Stratospheric chemistry and aerosol modeling in CAMS with the IFS-CB05-BASCOE-GLOMAP (ICBG) system

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Volcanic simulation: coupling of IFS-CB05-BASCOE with IFS-GLOMAP

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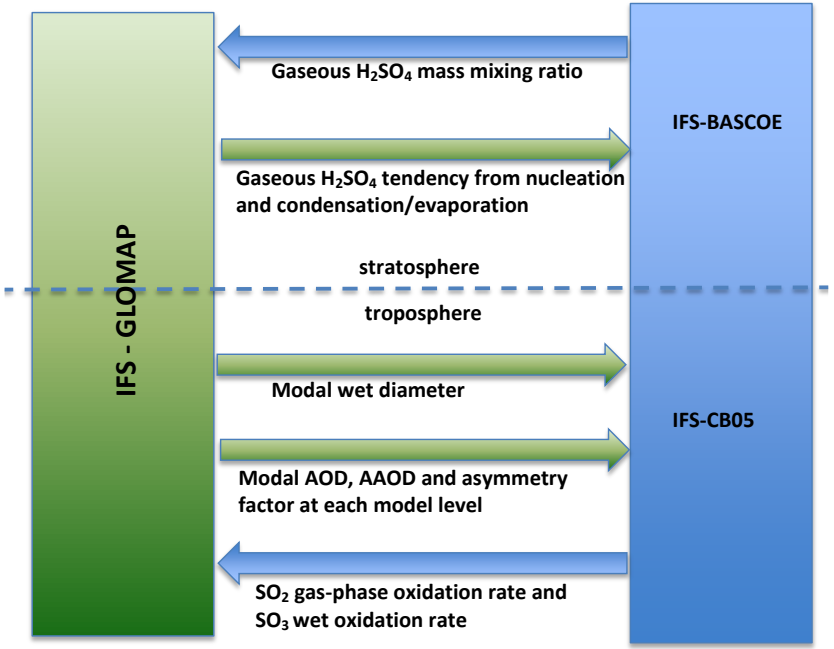
Original ICBG configuration (2019)

- Stratospheric sulphur cycle has been implemented in IFS-CB05-BASCOE
- Stratospheric coupling of IFS-CB05-BASCOE with IFS-GLOMAP
  - Sulphuric acid from IFS-BASCOE
  - Sulphuric acid tendencies from nucleation and condensation from IFS-GLOMAP
- Tropospheric coupling of IFS-CB05 with IFS-GLOMAP
  - SO_x oxidation rate from IFS-CB05
  - Aerosol wet diameter from IFS-GLOMAP to compute Surface Area Density (SAD) for heterogeneous chemical reactions
  - Aerosol optical properties to compute photolysis rates



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Volcanic simulation: coupling of IFS-CB05-BASCOE with IFS-GLOMAP

Stratospheric sulfur cycle has been implemented in IFS-CB05-BASCOE

Reaction	Rate expression
$OCS + O \rightarrow CO + SO_2$	$2.1 \times 10^{-11} \exp(-2200/T)$
$OCS + OH \rightarrow CO_2 + SO_2$	$1.1 \times 10^{-13} \exp(-1200/T)$
$SO_2 + OH \rightarrow SO_3 + HO_2$	Third-body reaction (3.3×10^{-31}, 4.3, 1.6×10^{-12}, 0.)
$SO_2 + O_3 \rightarrow SO_3$	$3.0 \times 10^{-12} \exp(-7000/T)$
$SO_3 + H_2O \rightarrow H_2SO_4$	$8.5 \times 10^{-41} \exp\left(-\frac{6540}{T}\right) \cdot [H_2O]$

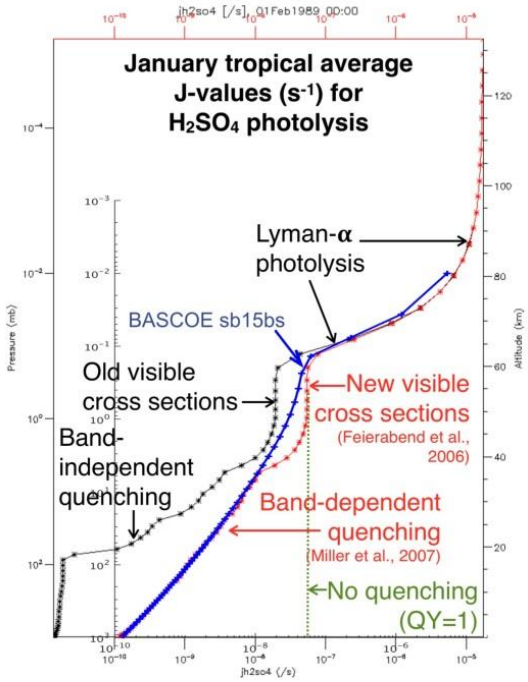
Tropospheric sulfur cycle has been modified as well:

Reaction	Rate expression
$OCS + OH \rightarrow CO_2 + SO_2$	$1.1 \times 10^{-13} \exp(-1200/T)$
$SO_2 + OH \rightarrow SO_3 + HO_2$	Third-body reaction (3.3×10^{-31}, 4.3, 1.6×10^{-12}, 0.)
$SO_2 + O_3 \rightarrow SO_3$	$3.0 \times 10^{-12} \exp(-7000/T)$
$SO_3 + H_2O \rightarrow$ particulate SO_4	$8.5 \times 10^{-41} \exp\left(-\frac{6540}{T}\right) \cdot [H_2O]$

Volcanic simulation: coupling of IFS-CB05-BASCOE with IFS-GLOMAP

Additional photolysis reactions in IFS-BASCOE:

Photolysis	Rate expression
$OCS + hv \rightarrow CO + SO_2$	(Burkholder et al. 2015)
$H_2SO_4 + hv \rightarrow SO_3 + H_2O$	(see below)
$SO_3 + hv \rightarrow SO_2 + O$	(Burkholder et al. 2015)



Intercomparison of H_2SO_4 photolysis as implemented in BASCOE (blue) against Miller et al. (2007) and Feierabend et al (2006) (red), as implemented in WACCM.

Volcanic simulation: coupling of IFS-CB05-BASCOE with IFS-GLOMAP

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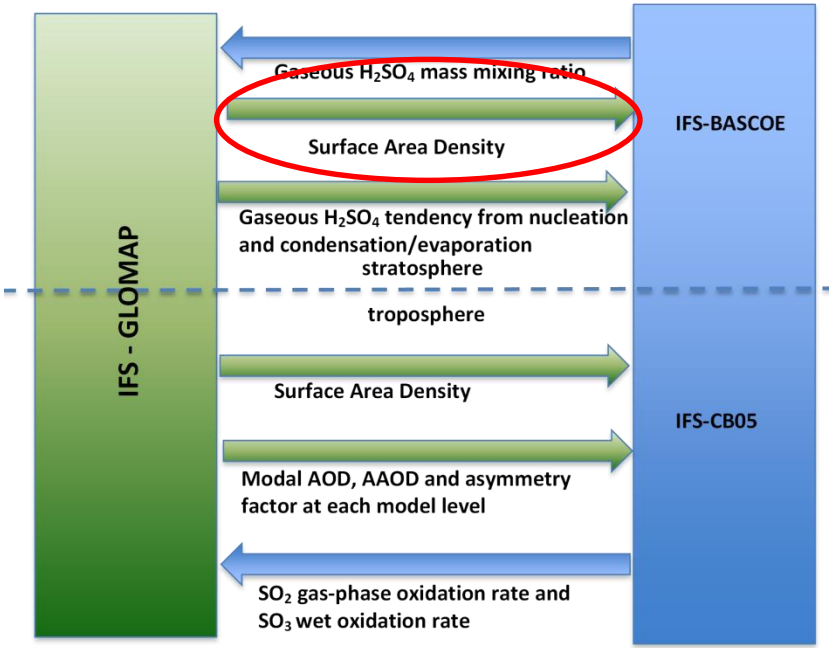
Recent updates of the ICBG system

- Use of Surface Area Density (SAD) from IFS-GLOMAP to estimate the rate of heterogeneous reactions in IFS-BASCOE
- Use of a climatological replenishment of Meteoric Smoke Particles (MSP): stratospheric sulfate particles also form from MSP.



