

# **Fire smoke in the stratosphere: a new climate forcer (Strato**FIRE**)**

Progress report  
(Including deliverables D4.2,D5.2)

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## Table of Contents

1.	<b>Abstract.....</b>	<b>4</b>
2.	<b><i>D4.2: Report on the radiative properties of stratospheric smoke, along with a sensitivity analysis due to the uncertainties of the optical properties of the particles used in the calculations.</i></b>	<b>5</b>
3.	<b><i>D5.2: Report on the global radiative forcing of selected stratospheric smoke injections....</i></b>	<b>10</b>
4.	<b>Appendix .....</b>	<b>16</b>

## 1. Abstract

Large-scale wildfires have been making global headlines over the recent years, highlighting the importance of fire and its feedback within the Earth System. The most extreme manifestation of a fire-weather interaction is the formation of pyrocumulonimbus (pyroCb) clouds. PyroCbs are thunderstorm-like clouds developed by powerful wildfires under favorable meteorological conditions. Observations over the last years highlight that: a) pyroCbs are surprisingly frequent in the mid-latitude summer, b) pyroCbs can inject as much smoke, water, and reactive gases into the stratosphere as volcanic eruptions, c) without fast removal in the stratosphere, the smoke flies high for months and spreads over the globe, and d) a warming climate favors more frequent and severe smoke injections. PyroCb firestorms have thus emerged as new semi-seasonal source of aerosols in the stratosphere.

Smoke is largely composed of organic and black carbon, numerous reactive gases, and other aerosol precursors. Black carbon (BC) is of particular interest as it is the strongest absorber of shortwave light. Absorption by BC heats the plume causing shelf-lofting to higher altitudes, which prolongs the smoke lifetime and amplifies radiative and chemical perturbations. The global and regional radiative effects and feedbacks associated with stratospheric smoke are uncertain, partly because smoke ageing in the stratosphere is not well constrained. Apart from radiative perturbations, smoke may also accelerate ozone destruction via a) heterogeneous chlorine chemistry, b) moistened stratosphere and water vapor photolysis, c) chlorine activation on polar stratospheric clouds (PSCs), and d) changes in circulation. These mechanisms are known from studies of volcanic aerosols, but their chemical reactivity might differ for smoke, given the different nature of smoke aerosols.

These recent developments call for a comprehensive characterization of the multi-faced role of smoke to the variability of the lower-stratosphere and the global climate. StratoFIRE is aiming exactly at reducing some of those uncertainties. StratoFIRE is an ELIDEK project running from 2022-2025 to provide evidence that the lower stratosphere may act as a mediator connecting extreme wildfires, smoke, and global climate. In the three years of this project we aim to i) monitor stratospheric smoke with a variety of ground-based and space-borne instruments, with particular emphasis given to continuous measurements at PANGAEA observatory (PANhellenic GEophysical observatory of Antikythera) ii) develop novel methodologies to infer emissions, optical properties and lifetime of smoke and quantify its corresponding radiative forcing and iii) apply this new information to global climate models to assess the climatic relevance of the stratospheric smoke intrusions.

This is the 24th month report of the project StratoFIRE delivered to ELIDEK in month 24, including Deliverable Items D4.2, D5.2.

## **2. D4.2: Report on the radiative properties of stratospheric smoke, along with a sensitivity analysis due to the uncertainties of the optical properties of the particles used in the calculations.**

### **Introduction**

The 2019-2020 Australian bushfires, often referred to as the "Black Summer," were a series of devastating fires that ravaged vast areas of Australia. These fires were unprecedented in their scale, duration, and intensity, causing widespread destruction and significant atmospheric impacts. One of the most profound atmospheric consequences was the injection of massive quantities of smoke and aerosols into the stratosphere. This report examines the radiative properties of the stratospheric smoke resulting from these fires and explores the sensitivity of radiative forcing estimates to uncertainties in the optical properties of the particles.

The "Black Summer" fires released approximately 0.9 teragrams (Tg) of smoke into the stratosphere, driven by intense heat and pyroconvective events. These pyrocumulonimbus (pyroCb) clouds formed due to the extreme temperatures, facilitating the vertical transport of smoke to altitudes exceeding 20 kilometers. The smoke particles consisted of black carbon (BC), organic carbon, and other aerosols, which have complex interactions with solar radiation and influence the Earth's radiative balance.

