The effects of SO2, volcanic ash and sulfate aerosols on photolysis rates and the sulfate chemical production following the volcanic eruptions

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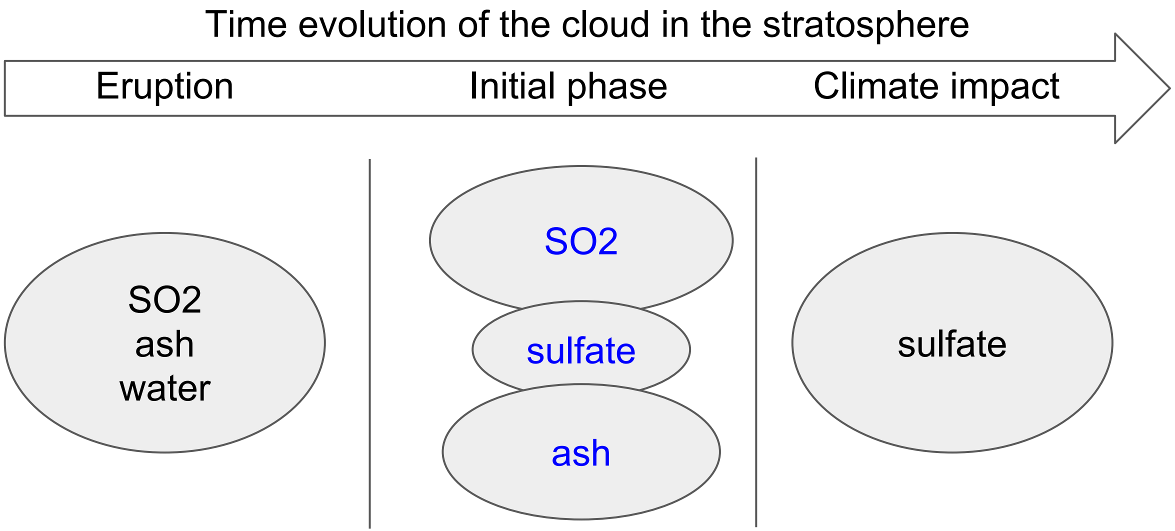
SO_2 key points:

1. greenhouse gas effect
2. SO_2-sulfate cloud heating
3. photolysis rates reduction

Introduction



The evolution of the SO_2-sulfate cloud depends on the atmospheric **dynamics, photochemistry**, and microphysics.

We account that SO_2, sulfate and ash are optically active in SW and LW including the UV range.

Pinatubo, 1991, 20Mt of SO2
8 ppmv, 400 km x 2000 km x 14-21 km

Toba, 75 000 years ago, 100x Pinatubo, 2000 Mt of SO2
15.5 ppmv, equatorial belt 15S-15N x 10-50 hPa

Modelling setup

Standalone models:

1. Line-by-line AER LBLRTM coupled to DISORT
2. Column AER SW/LW RRTM/RRTMG with added SO_2 effects (thanks to Mike Iacono, Eli Mlawer, Karen Cady-Pereira)

Initial phase

NCAR WRF Chem v3.7.1:

Global 1°x1° setup, spectral U&V nudging
Prescribed SST
Interactive chemistry: GOCART(sulphur cycle) coupled with RACM-KPP, fixed climatological ozone

Aerosols are coupled to the chemistry.

Modifications:

1. SO_2 included in RRTMG SW&LW (AER custom version, *dynamics*)
2. SO_2 included in Fast-j actinic flux (*photochemistry*)
3. Double radiation and chemistry calls for instantaneous online diagnostics
4. Volcanic emissions added though the IC or inventory

Climate impact

NASA GISS ModelE2:

Global 2°x2.5° setup
Coupled ocean (Russel) and sea-ice models
Interactive chemistry (Shindell et al. [2006])

Aerosols are coupled to the chemistry.