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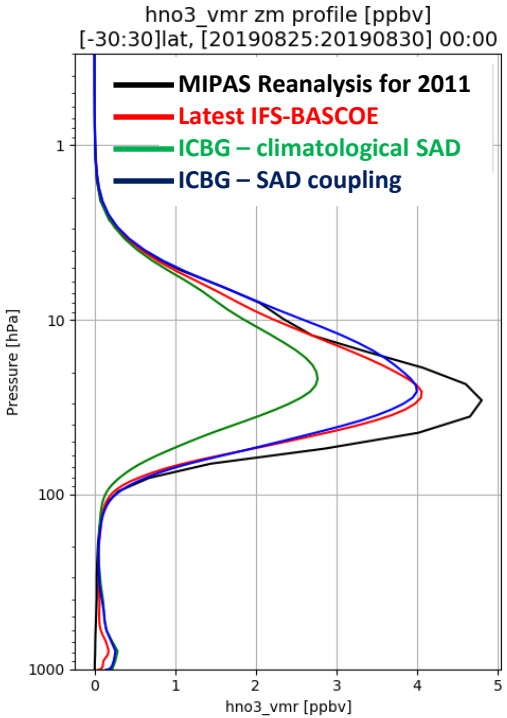
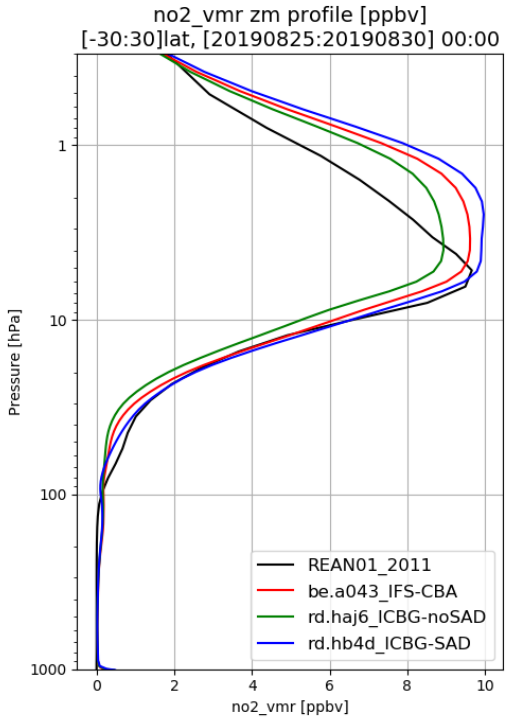
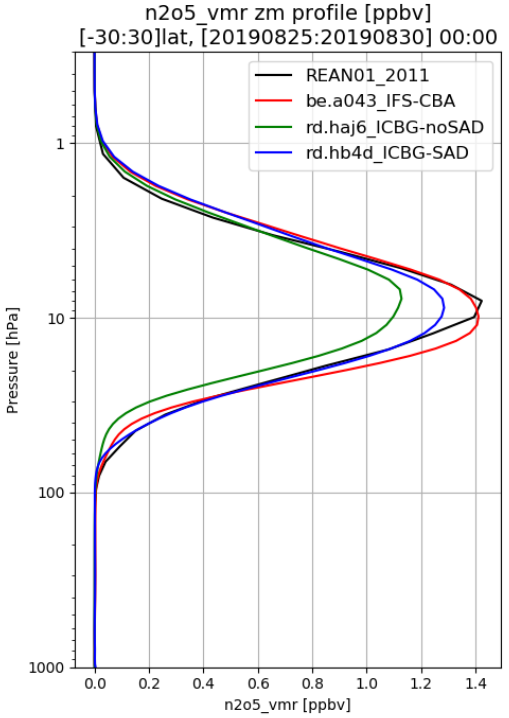
Impact of SAD from IFS-GLOMAP on simulated stratospheric trace gases

The impact of using SAD from IFS-GLOMAP is significant







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ICBG – evaluation of simulated stratospheric sulfate

The ICBG system has been tested in quiescent conditions and on the following volcanic eruptions:

- Pinatubo (June 1991, ~14Tg SO_2 release)
- Raikoke (June 2019, ~1.5 Tg SO_2 release)

In quiescent conditions, sensitivity tests on the role of particulate evaporation onto sulphuric acid have been carried out.

The simulated total stratospheric particulate sulfate burden varies between 0.12 TgS (no evaporation) and 0.2 TgS (evaporation), which agrees quite well against estimates from SAGE: 0.16 TgS for Sage - 3λ and 0.12 TgS for Sage - 4λ .

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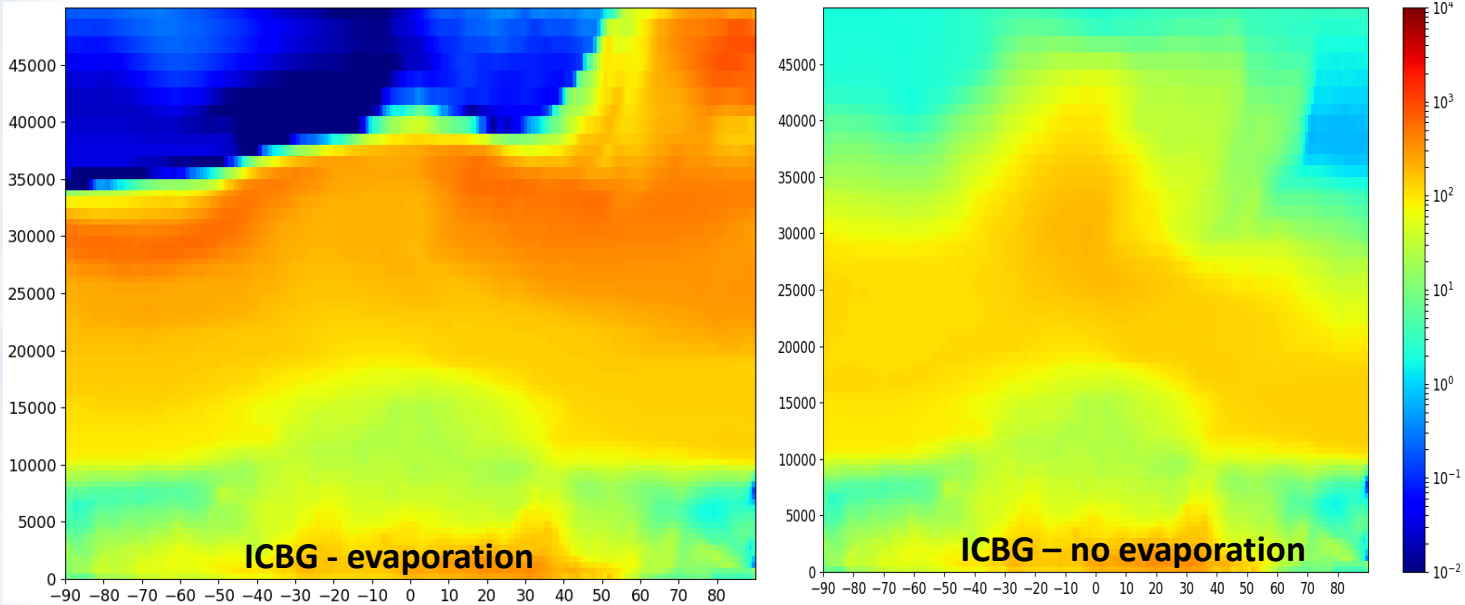
ICBG – evaluation of simulated stratospheric sulfate

