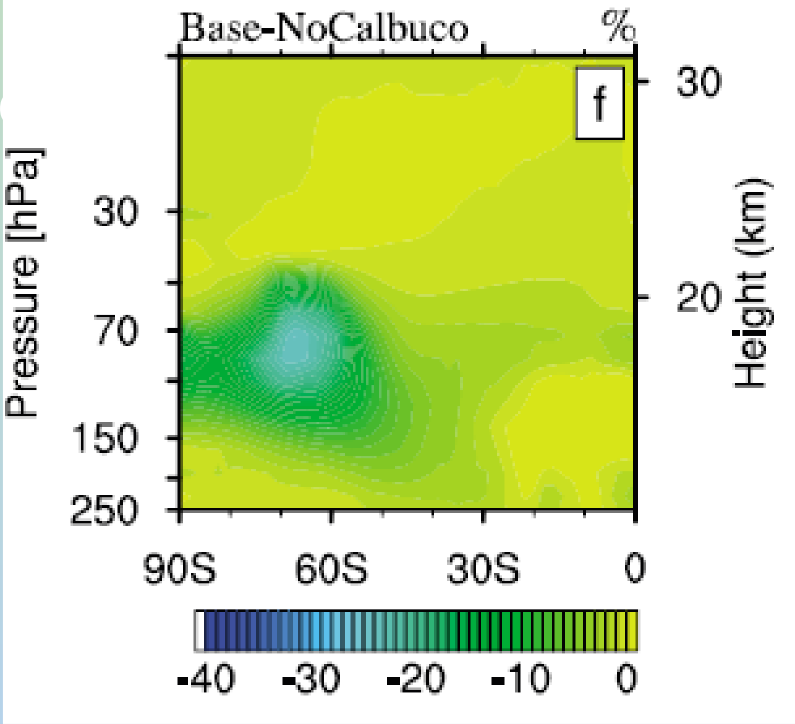
Hoffmann et al. GRL, 1994

Volcanic aerosols from moderate eruptions: chemical impacts

Observations by CALIOP/Calipso
Scattering ratio (SR)



change %



Zhu et al., JGR, 2018

Impact of Calbuco volcano aerosols

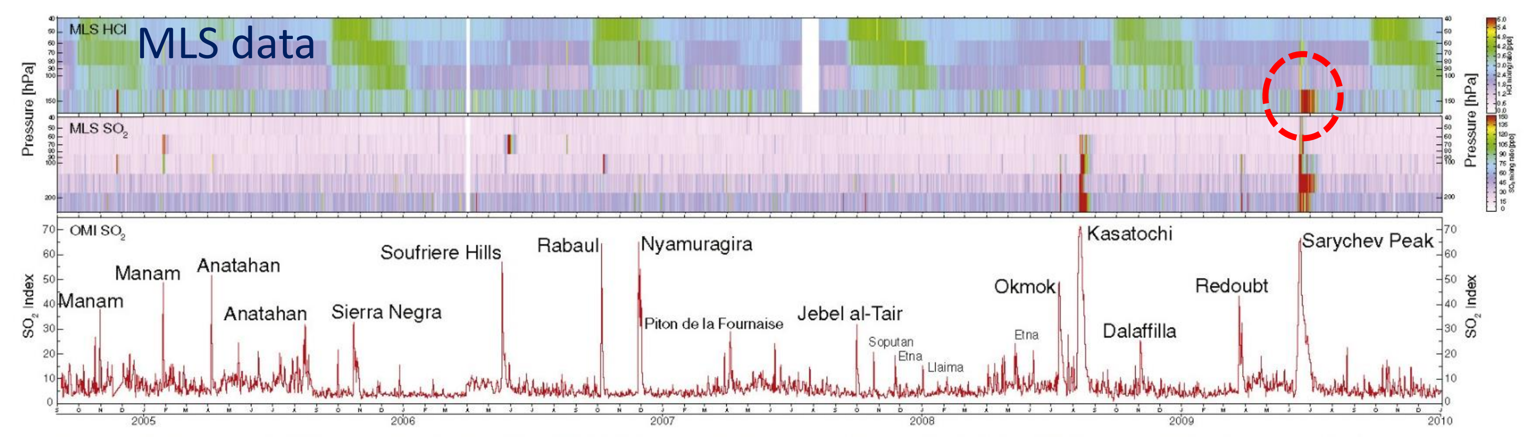
CESM1(WACCM)-CARMA model

Antarctic polar vortex

15-30 September 2015

→ **~25% of further ozone depletion (zonal mean) due to volcanic aerosol presence in the Antarctic polar vortex**