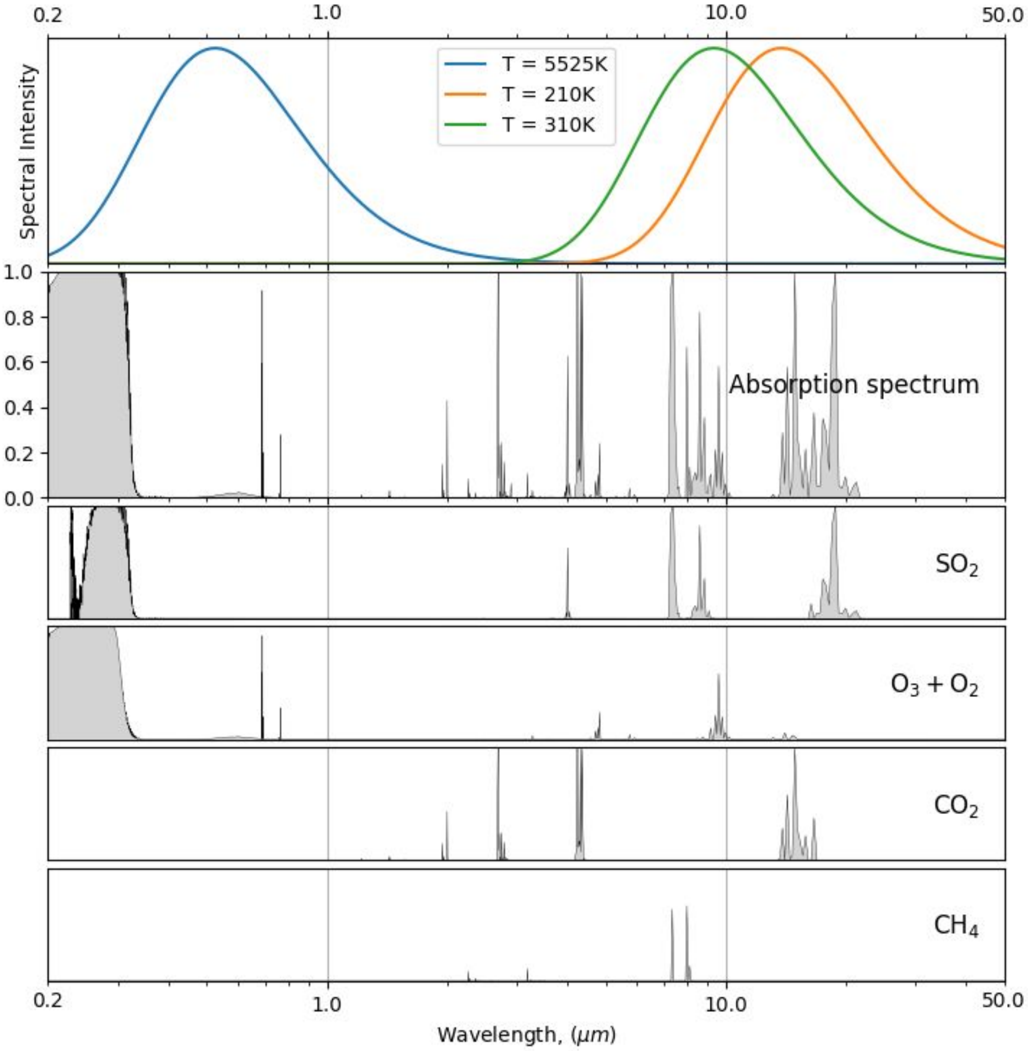
Modifications (future work):

1. SO_2 effect in SW (currently only in LW)
2. SO_2 effect in Fast-j actinic flux

Motivation

10-50 hPa line-by-line absorption spectrum. Realistic equatorial atmosphere + Toba eruption (15.5 ppmv SO_2)

SO_2 is a strong greenhouse gas.
SO_2 competes with O_3 in UV absorption.