The ICBG system has been tested in quiescent conditions: 1998-2002 simulations, without MSP replenishment. The impact of particulate sulfate evaporation is clear, especially in the higher stratosphere.



January 1999, vertical-zonal cross section of sulfate aerosol (accumulation soluble mode) volume mixing ratio in pptv

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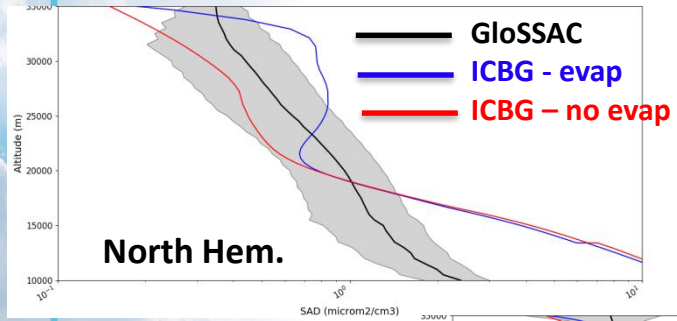
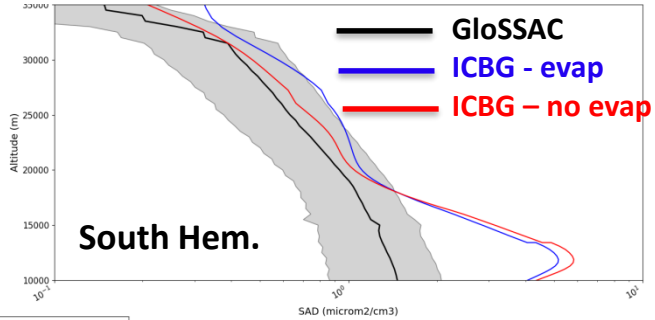
Atmosphere Monitoring

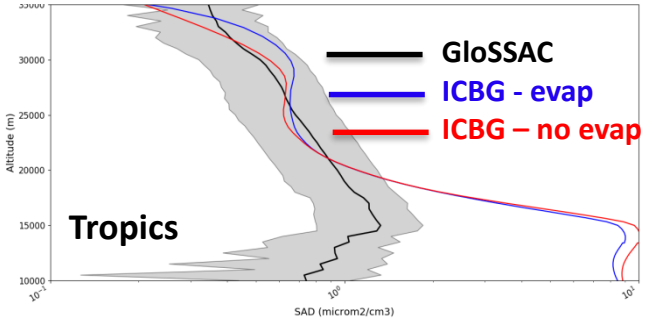
ICBG – evaluation of simulated stratospheric sulfate

The ICBG system has been tested in quiescent conditions: 1998-2002 simulations, without MSP replenishment

Evaluation of surface area density (SAD) against GloSSAC







June 1998, comparison of retrieved Surface Area Density from GloSSAC v2.0 (black, with standard deviation shaded gray) with simulated values from ICBG

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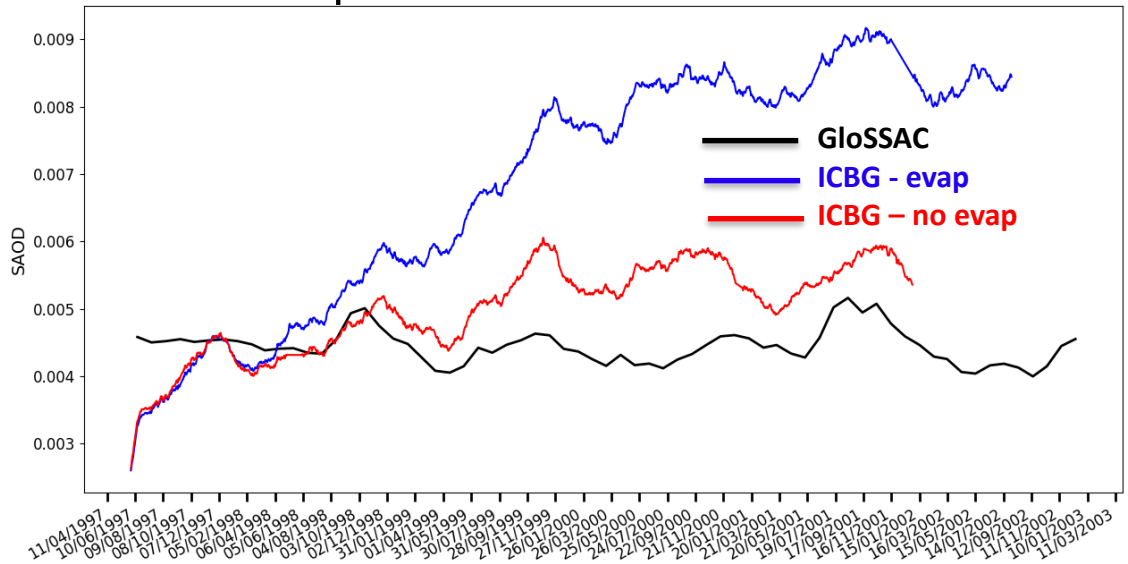
ICBG – evaluation of simulated stratospheric sulfate

The ICBG system has been tested in quiescent conditions: 1998-2002 simulations, without MSP replenishment

Evaluation of stratospheric AOD at 550nm against GloSSAC: overestimation with evaporation on.



Timeseries of monthly mean of mid-visible global-mean-strat-AOD (77.5˚S to 77.5˚N) from the 1998-2002 period within GloSSAC v1.1 (525nm) and from ICBG experiments (550nm).