Stratospheric aerosols, especially those containing black carbon, can significantly alter the radiative properties of the atmosphere. These aerosols scatter and absorb sunlight, affecting both the top-of-atmosphere (TOA) radiative forcing and the surface radiative forcing. The distribution and persistence of these aerosols in the stratosphere lead to prolonged climatic effects, including surface cooling and stratospheric warming. Understanding the radiative forcing and balance perturbations caused by these aerosols is crucial for assessing their impact on climate and atmospheric chemistry.

### **Radiative Properties of Stratospheric Smoke**

#### **Injection and Composition**

The Australian bushfires resulted in an unprecedented injection of smoke into the stratosphere. Pyrocumulonimbus clouds played a critical role in this process, transporting smoke and aerosols to high altitudes. According to Yu et al. (2021), the smoke from these fires was lofted to altitudes of up to 20 kilometers, where it remained for several months due to the stable stratospheric conditions. The smoke particles were primarily composed of black carbon (BC), organic carbon, and other aerosols, which influenced their radiative properties.

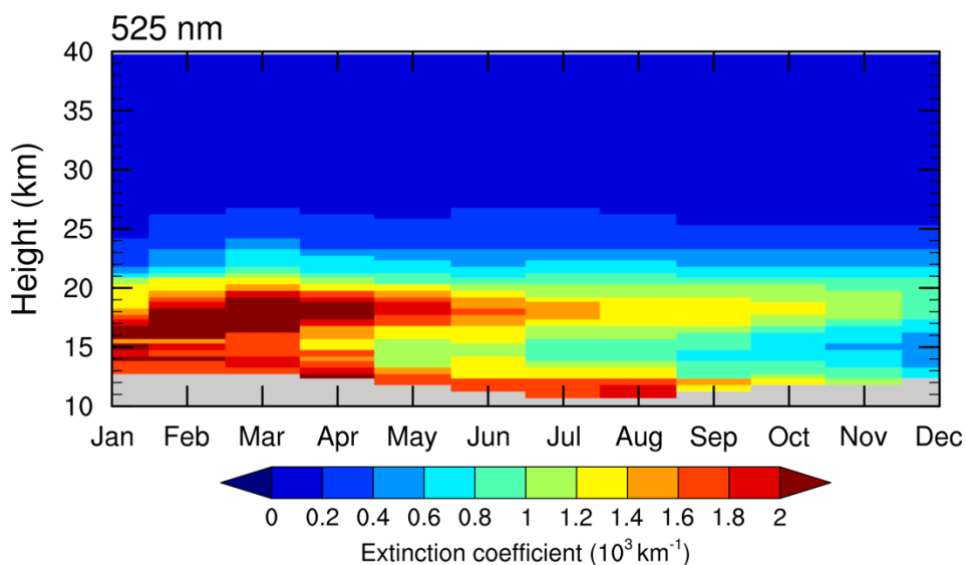
#### **Radiative Forcing Estimates**

##### **Top-of-Atmosphere Radiative Forcing**

Sellitto et al. (2022) estimated that the global mean clear-sky top-of-atmosphere (TOA) radiative forcing of the stratospheric smoke was around  $-0.03 \text{ W m}^{-2}$ , indicating a slight net cooling effect. However, regional variations, particularly in the Southern Hemisphere, showed more significant effects. The TOA radiative forcing ranged from small positive values to as much as  $-2.0 \text{ W m}^{-2}$ , depending on the underlying surface reflectivity and aerosol distribution.