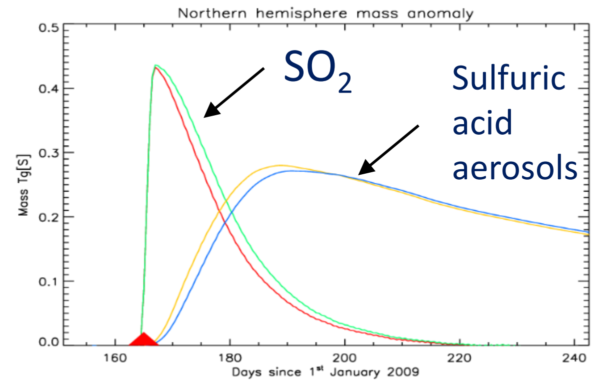
Co-injection of halogens



Northern hemisphere

HCl injection (27 Gg)

Carn et al., J. of Volc. And Geoth. Res., 2016



Sarychev eruption

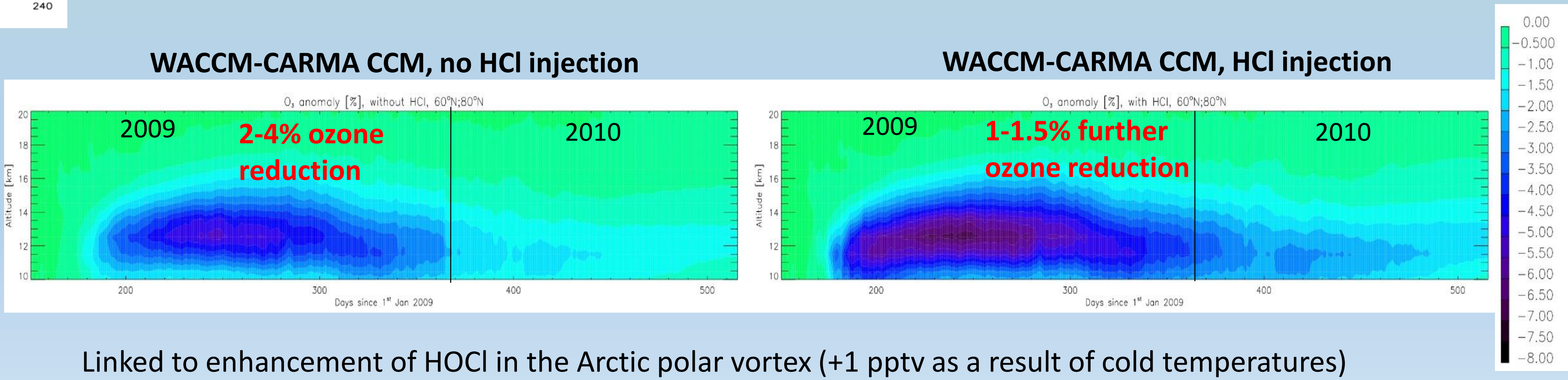
HCl injection → Slow down in the oxidation of SO_2 and sulfate aerosol formation: ~2-day delay

Combined effects of:

$HCl + OH \rightarrow Cl + H_2O$

$SO_2 + OH + M \rightarrow HSO_3 + M \ldots \rightarrow H_2SO_4 + H_2O$

Lurton et al., ACP, 2018


WACCM-CARMA CCM, no HCl injection

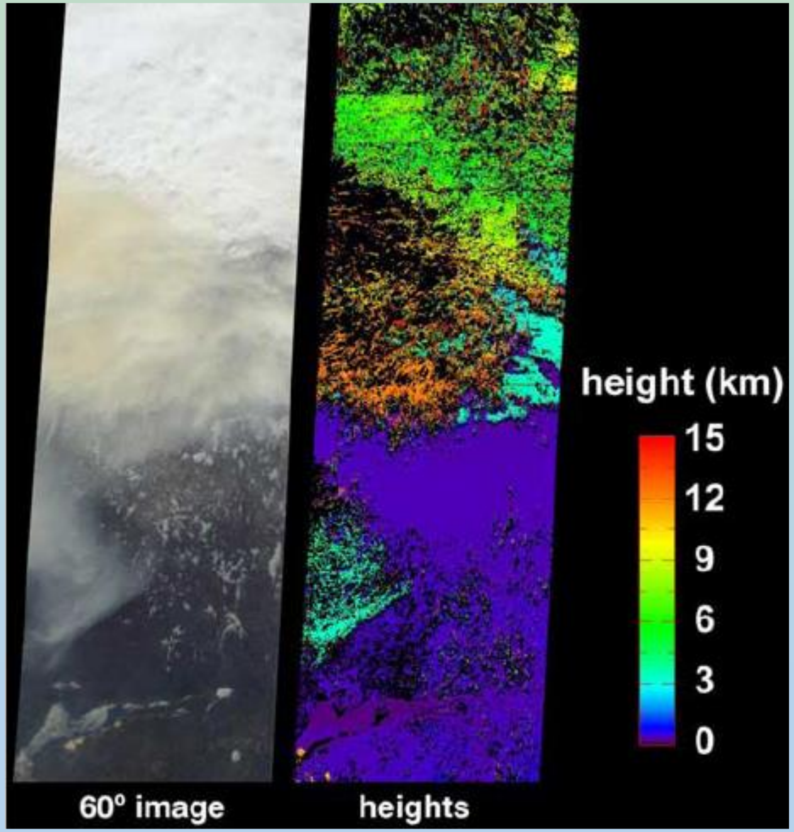
WACCM-CARMA CCM, HCl injection

Linked to enhancement of HOCl in the Arctic polar vortex (+1 pptv as a result of cold temperatures)

FIRE SMOKE

Regular Pyrocumulonimbus (PyroCb) injections in the UTLS region

MISR images May 2001,
Alberta, Canada

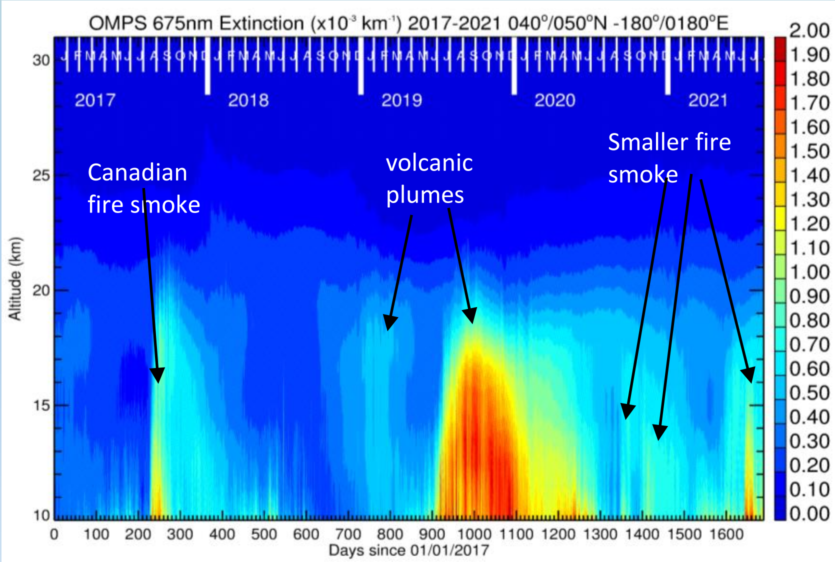


Fromm et al., JGR, 2008

See *Fromm et al., BAMS, 2010* for a review

For example (strong events):

Norman Wells pyroCb, August **1998**, NW Canada
Chilsom fire, May **2001**, Alberta, Canada
PyroCbs of January **2003**, SE Australia
Black Saturday, February **2009**, Australia
Canadian fires, August **2017**
Australian bushfires, December **2019-**January **2020**