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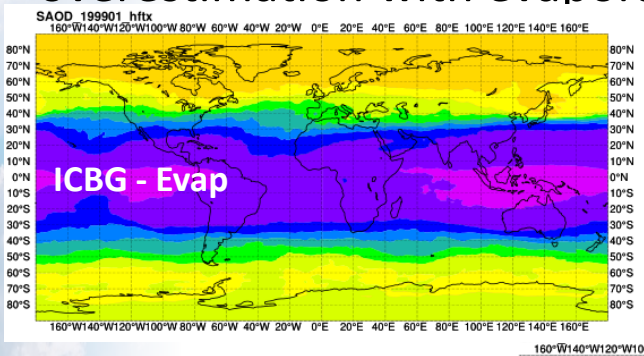
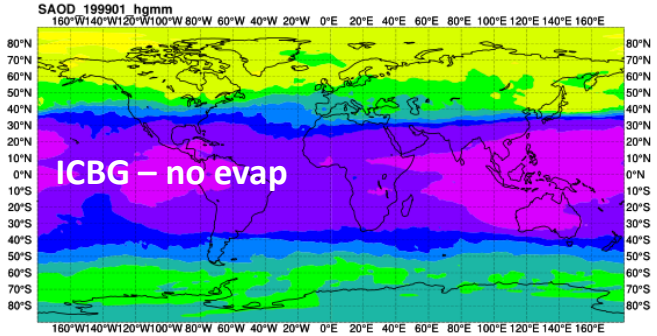
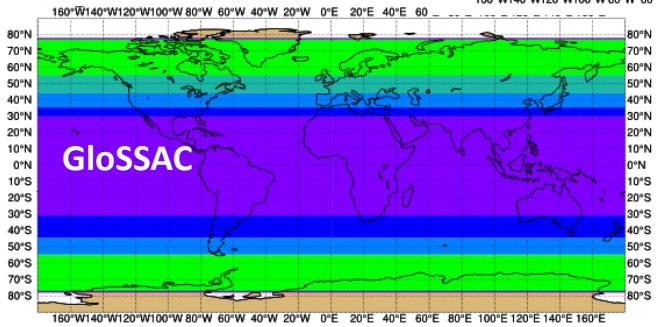
ICBG – evaluation of simulated stratospheric sulfate

The ICBG system has been tested in quiescent conditions: 1998-2002 simulations, without MSP replenishment

Evaluation of stratospheric AOD at 550nm against GloSSAC (January 99): overestimation with evaporation on.







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Simulation of Pinatubo with ICBG

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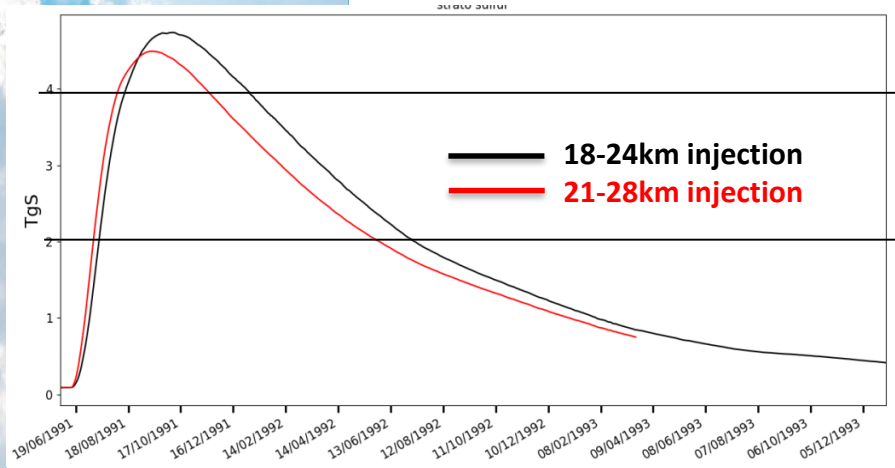
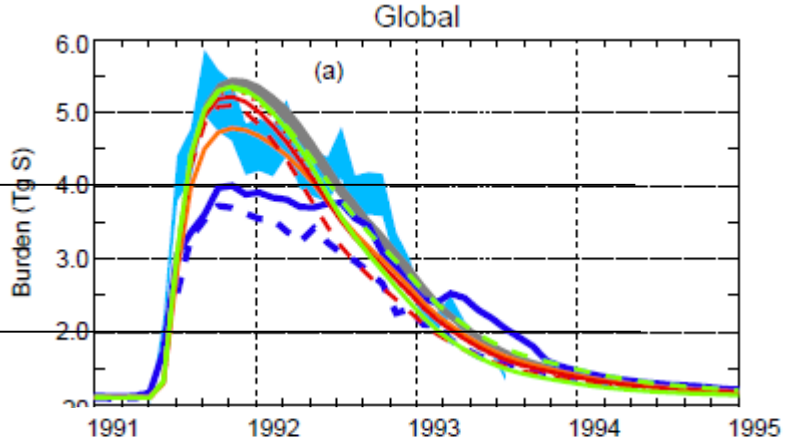
Simulation of the 1991 Pinatubo eruption with 7 TgS emitted :

- between 18 and 24km,
- between 21 and 28km

No interaction between radiation and aerosols



Stratospheric particulate sulfate from ICBG simulations



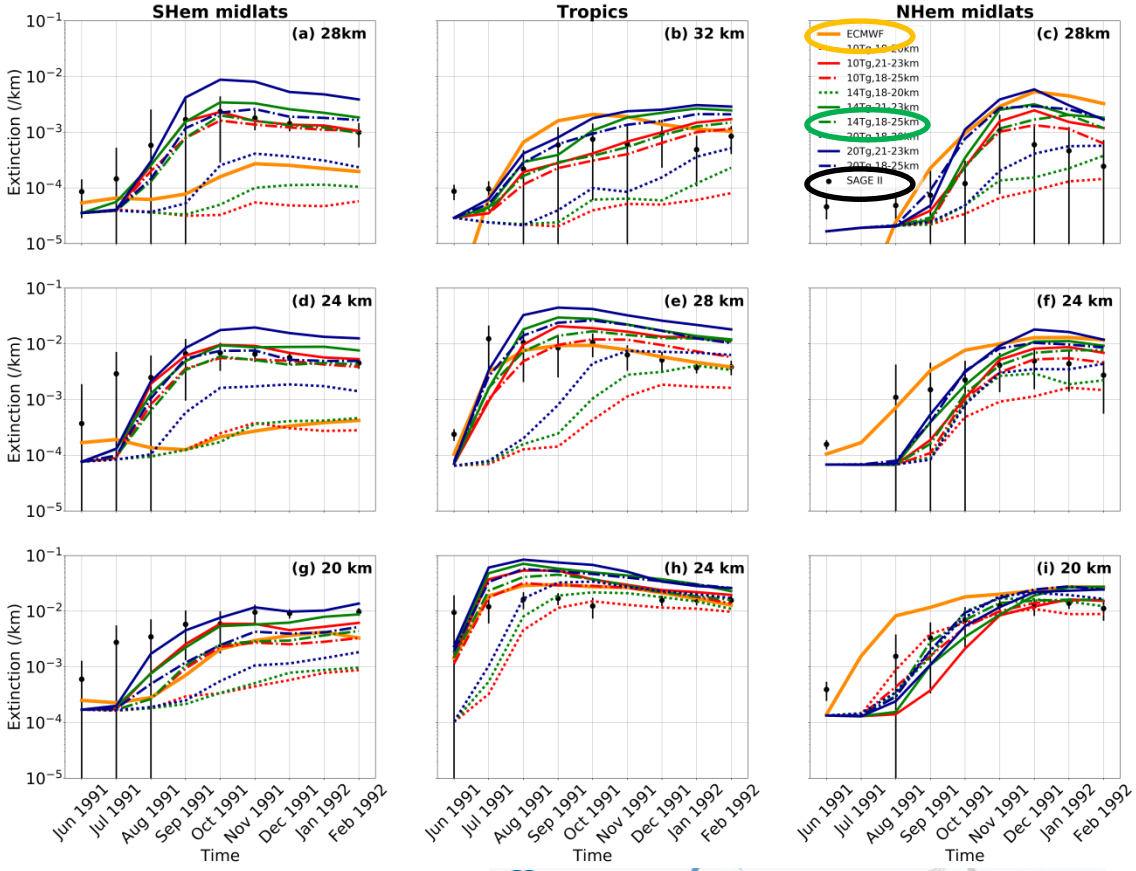
Stratospheric particulate sulfate from SOCOL-AER (Sukhodolov et al. (2018)). Light/dark blue = retrievals.