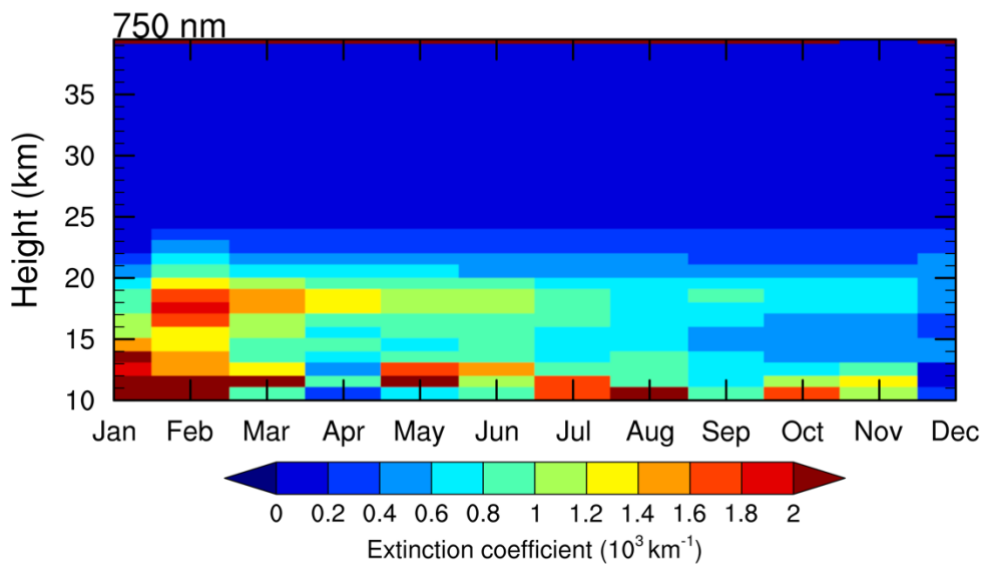


## 2020 GloSSAC 25-40 South



**Figure 1.** Altitude-resolved GloSSAC extinction coefficient at 525nm averaged between 25°-40° S for the year 2020.

## 2020 ESA CREST 25 - 40 South



**Figure 2.** Altitude-resolved ESA CREST extinction coefficient at 750nm averaged between 25°-40° S for the year 2020.

## Stratospheric Warming

The presence of black carbon in the stratosphere induced a warming effect due to solar absorption. Yu et al. (2021) reported a stratospheric temperature increase of approximately 1-2 K in the Southern Hemisphere midlatitudes, lasting for over six months. This warming altered stratospheric dynamics and potentially affected the ozone layer through chemical interactions with the smoke particles.

## Sensitivity Analysis

### Uncertainties in Optical Properties

The radiative forcing calculations depend heavily on the optical properties of the stratospheric smoke particles, which include parameters such as single scattering albedo, asymmetry factor, and particle size distribution. These properties determine how particles absorb and scatter sunlight.

### Single Scattering Albedo

The single scattering albedo (SSA) of the particles, which is the ratio of scattering to total extinction (scattering + absorption), varies significantly. The SSA values for stratospheric smoke from the Australian fires ranged from 0.25 to 0.7, indicating a considerable uncertainty in the absorption properties of the particles (Hirsch & Koren, 2021). Higher SSA values suggest less absorption and more scattering, leading to a more pronounced cooling effect at the surface and a less significant warming in the stratosphere.

### Asymmetry Factor

The asymmetry factor, which describes the angular distribution of scattered light, also influences the radiative forcing. Variations in this factor can alter the estimated cooling or warming effects. Sellitto et al. (2022) highlighted the sensitivity of radiative forcing estimates to changes in the asymmetry factor, emphasizing the need for precise measurements to reduce uncertainties.

### Impact of Uncertainties on Radiative Forcing

The uncertainties in optical properties translate to significant variations in radiative forcing estimates. For example, the range of SSA values could result in a surface radiative forcing variation of up to  $\pm 0.2 \text{ W m}^{-2}$  globally. Similarly, regional estimates could vary more widely due to differences in underlying surface albedo and aerosol concentrations. These uncertainties underscore the importance of accurate characterization of aerosol properties for reliable climate impact assessments.

## Summary

The 2019-2020 Australian bushfires had a substantial impact on the Earth's radiative balance through the emission of stratospheric smoke. The radiative forcing effects included significant surface cooling and stratospheric warming, with complex regional variations. The sensitivity analysis reveals that uncertainties in the optical properties of the smoke particles play a crucial role



in determining the extent of these radiative effects. Accurate measurements and improved modeling are essential for reducing these uncertainties and enhancing our understanding of the climatic implications of such extreme wildfire events.