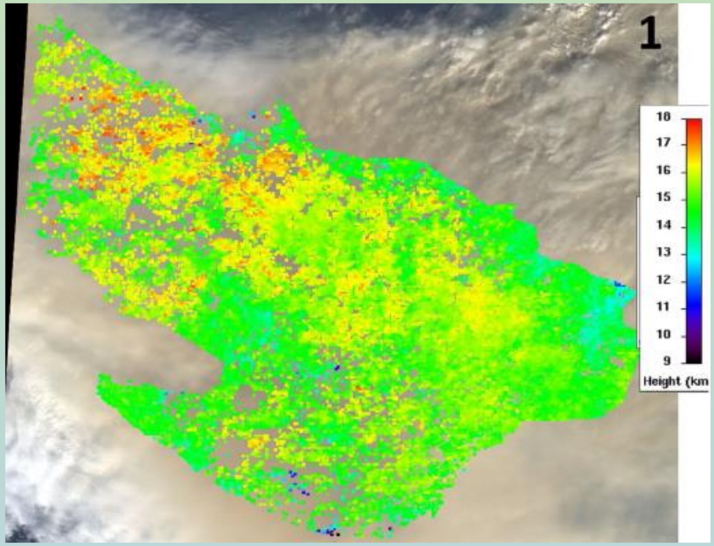


OMPS midlatitudes

+ smaller smoke injection events

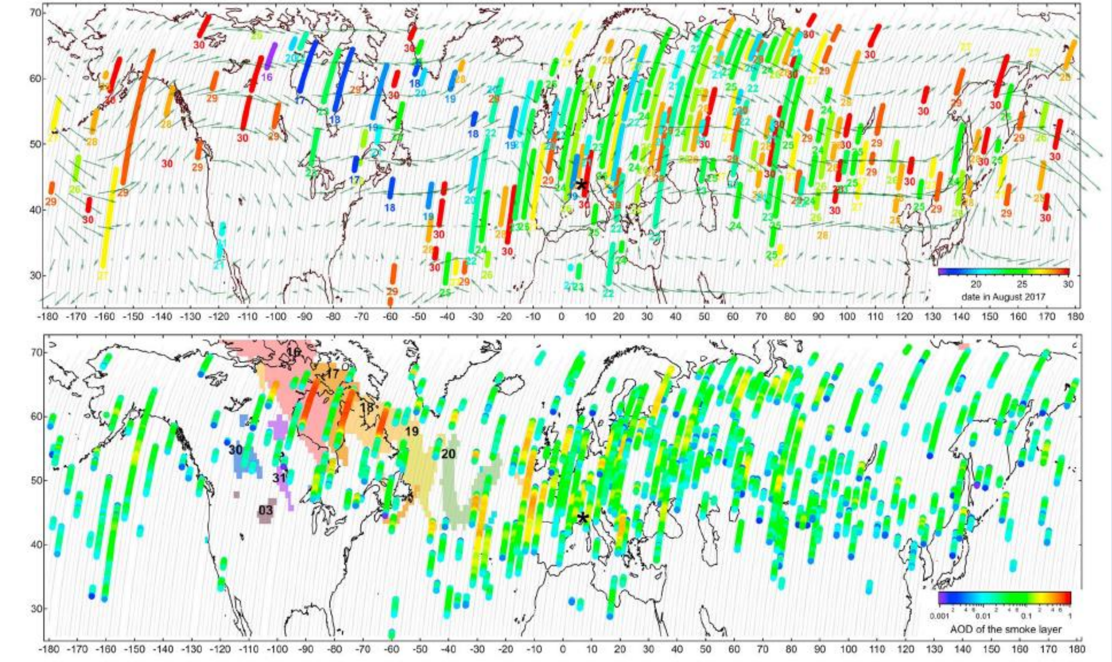
Plume transport: example of the Canadian fires, August 2017