SO_2 effects:

1. **Radiative forcing**
2. **Dynamics**
3. **Photochemistry**

SO_2 radiative forcing, Toba, Wm^{-2}

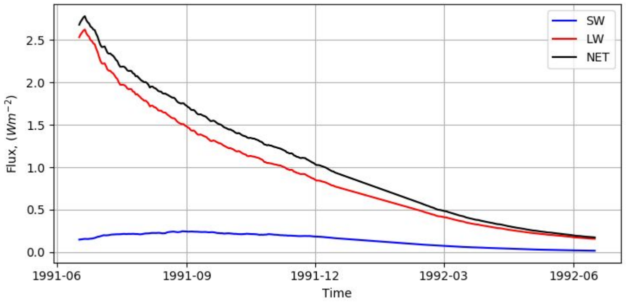
Single profile (RRTM column model)
15.5 ppmv of SO2, 10-50 hPa

TOA	SW	0.69	BOA	SW	-1.08
	LW	12.66		LW	0.32
	NET	13.35		NET	-0.77

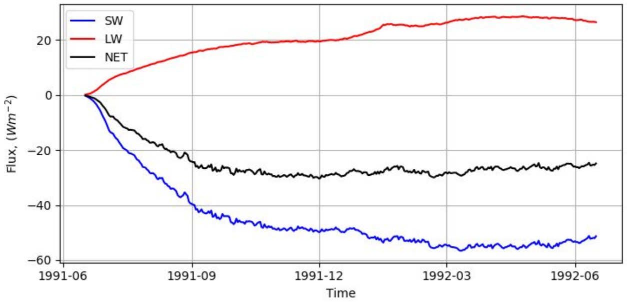
Global average (WRF-Chem)
15.5 ppmv of SO2, 10-50 hPa, 15S-15N

TOA	SW	0.16	BOA	SW	-0.12
	LW	2.62		LW	0.03
	NET	**2.78**		NET	-0.08

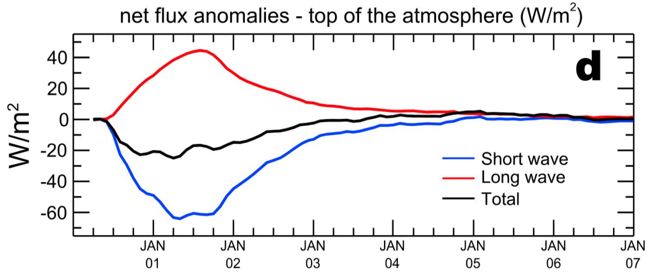
SO_2 TOA forcing
WRF-Chem



sulfate TOA forcing
small r_{eff}, WRF-Chem



sulfate TOA forcing
large r_{eff}, Timmreck et. al., 2010



Max **NET TOA forcing** of SO_2 represents **10-15%** of sulfate forcing (2.8 Wm^{-2} vs 20-30 Wm^{-2})

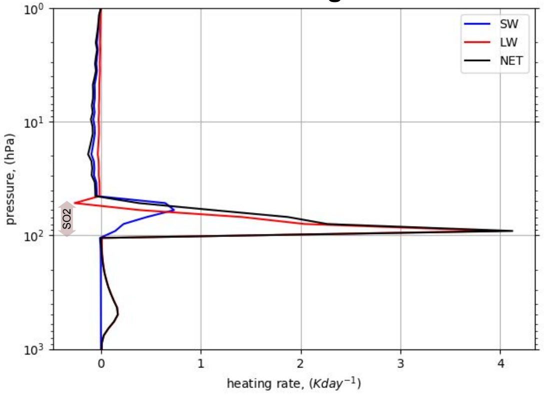
SO_2 effect on dynamics: heating rates

Poster P-28

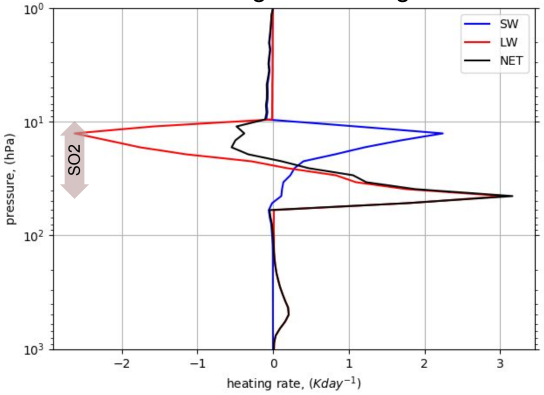
SO_2 daily mean heating rates, column RRTM model, Toba