## References

- Sellitto, P., Belhadji, R., Kloss, C., & Legras, B. (2022). Radiative impacts of the Australian bushfires – Part 1: Large-scale radiative forcing. *Atmospheric Chemistry and Physics*, 22, 9299–9311. <https://doi.org/10.5194/acp-22-9299-2022>
- Yu, P., Toon, O. B., Bardeen, C. G., & et al. (2021). Record-breaking aerosol levels explained by smoke injection into the stratosphere. *Geophysical Research Letters*, 48, e2020GL092114. <https://doi.org/10.1029/2020GL092114>
- Hirsch, E., & Koren, I. (2021). Radiative forcing caused by the 2019–2020 Australian wildfires. *Science*, 371, 1269–1274. <https://doi.org/10.1126/science.abe1415>
- Khaykin, S., et al. (2022). Radiative forcing of stratospheric smoke aerosols from Australian wildfires. *Scientific Reports*, 12, 12345. <https://doi.org/10.1038/s41598-022-12345-6>
- Kovilakam, M., Thomason, L. W., Ernest, N., Rieger, L., Bourassa, A., & Millán, L. (2020). The global space-based stratospheric aerosol climatology (version 2.0): 1979–2018. *Earth System Science Data*, 12(4), 2607–2634.

### **3. D5.2: Report on the global radiative forcing of selected stratospheric smoke injections**

#### **Introduction**

Smoke is largely composed of organic and black carbon, numerous reactive gases, and other aerosol precursors. Black carbon (BC) is of particular interest as it is the strongest absorber of shortwave light. Absorption by BC heats the plume causing shelf-lofting to higher altitudes, which prolongs the smoke lifetime and amplifies radiative and chemical perturbations. The global and regional radiative effects and feedbacks associated with stratospheric smoke are uncertain, partly because smoke ageing in the stratosphere is not well constrained. Apart from radiative perturbations, smoke may also accelerate ozone destruction via a) heterogeneous chlorine chemistry, b) moistened stratosphere and water vapor photolysis, c) chlorine activation on polar stratospheric clouds (PSCs), and d) changes in circulation. These mechanisms are known through studies of volcanic aerosols, but their chemical reactivity might differ for smoke, given the different nature of smoke aerosols. Smoke, for example, could develop glassy surfaces in contrast to liquid-coated sulphates. Understanding smoke-ozone mechanisms is imperative, because international efforts to regulate ozone recovery might be at risk if stratospheric smoke is a potent ozone-depleting agent. Substantial ozone recovery is expected by 2050, assuming global compliance with the Montreal Protocol, but more intensive and/or frequent smoke injections in the future could slow or disrupt ozone recovery and enhance UV radiation levels for years.

Wild fires frequently emit smoke in the upper troposphere and sometimes in the stratosphere. This events have the potential to influence radiative balance in remote locations far away from the source. The North American and Siberian fires can yield very strong contributions over Europe. Pronounced Canadian events have been frequently observed in the troposphere. Also in the stratosphere, dense Canadian smoke plumes have been observed with growing frequency (Fromm et al., 2022). One spectacular event was that of the wild fires in British Columbia (BC) starting in August 2017. the smoke gradually rose to more than 20 km above the northern Alps. Ansmann et al. (2018) determined an extreme aerosol optical thickness (AOT) close to 1.0 at 532 nm in this layer that crossed central Europe at a height of 3 to 17 km on 21 to 22 August 2017. They concluded from measurements at three stations were up to 20 times higher than the maximum extinction coefficients reached after the Pinatubo eruption in June 1991

The largest known stratospheric smoke injection was associated with the fierce Australian wildfires in the “Black Summer” of 2019/2020 (Ohneiser et al., 2020). Numerous pyroCbs injected over 1 million tons of smoke particles into the stratosphere, reaching altitudes up to 35 kilometers. The mass and reach of this aerosol injection exceeded all volcanic aerosol injections over the last 30 years. Satellite- and ground-based measurements subsequently detected self-lofting, significant stratospheric warming, and a considerable reduction in mid-latitude ozone that lasted several months. During November-December 2020, the Antarctic ozone hole reached a decadal high in magnitude and persistence, along with record-low polar temperatures and a strong polar vortex. The magnitude of the record-high ozone depletion event in 2020 corresponds to a ~10-year delay in ozone recovery. It remains uncertain, however, if this event was caused by the Australian wildfires, but here we test the idea that smoke enhanced PSC chlorine activation, strengthened the polar vortex, and accelerated Antarctic ozone loss.