MISR red-band data
Source region



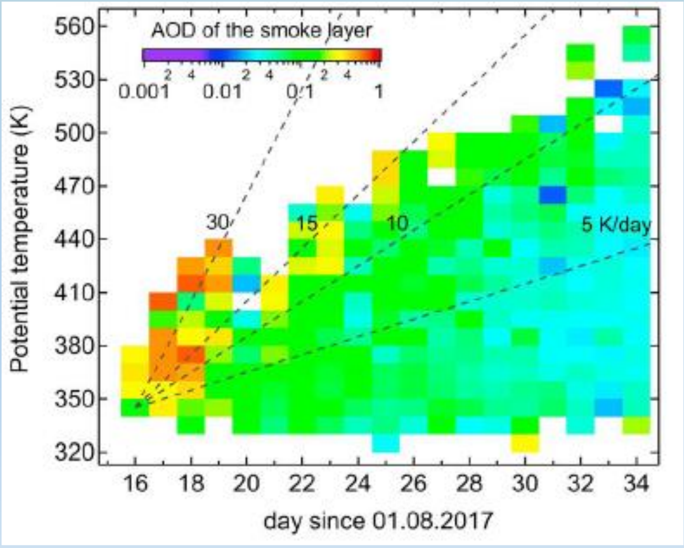
Fromm et al., JGR, 2021

CALIOP/Calipso



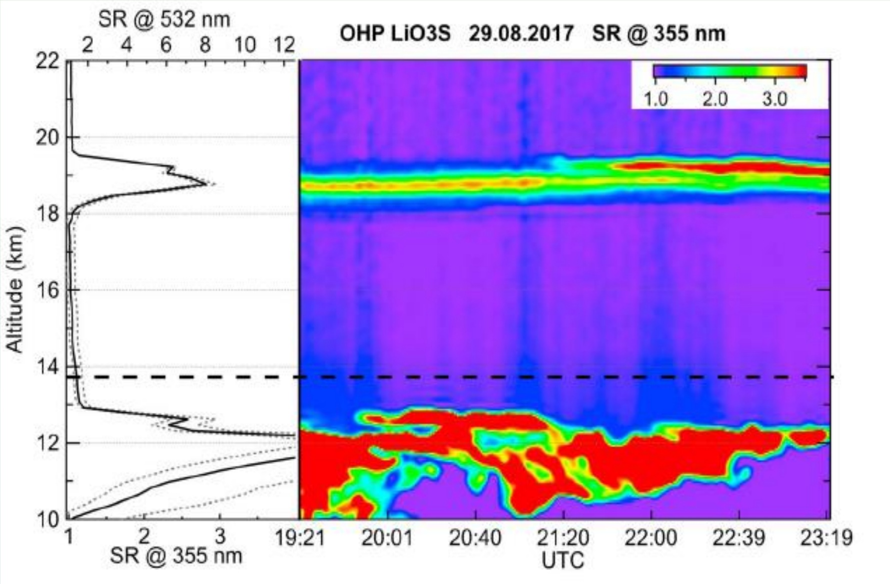
Khaykin et al., GRL, 2018

Plume rise consistent with localized heating due to the radiative absorption by the smoke particles