Zhong et. al., 1996

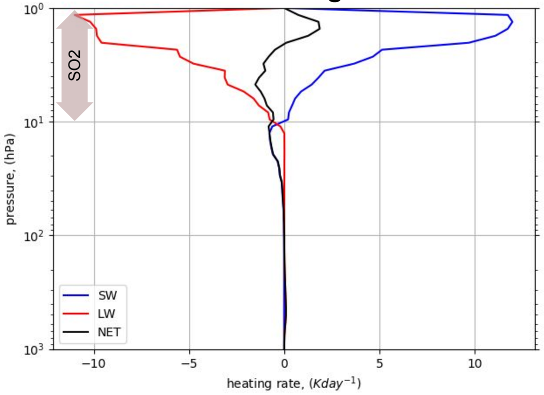
100-50 hPa SO_2
LW heating



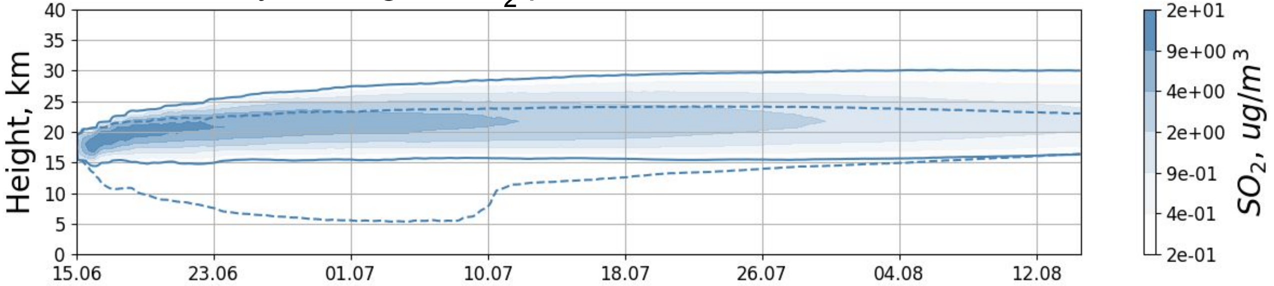
50-10 hPa SO_2
LW heating and cooling



10-1 hPa SO_2
LW cooling



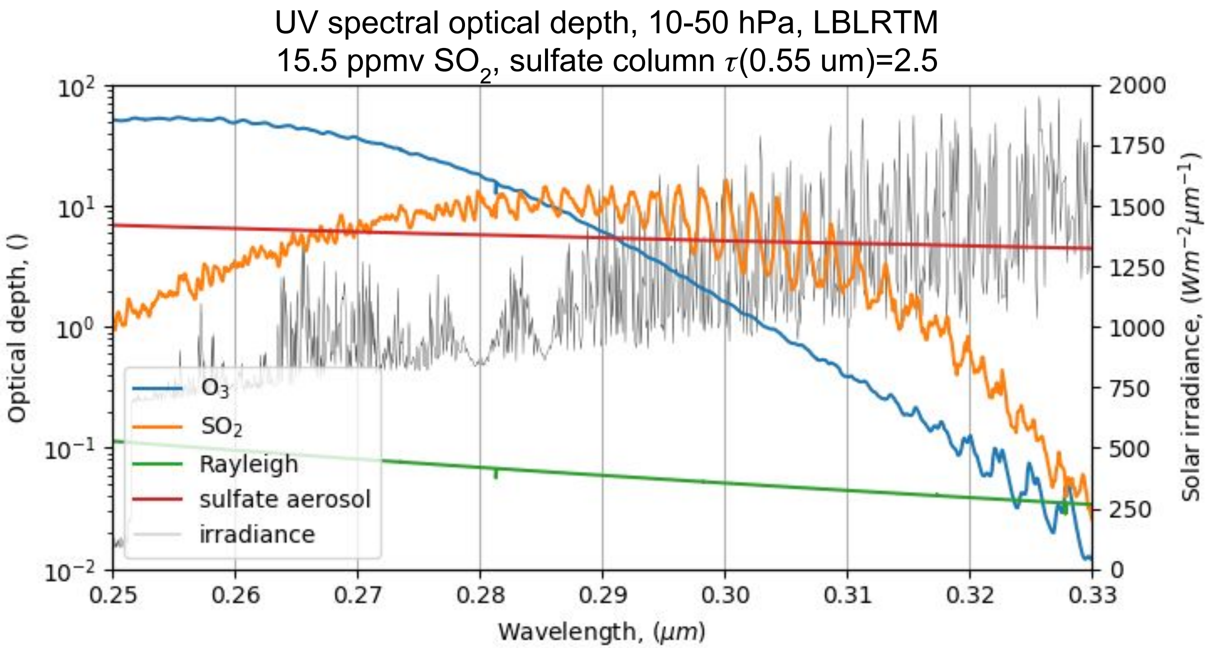
Globally averages SO_2 profile, Pinatubo, WRF-Chem