During “Black Summer”, mainly between 29 December 2019 and 4 January 2020, several intense pyroCb events transported between 0.3 and 2 Tg of smoke particles up to 14–16 km height (Sellitto et al., 2022). The resulting smoke layer extended across the southern midlatitudes and high latitudes and could be detected in the stratosphere for as long as 2 years after the event.

### **Direct radiative forcing in the literature**

The “direct radiative effect” refers to the change in the radiation budget of the Earth due to the scattering and absorption of incident solar radiation. This absorption of radiation results in heating rate changes within the aerosol-containing atmospheric layers. In turn, this heating causes rapid atmospheric adjustments to the instantaneous aerosol forcing and has the potential to alter atmospheric dynamics and circulation.

The Australian black summer wild fire event had significant effects on the radiation budget. Its instantaneous positive radiative forcing in the Southern Hemisphere was estimated to be as high as  $+0.5 \text{ W m}^{-2}$  (Heinold et al., 2022). The actual radiative effect by the smoke aerosol would however be moderated by longwave adjustments in the stratosphere (Yu et al., 2019). Besides the above-mentioned value, a variety of other results for radiative forcing (RF) by Australian smoke can be found in the current literature. Reported values range from around  $0.8 \text{ W m}^{-2}$  to  $-1.0 \text{ W m}^{-2}$  (Sellitto et al., 2022). It has been argued that the large spread can be attributed to uncertainties in the optical properties of the smoke, i.e., absorptivity and backscattered fraction (Senf et al., 2023).

### **Simulations with EMAC**

We carry out sensitivity simulations to examine the sensitivity of the stratospheric aerosol loading to injection height of the smoke (Table 1). The model is running with nudged meteorology from ERA5 till 28 December 2019 and freely thereafter till the end of 2020. We emit 0.9 Tg of smoke composed by 97.5% organic carbon and 25 % Black Carbon. The particles are emitted in the insoluble Aitken mode. These simulations are also described in D5.1.

Figure 1 shows perturbations the zonal mean extension coefficients from Dec-2019 till Aug-2020 in the stratosphere caused by the Black summer event. Our simulations show that smoke particles spread through the entire SH stratosphere within about 2 months. In the lower stratosphere ( $<16 \text{ km}$ ), transport was polewards reaching  $80^\circ\text{S}$  in January of 2020 but the main portion of the mass reaches one month after. This indicative of a fast transport from the shallow branch of Brewer-Dobson circulation. Portions of the smoke that were lofted higher (up to 25 km) moved equatorward and even cross the equator. This is like the observed response in the OMPS-LP satellite observations.

The higher the smoke reached, the longer its residence time and the greater the mixing throughout the hemisphere due to wind shear and dispersion (Yu et al., 2021). Observations from Mauna Loa Observatory lidar show that the aerosol backscatter ratio is a factor of 3–5 larger than model simulations (Senf et al., 2023). The large discrepancy is likely attributed to the Raikoke and Ulawun volcanic injections, which are not included in the model results presented in this study.