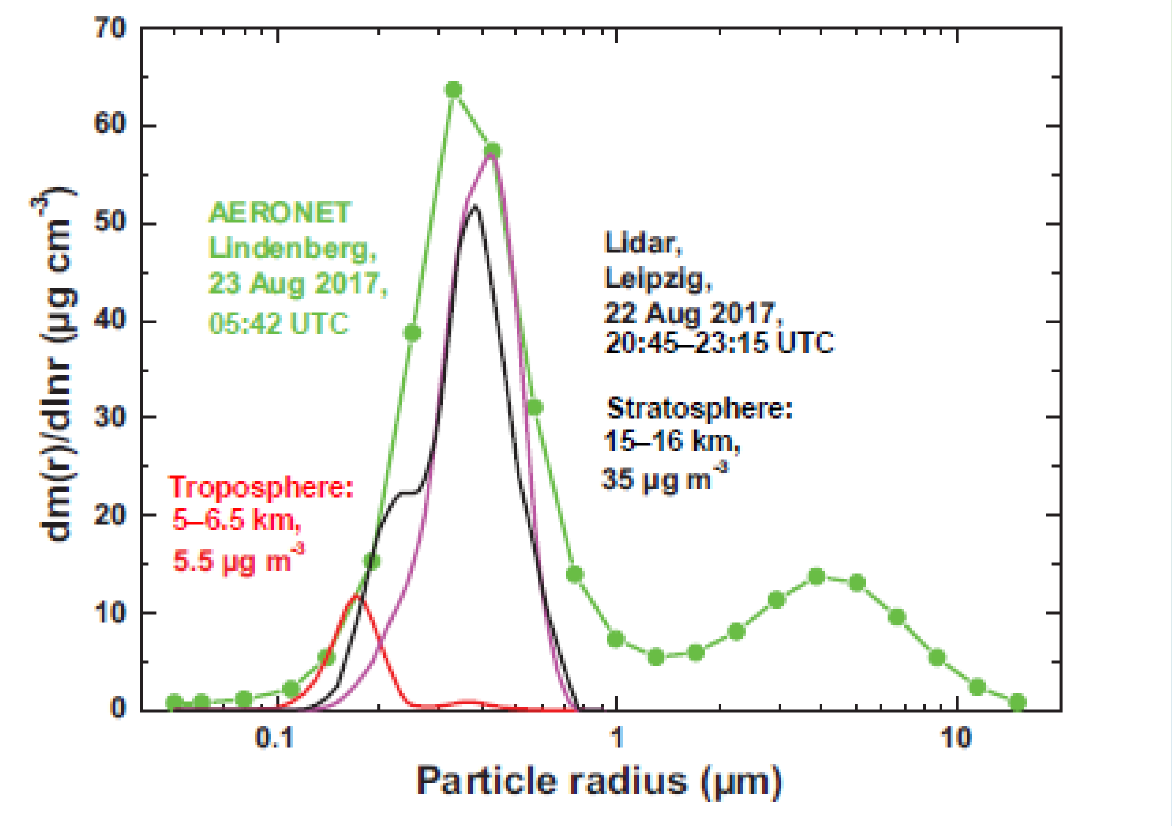


Information from ground-based observations

OHP lidar, France, 29/08/2017



Khaykin et al., GRL, 2018



Haarig et al., ACP, 2018

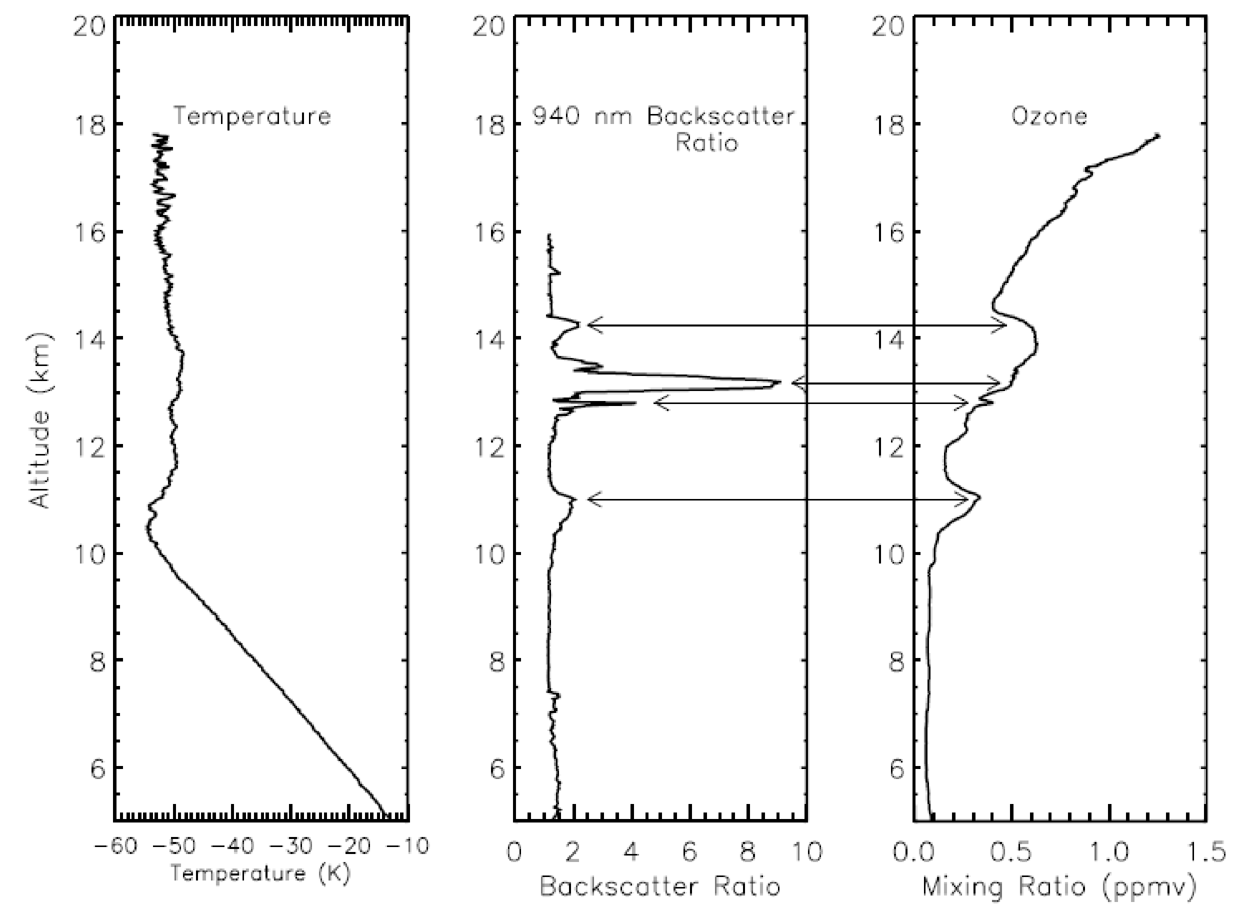
-High depolarization ratios of 22% at 355 nm and 18% at 532 nm