Radiative feedback on dynamics
solid lines: on
dashed lines: off

Different cloud evolution and transport due to SO_2 heating rates

SO_2 effect on the photochemistry: UV spectrum and O_3



O_3 photolysis relevant range, $\phi > 0$ ← → $\phi = 0$

A + hv -> B + C

$$\frac{d[A]}{dt} = -J_A[A] = -\int \sigma_A(\lambda)\phi_A(\lambda)F(\lambda)d\lambda \times [A]$$

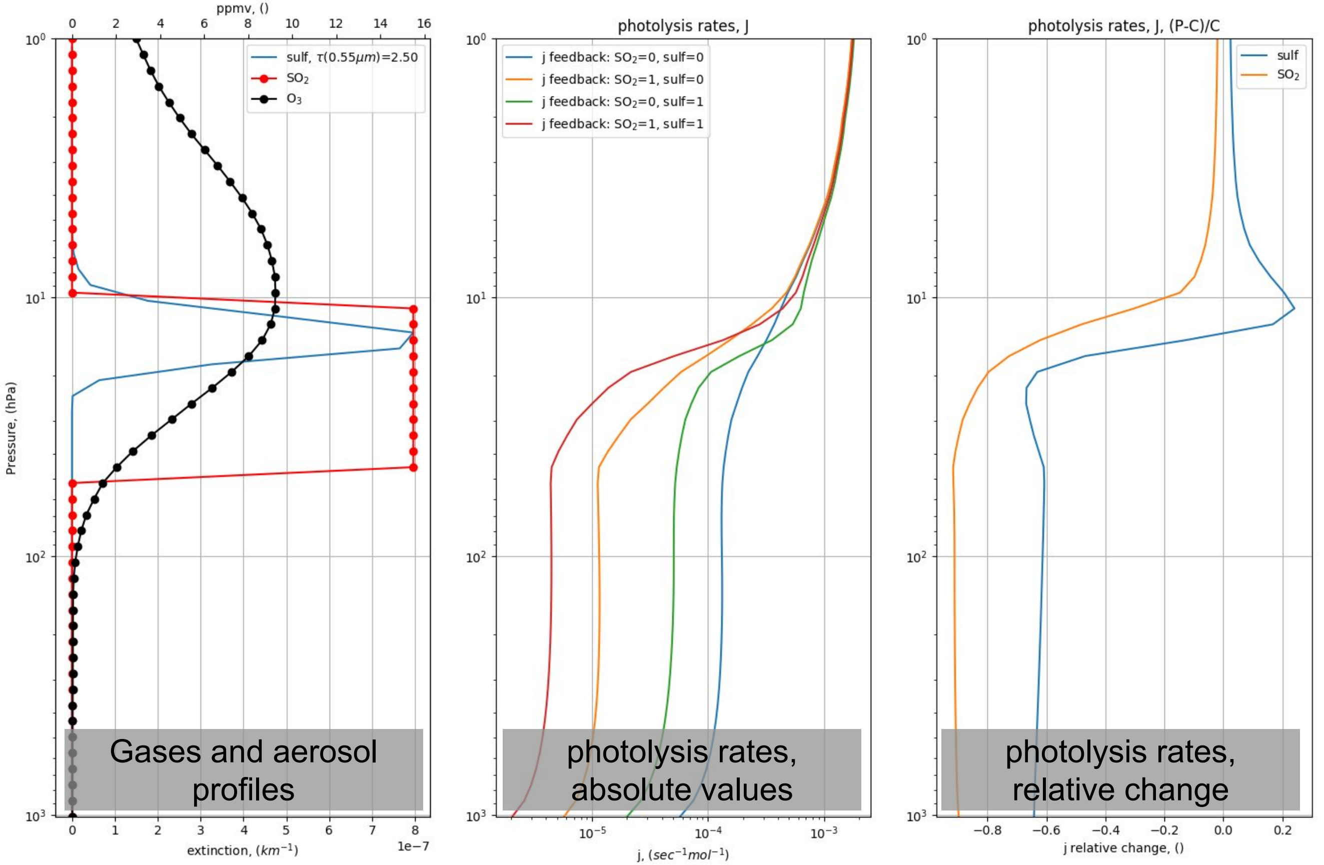
σ - absorption cross section of A
ϕ - quantum yield
F - actinic flux