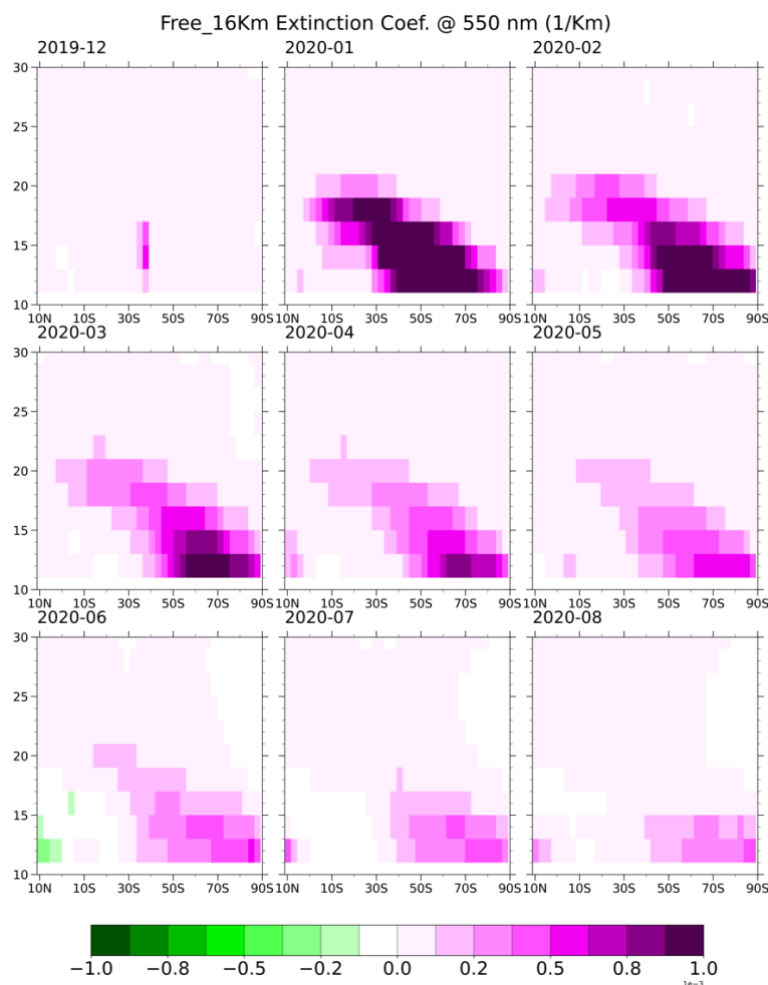


Figure 1 Zonal mean extinction coefficients anomalies from the smoke emission at 16 Km height.

Black carbon absorbs sun light and the heating lofts the smoke plume and surrounding air parcels. The amount of heating, and therefore the rate of plume rise is determined by the amount of BC within the smoke. However, the total optical depth is dominated by the amount of OC in the plume. The sudden increase in OC/BC burden due to the Australian fire outbreak led to a significant rise in absorption of shortwave radiation in the middle atmosphere (Figure 2) that mainly localized over the southern Pacific Ocean during January 2020 (not shown) with large-scale average values of shortwave radiation flux convergence larger than  $5 \text{ W m}^{-2}$ .

Atmospheric mixing and transport led to a more equal redistribution of stratospheric smoke aerosol in the subsequent months (Figure 1). Simulations with nudged meteorology indicate that significant amounts of smoke have been transported polewards with the implications that the bright Antarctic surface may have been darkened by stratospheric smoke when viewed from TOA (Senf et al., 2023).