-Pronounced accumulation mode (particle mass) centered at a particle radius of 0.35–0.40 µm.

-Effective radius : 0.32 µm (stratosphere), 0.17 µm (troposphere)

-Mass concentrations of ~5.5 µg.m-3 (tropospheric layer) and ~40 µg.m-3 (stratospheric layer) on 22/08/2017

Impact on ozone



Ozone production

depending on the levels of NOx, VOCs, Hox

But what is the detailed chemistry?

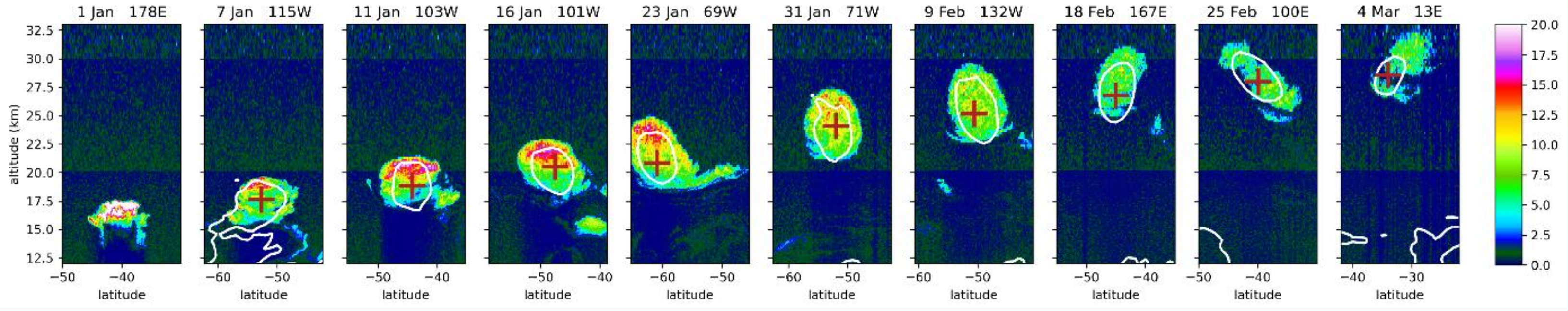
August 1998, Canada
(52°N; 107°W)

Fromm et al., JGR, 2005

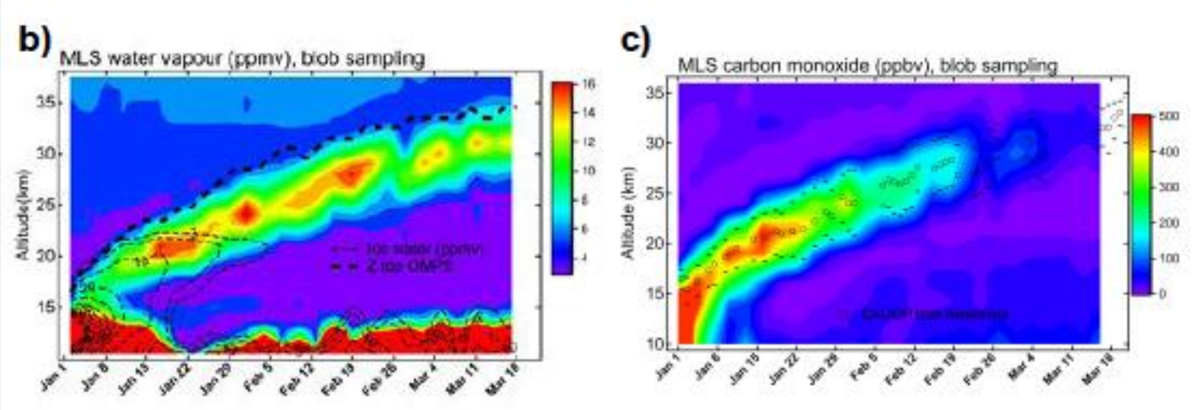
Self-maintained anticyclonic vortex of smoke particles