Simulation of Pinatubo with ICBG

Simulation of the 1991 Pinatubo eruption with 7 TgS emitted between 18 and 24km

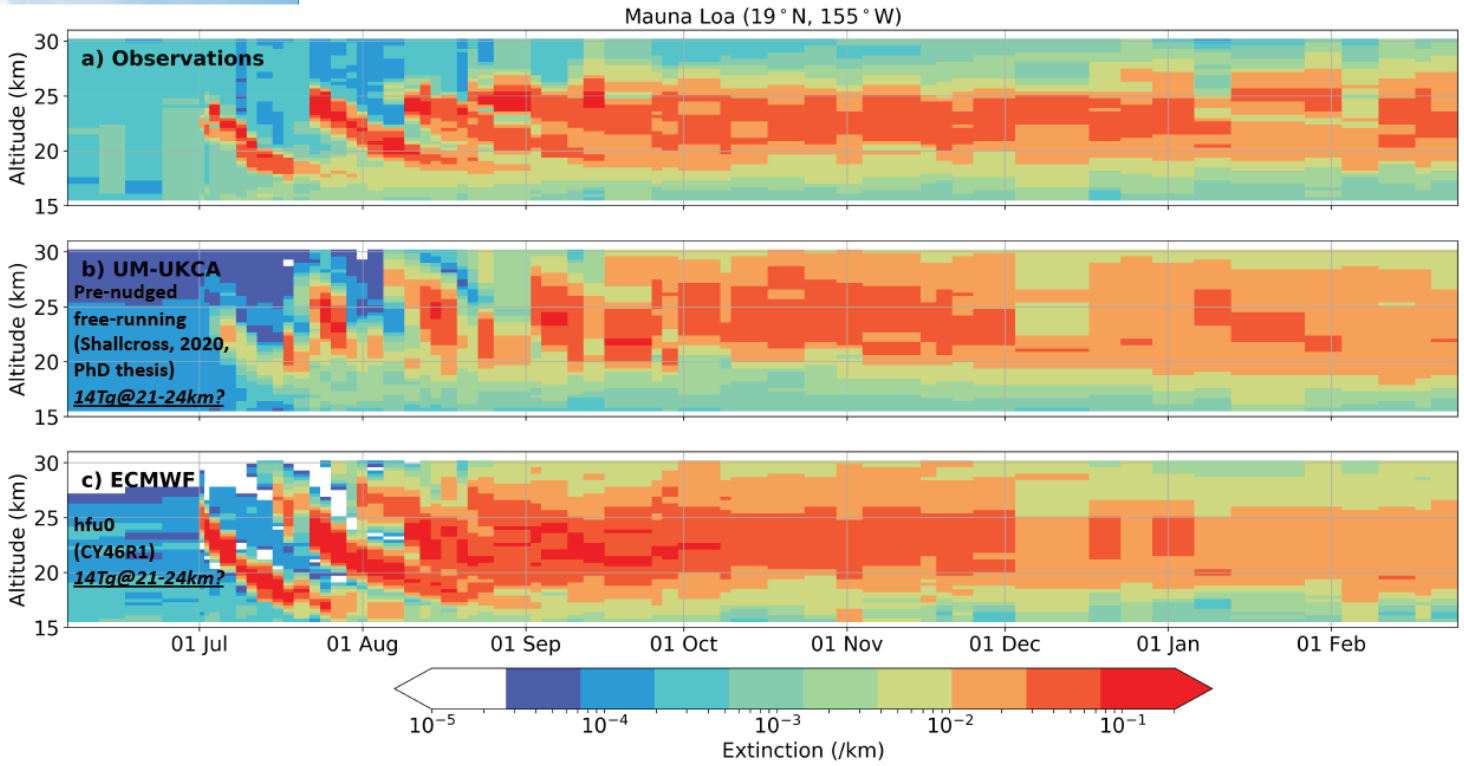
Comparison of ICBG simulation (**orange**) with UM-UKCA runs (most similar: **green dashed**) and SAGE-II obs (**black dots**).

UM-UKCA includes aerosol-radiation interaction; ICBG does not.