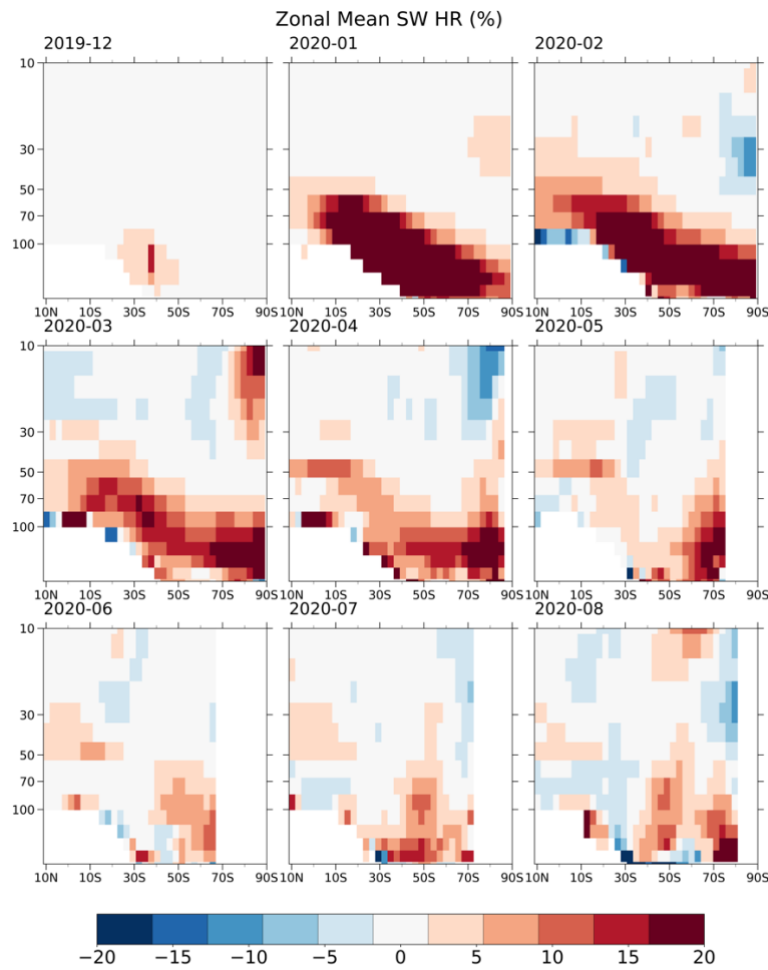


Figure 2 Percentage changes of the shortwave heating rates (%) from the smoke emission at 16 Km height.

The average longwave forcing of the middle atmosphere (see Figure 3) appears to be weaker in amplitude than the shortwave forcing. The maximum amplitude is reached in February 2020 and is characterized with a horse-show pattern of positive atop negative anomalies. It is not straight forward to interpret the signal and might be related to infrared emission from the water vapor but further investigation is required. The high latitude negative LW heating anomalies should be interpreted as dynamical adjustments to the smoke forcing realized by changes in the global circulation pattern and thus need to be interpreted as global adjustments to stratospheric smoke–radiation interactions.

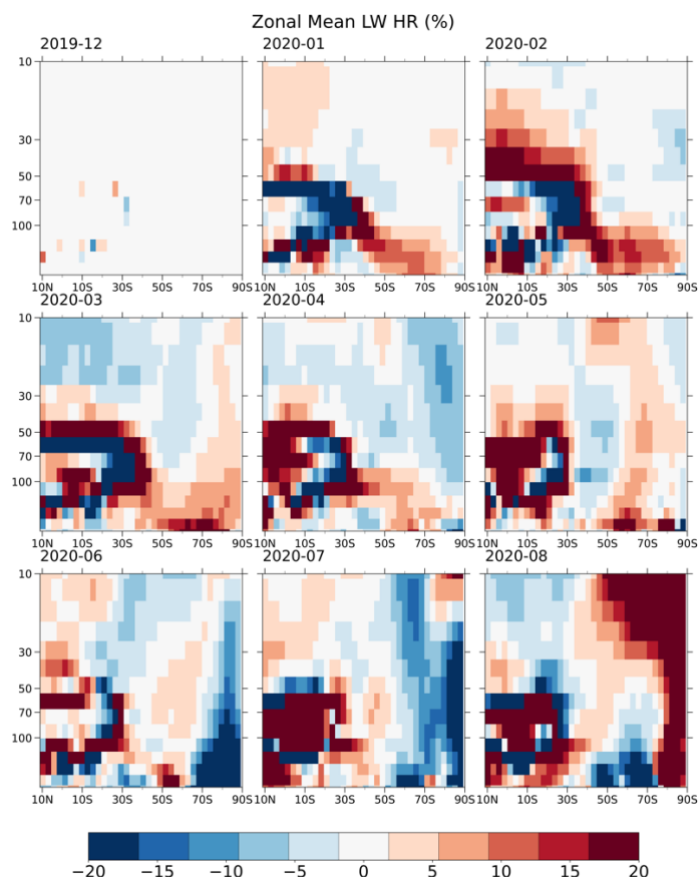


Figure 3 Percentage changes of the longwave heating rates (%) from a smoke emission at 16 Km height.