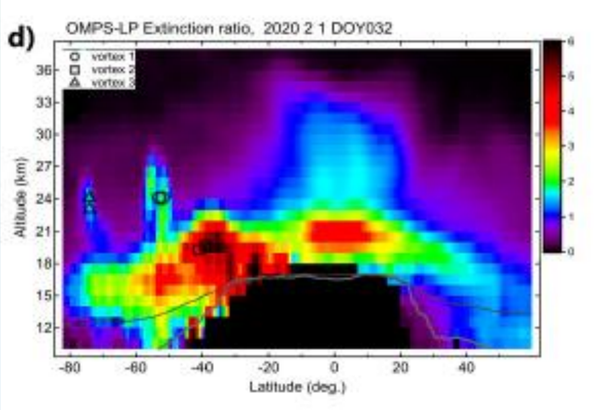
Australian fires in 2019-2020 but also observed for the Canadian fires in 2017 (see *Lestrelin et al., ACP, 2021*)

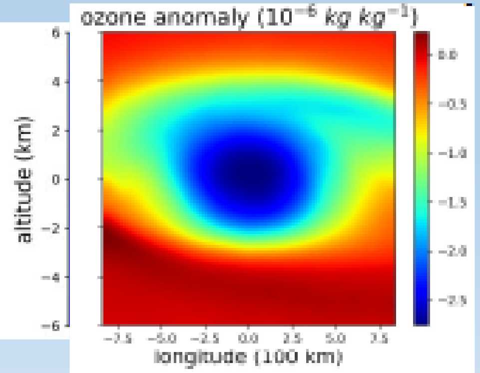
532nm scattering ratio of the CALIOP lidar



1,000 km diameter
Persistence over 13 weeks, 66,000 km travel
Confined bubble of smoke and moisture
+ shorter-lived companions







Time-averaged composite section of the ozone anomaly (ECMWF analysis)

-> deep mini ozone hole, depleted by up to 100 DU

Khaykin et al., Communic. Earth&Envir., 2020

Conclusions

- Global stratospheric aerosol burden largely modulated by volcanic eruptions and by wildfires (which have tended to increase over the recent years) but other particles types have been reported (e.g. organics in the tropical lower stratosphere)

- Detection of the ATAL aerosol layer from in situ observations confined in the summer monsoon anticyclone confirming the observations by satellite instruments. → mainly small particles

- The amplitude of the impact of volcanic eruptions critically depend on the latitude and altitude of injection, on the amount of SO_2 and on the timing of the eruption. Monomodal size distributions for moderate volcanic eruptions. Important role of ash particles which perturb the kinetics of aerosol formation and possibly plume dispersion.

- Volcanic aerosol plumes quite well captured by global models. Uncertainties mainly resulting from differences in resolution and uncertainties in the knowledged of injection parameters.

- Fires: difficult to make conclusions regarding the generality of smoke vortice structures and what their global impact may be. Compact and vortex-like structures possibly reported after the 2019 eruption of Raikoke (Chouza et al., 2020).

- However, comparisons between the model and in situ observations show contrasted results and not systematically consistent between model and satellite observations.

  → Difficult to draw robust conclusions if comparisons are not statistically significant.

- No reference instrument for in situ observations of aerosol concentrations and size distributions. Simultaneous observations of different optical particle counters needed and initiated by HEMERA.

- OPC observations not sufficient. Need for chemical composition characterization. Ongoing cooperation with NASA-Langley using new balloon-borne aerosols collectors (JP Vernier)