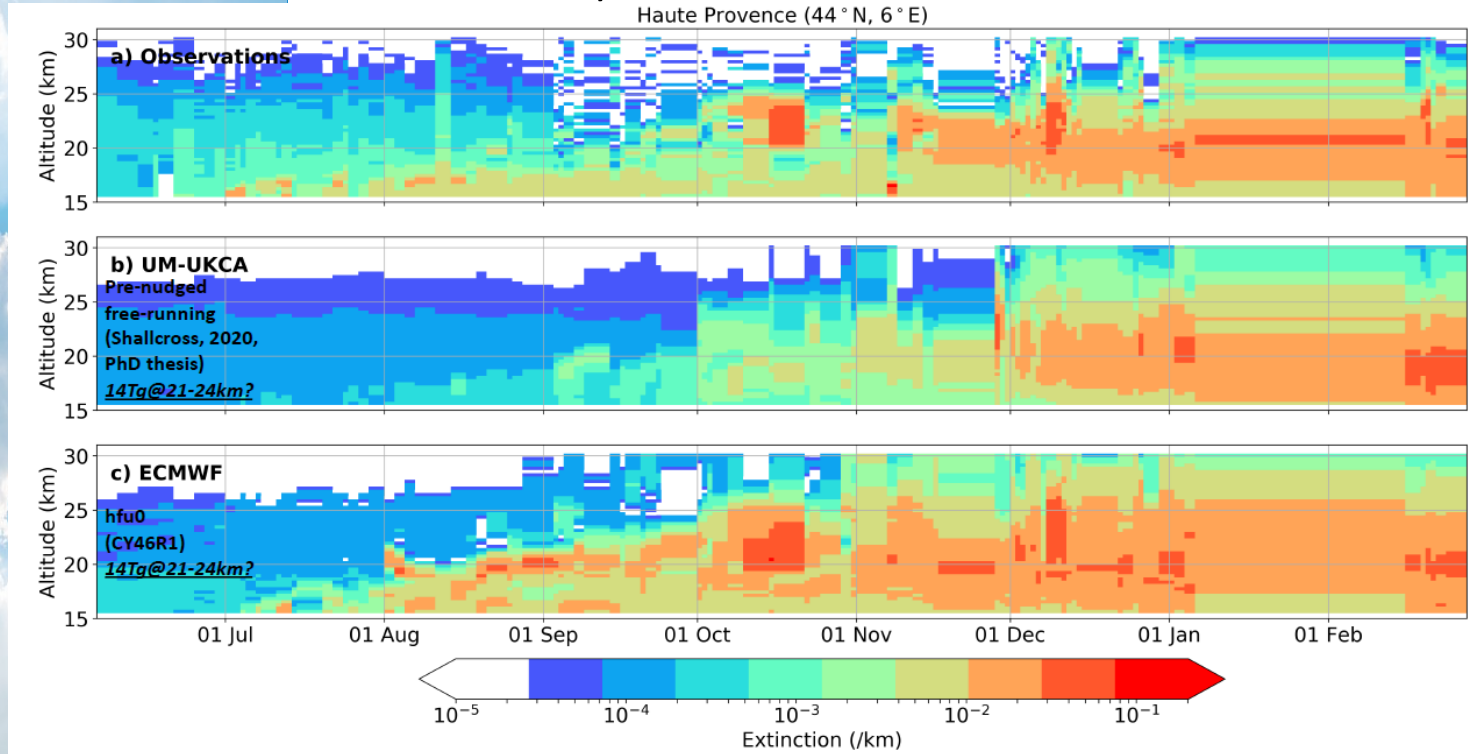
Simulation of Pinatubo with ICBG

Simulation of the 1991 Pinatubo eruption with 7 TgS emitted between 18 and 24km. Comparison of ICBG simulations with UM-UKCA runs and lidar data (Mauna Loa).



Simulation of Pinatubo with ICBG

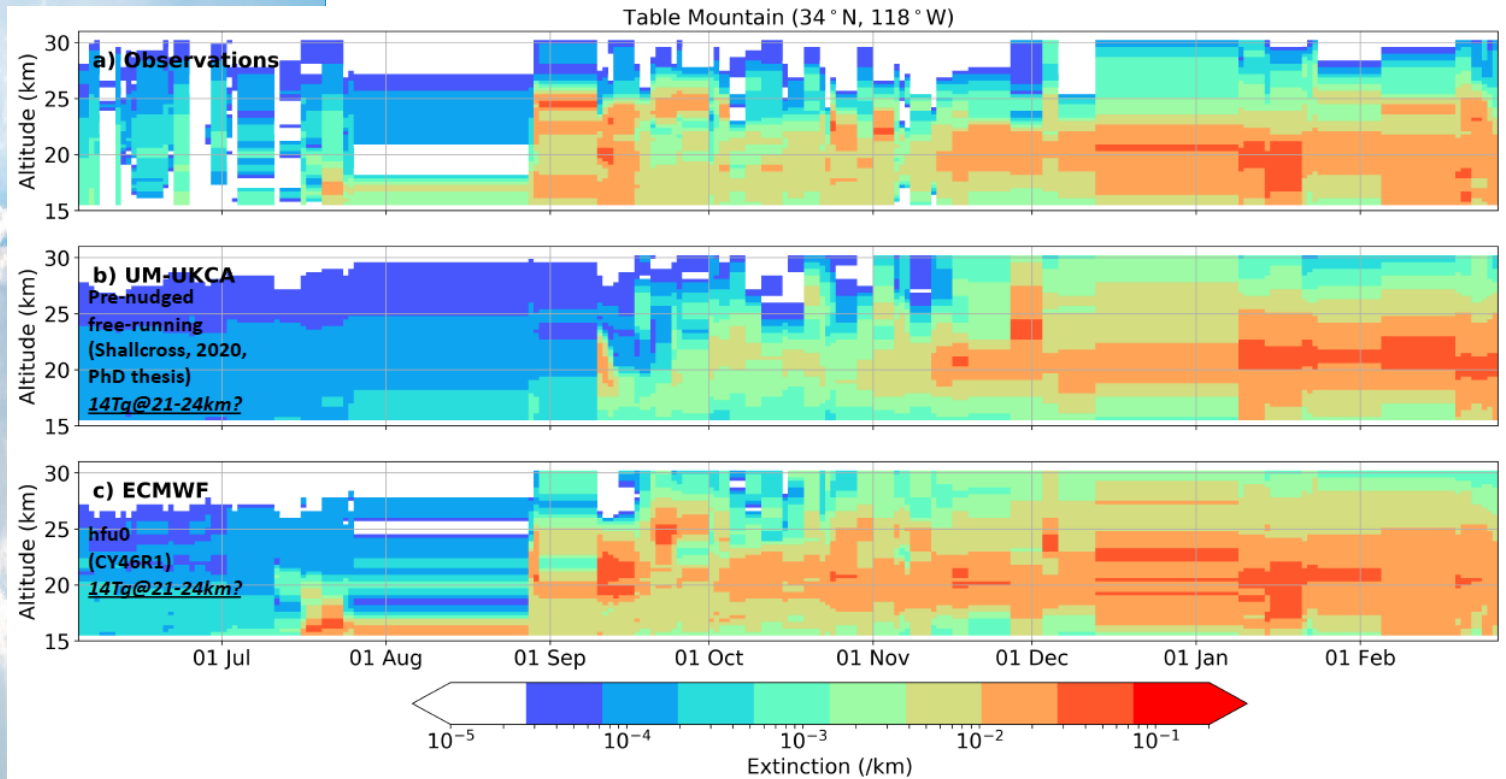
Simulation of the 1991 Pinatubo eruption with 7 TgS emitted between 18 and 24km. Comparison of ICBG simulations with UM-UKCA runs and lidar data (Observatoire de Haute Provence).



Simulation of Pinatubo with ICBG

Simulation of the 1991 Pinatubo eruption with 7 TgS emitted between 18 and 24km:

ICBG and UM-UKCA simulations versus LIDAR observations (Table Mountain, CA, USA)



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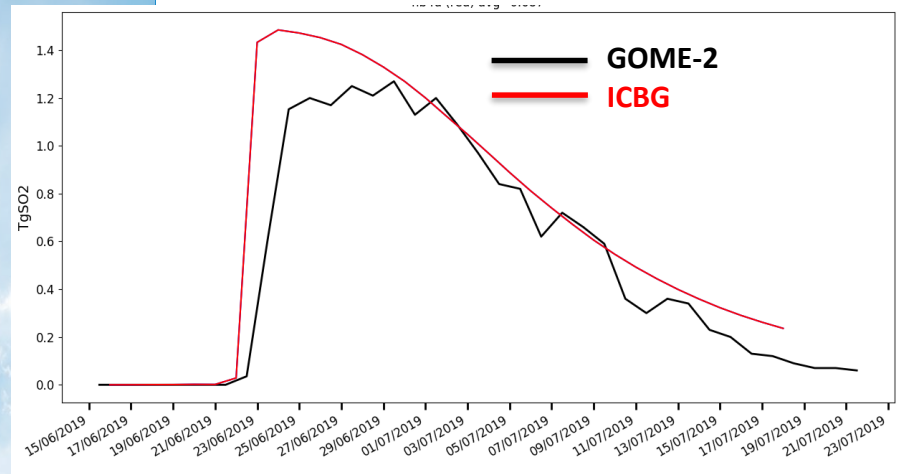
Simulation of Raikoke with ICBG

Simulation of the 21st of June 2019 Raikoke eruption: 1.5 TgS emitted, mostly between 12 and 14km height.