O_3 photolysis is a primary source of OH, which drives the SO2 oxidation.

Actinic flux reduction due to SO_2 absorption reduces O_3 photolysis rates, SO_2 depletion and sulfate production

SO_2 and sulfate effect on photolysis rates J, Toba

daylight mean photolysis rates, LBLRTM+DISORT



OH is a short-lived radical. Changes in photolysis rates J are only relevant for sulfate production where SO_2 is present.

relative change = (P-C)/C

P: both SO_2 and sulf

C: only one (SO_2 or sulfate)

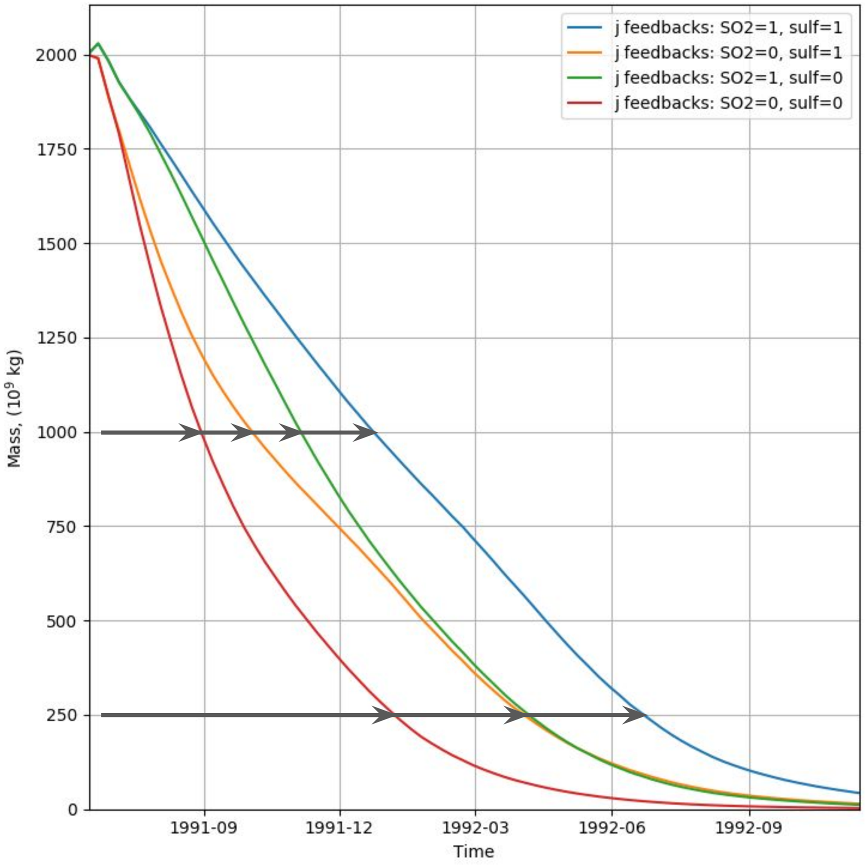
J relative change due to:

SO2: -90% to -20%

sulfate: -70% to 20%

SO_2 depletion time evolution, Toba

Globally integrated SO2 mass, WRF-Chem



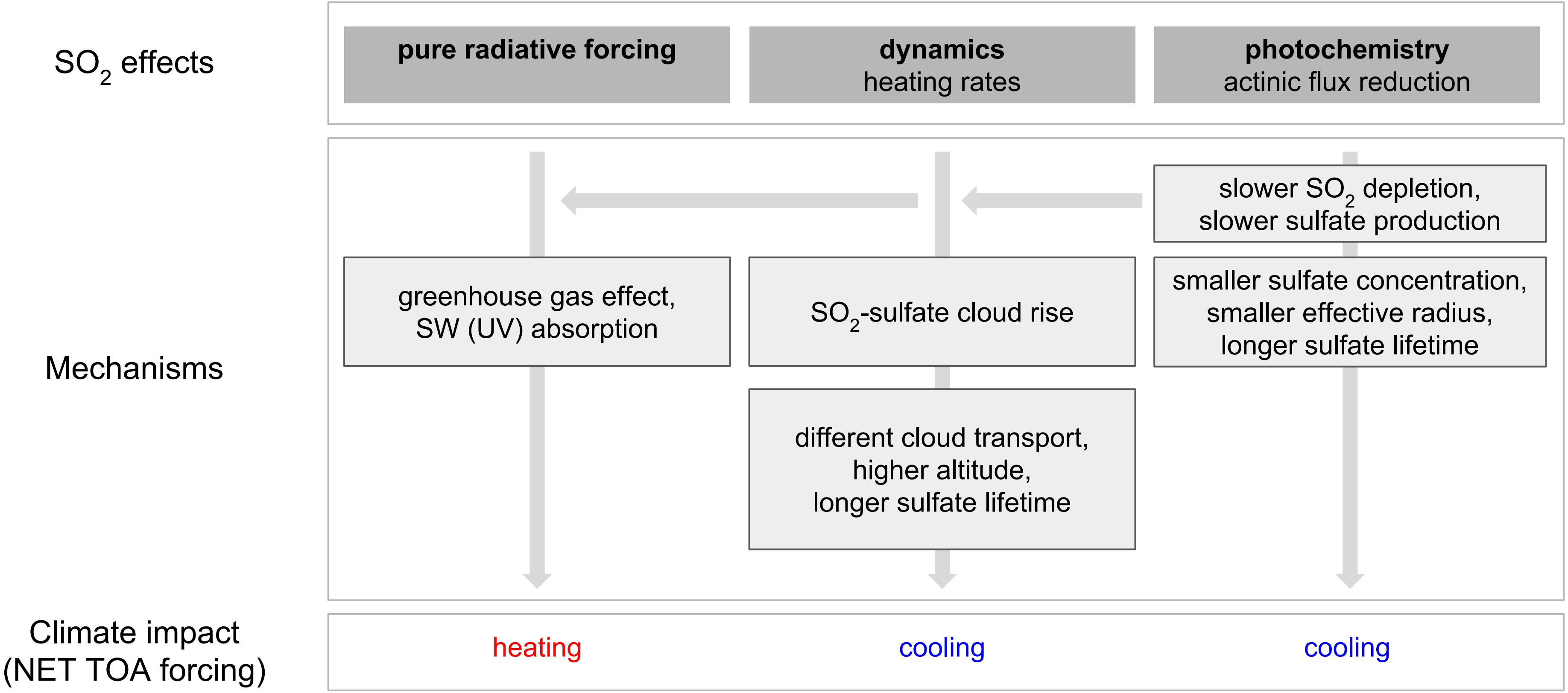
SO_2 time scales

w.r.t experiment without feedbacks

interval	-	sulf	SO_2	sulf, SO_2
e-time (1st month)	x1 4 months	x1.15 4.6 months	**x2.3** 9.2 months	x2.3 9.3 months
50% SO_2	x1 2.6 months	x1.3 3.7 months	**x1.6** 4.9 months	x2.3 6.5 months
12.5% SO_2	x1 7 months	x1.4 9.8 months	**x1.4** 10 months	x1.9 12.6 months

1.4 - 2.3 times slower SO_2 depletion and sulfate production due to effect on photochemistry

Conclusions, Toba



Conclusions, Pinatubo