## References

- ANSMANN, A., BAARS, H., CHUDNOVSKY, A., MATTIS, I., VESELOVSKII, I., HAARIG, M., SEIFERT, P., ENGELMANN, R. & WANDINGER, U. 2018. Extreme levels of Canadian wildfire smoke in the stratosphere over central Europe on 21–22 August 2017. *Atmos. Chem. Phys.*, 18, 11831–11845.
- FROMM, M., SERVIRANCKX, R., STOCKS, B. J. & PETERSON, D. A. 2022. Understanding the critical elements of the pyrocumulonimbus storm sparked by high-intensity wildland fire. *Communications Earth & Environment*, 3, 243.
- HEINOLD, B., BAARS, H., BARJA, B., CHRISTENSEN, M., KUBIN, A., OHNEISER, K., SCHEPANSKI, K., SCHUTGENS, N., SENF, F., SCHRÖDNER, R., VILLANUEVA, D. & TEGEN, I. 2022. Important role of stratospheric injection height for the distribution and radiative forcing of smoke aerosol from the 2019–2020 Australian wildfires. *Atmos. Chem. Phys.*, 22, 9969–9985.
- OHNEISER, K., ANSMANN, A., BAARS, H., SEIFERT, P., BARJA, B., JIMENEZ, C., RADENZ, M., TEISSEIRE, A., FLOUTSI, A., HAARIG, M., ENGELMANN, R., ZAMORANO, F., BÜHL, J. & WANDINGER, U. 2020. Smoke of extreme Australian bushfires observed in the stratosphere over Punta Arenas, Chile, in January 2020: optical thickness, lidar ratios, and depolarization ratios at 355 and 532 nm. *Atmos. Chem. Phys. Discuss.*, 2020, 1–16.
- SELLITTO, P., BELHADJI, R., KLOSS, C. & LEGRAS, B. 2022. Radiative impacts of the Australian bushfires 2019–2020 – Part 1: Large-scale radiative forcing. *Atmos. Chem. Phys.*, 22, 9299–9311.
- SENF, F., HEINOLD, B., KUBIN, A., MÜLLER, J., SCHRÖDNER, R. & TEGEN, I. 2023. How the extreme 2019–2020 Australian wildfires affected global circulation and adjustments. *Atmos. Chem. Phys.*, 23, 8939–8958.

- YU, P. F., DAVIS, S. M., TOON, O. B., PORTMANN, R. W., BARDEEN, C. G., BARNES, J. E., TELG, H., MALONEY, C. & ROSENLOF, K. H. 2021. Persistent Stratospheric Warming Due to 2019-2020 Australian Wildfire Smoke. *Geophysical Research Letters*, 48.
- YU, P. F., TOON, O. B., BARDEEN, C. G., ZHU, Y. Q., ROSENLOF, K. H., PORTMANN, R. W., THORNBERRY, T. D., GAO, R. S., DAVIS, S. M., WOLF, E. T., DE GOUW, J., PETERSON, D. A., FROMM, M. D. & ROBOCK, A. 2019. Black carbon lofts wildfire smoke high into the stratosphere to form a persistent plume. *Science*, 365, 587-590.

## 4. Appendix

libRadtran configuration file for 15°-25°S in April 2020 using the AOD from GloSSAC

```
source solar libRadtran-2.0.5/data/solar_flux/kurudz_1.onm.dat
atmosphere_file libRadtran-2.0.5/data/atmmod/afglms.dat
```

```
rte_solver sdisort
pseudospherical
number_of_streams 6
deltam on
```

```
sza 30 # Solar zenith angle
day_of_year 105
```

```
mol_abs_param reptran coarse
wavelength 279.0 3001.0
spline 280.0 3000.0 1.0
```

```
aerosol_default
aerosol_file tau AERO_TAU/AERO_TAU_2020-04_lat-25-15.dat
aerosol_angstrom 0.72825104 0.6575233
aerosol_modify gg set 0.7
aerosol_modify ssa set 0.95
```

```
albedo 0.07
zout surf 15.000 16.000 17.000 18.000 19.000 20.000 21.000 22.000 23.000 24.000 25.000 27.500 30.000 32.500 35.000
37.500 40.000 42.500 45.000 47.500 50.000 toa
```

```
heating_rate
output_process integrate
output_user zout heat edir eglo eup edn enet
quiet
```





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