Global volcanic SO_2 burden from GOME-2 (black) and ICBG (red)

Simulation of Raikoke with ICBG

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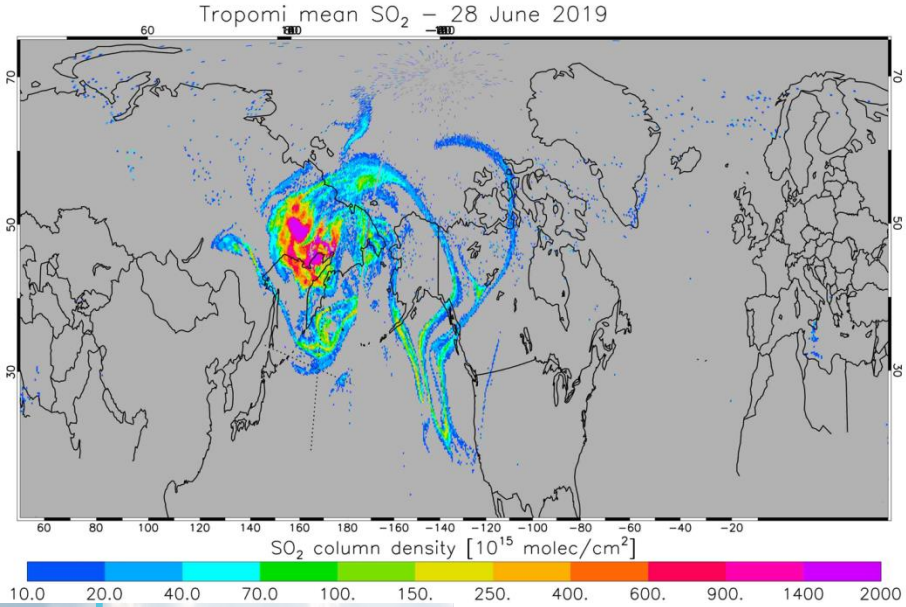
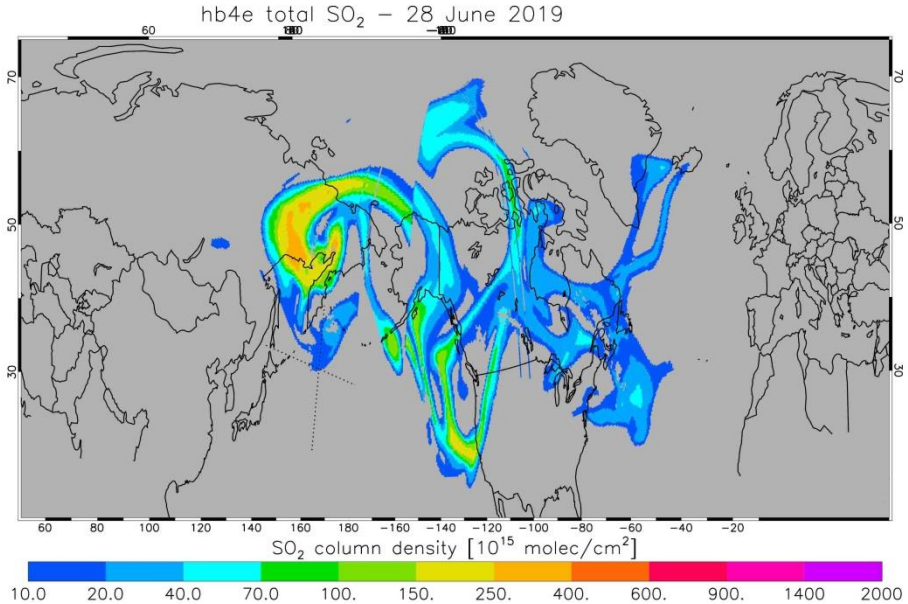
Simulation of the June 2019 Raikoke eruption: 1.5 TgS emitted, mostly between 12 and 14km height.

Evaluation of volcanic SO_2 total column





Volcanic SO_2 column density retrieved by TropOMI (left) and simulated by ICBG on 28/6/19 (right)

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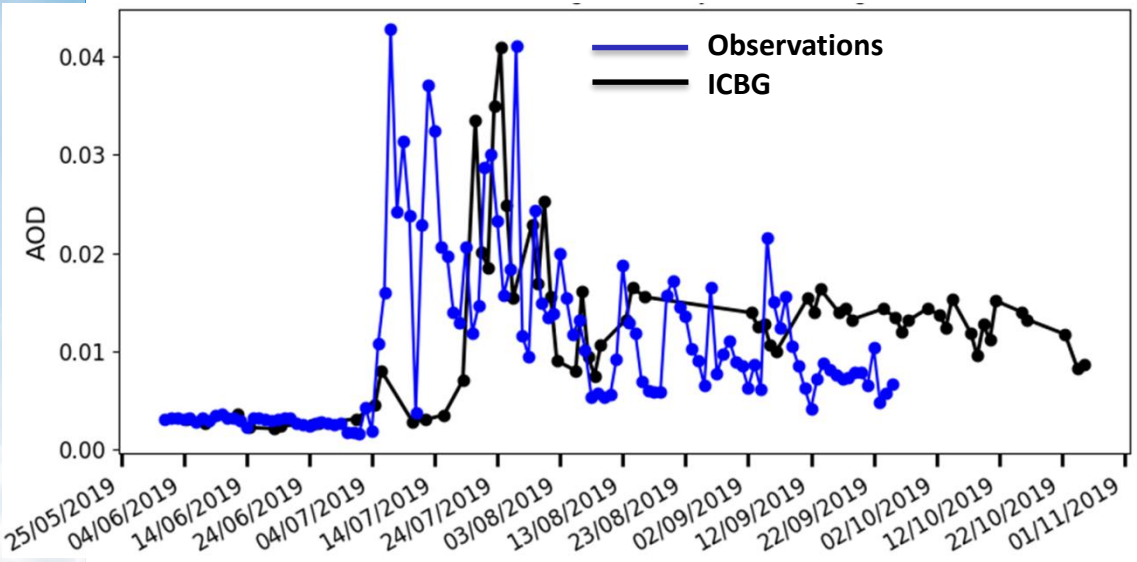
Simulation of Raikoke with ICBG

Simulation of the June 2019 Raikoke eruption: 1.5 TgS emitted, mostly between 12 and 14km height.

Evaluation of stratospheric AOD



Observed (black) and simulated by the ICBG (blue) stratospheric AOD at 532nm at the Observatoire de Haute Provence (France)

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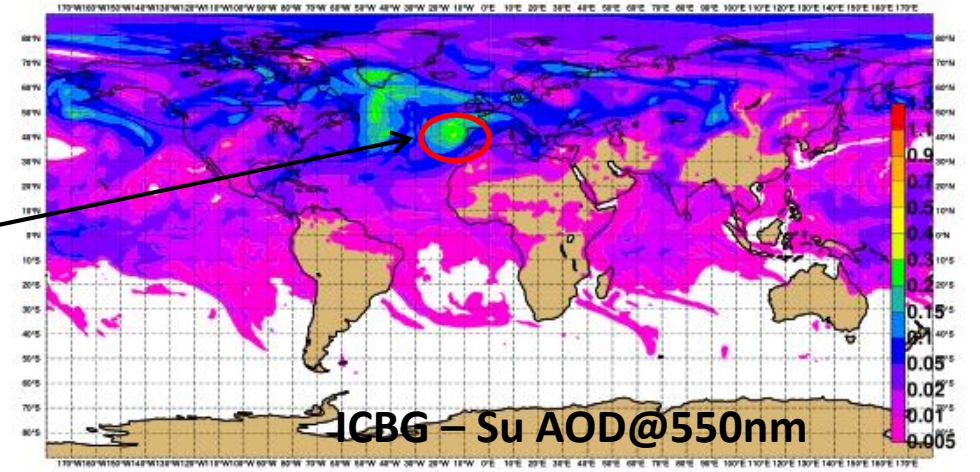
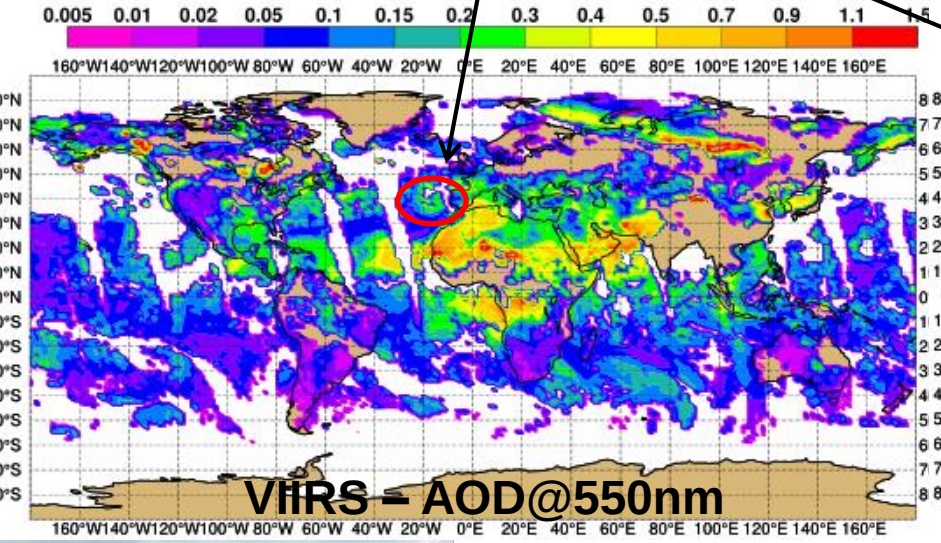
Simulation of Raikoke with ICBG

Evaluation of AOD (stratospheric signal weak as compared to tropospheric contribution)

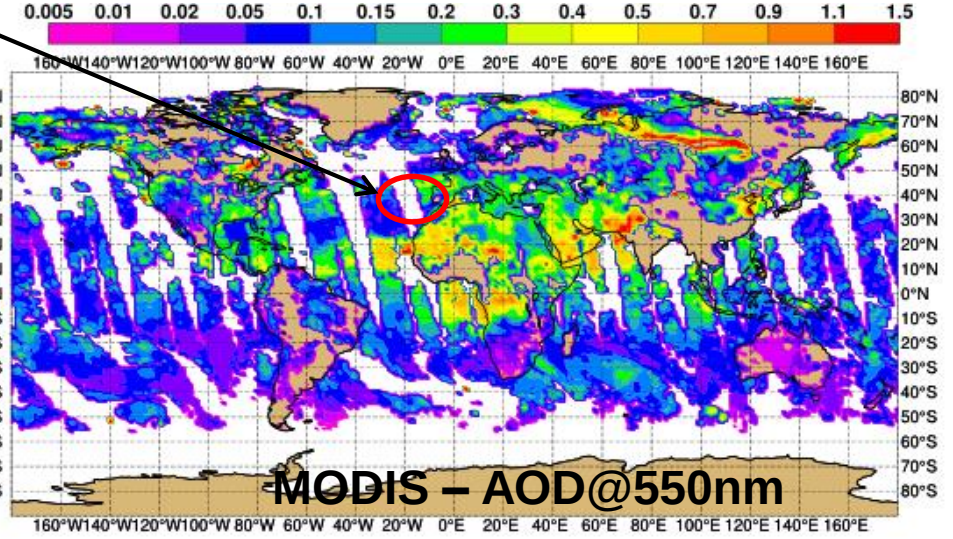
Signal W of Spain on 6th of July 2019



ICBG – Su AOD@550nm



VIIRS – AOD@550nm



MODIS – AOD@550nm

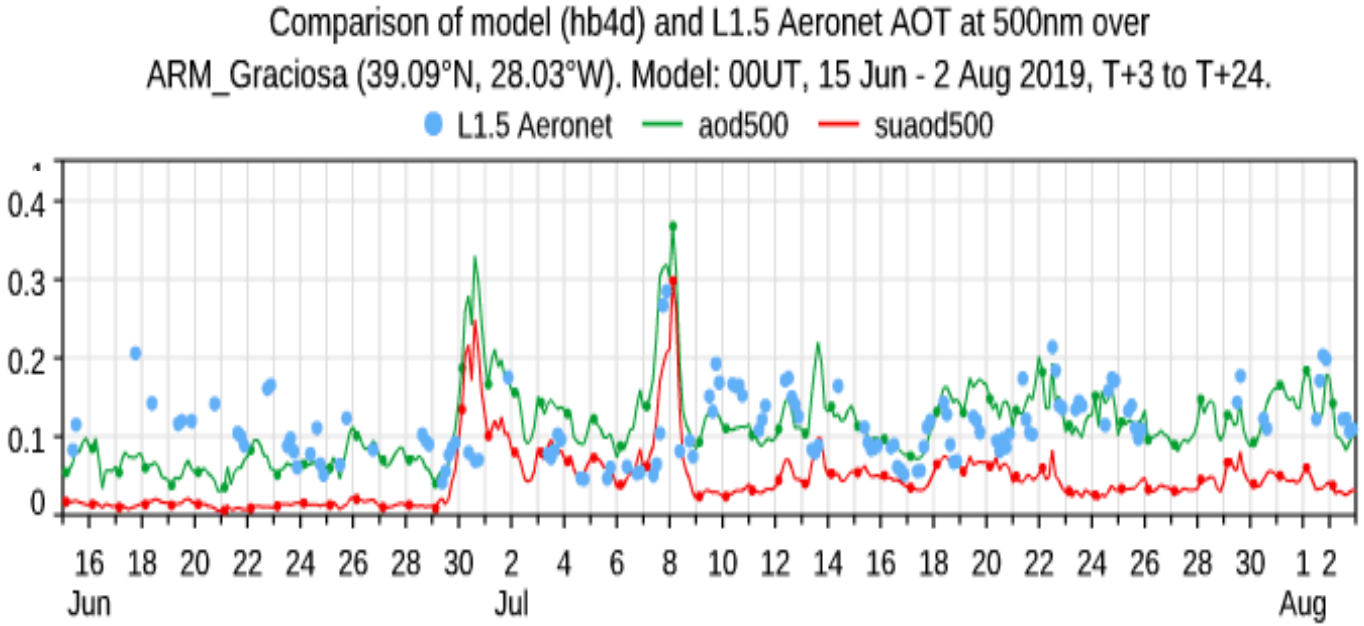
Simulation of Raikoke with ICBG

Atmosphere Monitoring

- Evaluation of AOD (stratospheric signal weak as compared to tropospheric contribution)
- Signal W of Spain

ICBG AOD at 500nm

ICBG stratospheric sulfate AOD at 500nm



Summary - outlook

ICBG shows a good ability to represent stratospheric sulfate, in quiescent conditions and for the Raikoke and Pinatubo eruptions.

More evaluation needed for volcanic simulations, as well as impact on stratospheric chemistry.