2. D4.1: Microphysical and optical properties of stratospheric smoke

This report synthesizes key findings of the published literature that investigate the complex dynamics and optical properties of smoke particles injected into the atmosphere during wildfire events in the Pacific Northwest in 2017 as well as the evolution of smoke particles' optical properties and the radiative impact of the record-breaking Australian fires in 2019-2020.

The optical and microphysical properties of wildfire aerosols reaching into the stratosphere are crucial for understanding their interaction with radiation fields. These properties, not directly accessible from observations, are modelled to enhance radiative transfer simulations.

Useful definitions and summary of the key optical properties of an aerosol layer:

- **Single Scattering Albedo (SSA):**
  - Defines the ratio of scattering to extinction cross-sections.
  - Quantifies the fraction of extinction due to scattering and absorption.
- **Scattering Phase Function:**
  - Represents the angular distribution of scattered radiation.
  - Compact representation by the asymmetry parameter (g).
  - Larger particles, according to Mie theory for spherical particles, exhibit more forward scattering, resulting in larger values of g.

Key considerations from the literature include but are not limited to:

1. **Absorptivity Modelling:**
   - The absorptivity of the aerosol layer is represented by the single scattering albedo (SSA).
   - SSA is varied from 0.80 to 0.95 in 0.05 steps, covering a spectrum of possible biomass burning aerosol configurations.
   - Biomass burning aerosols are more absorbing in fresh plumes, dominated by black carbon (BC), evolving into less absorbing layers as BC ages and mixes with other species.
2. **Scattering Angular Distribution Modelling:**
   - The angular distribution of scattering is modelled using Henyey–Greenstein phase functions with asymmetry parameters (g) of 0.50 and 0.70.
   - These parameters are chosen to cover a reasonable variability for biomass burning aerosols in the shortwave spectral range.
3. **Evolution and Mixing of Aerosol Layers:**
   - Representing the atmospheric evolution of biomass burning aerosols is challenging.
   - Fresh plumes are characterized by smaller, more absorbing particles (low SSA, low g), predominantly BC.
   - As BC ages and mixes, aerosols can evolve towards brown carbon aerosol layers (BrC), with larger, less absorbing particles (high SSA, high g).
   - Online aerosol/climate models tend to represent biomass burning aerosols as too absorptive.
4. **Stratospheric Smoke Properties:**
   - The unique optical properties of stratospheric smoke from pyrocumulonimbus activity are assumed to have near-spherical shapes consistent with past studies showing that such particles can exhibit high polarized lidar ratios (PLDR).

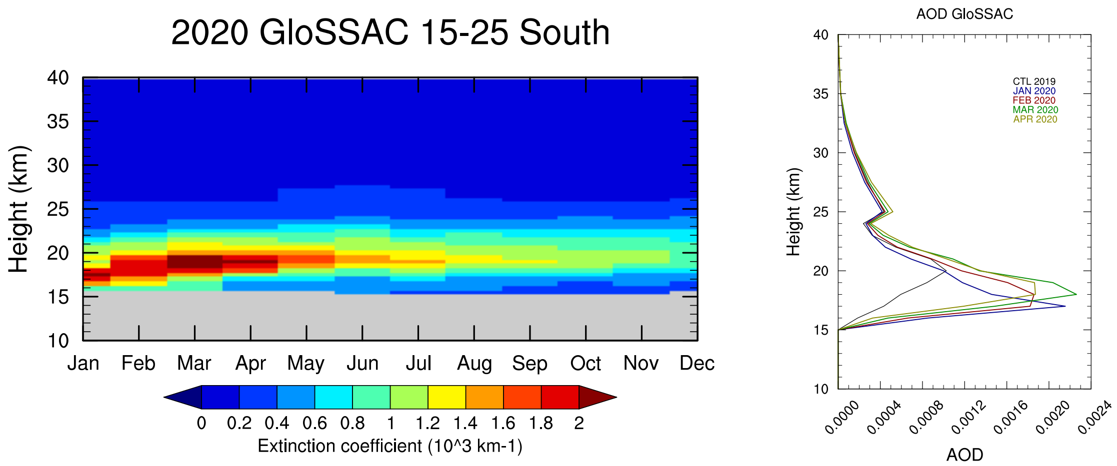

Figure 1. Left. Aerosol extinction coefficient at 525 nm from GloSSAC dataset for a representative latitude band (15°–25° South) in the 2020 wildfire season showcasing the enhancement of wildfire-related injection of smoke aerosols into the stratosphere. Right. Computation of Aerosol Optical Depth (AOD) from GloSSAC for the first four months of 2020, during the 2019–2020 wildfire season, known as the Black Summer in Australia.

Model Configuration:

1. **Radiative Transfer Equation Solver:**
    - libRadtran employs the SDISORT (spherical DISORT) scheme to solve the radiative transfer equation.
2. **Wavelength Range:**
    - Radiative forcing (RF) and heating rate estimates are integrated across the spectrum between 300 and 3000 nm.
    - Spectral resolution begins at 0.1 nm.
3. **Solar Flux Input:**
    - Solar flux input is based on the Kurucz (1992) model.
4. **Atmospheric State:**
    - Extra-aerosol atmospheric conditions are determined using the AFGL (Air Force Geophysics Laboratory) summer mid- or high-latitude climatological standards, depending on the latitude range (Anderson et al., 1986).
5. **Surface Albedo:**
    - Clear-sky conditions are primarily considered.
    - Shortwave surface albedo is set to 0.07, a typical value for sea surfaces, given that most of the fire plume disperses over the ocean.

Considerations:

- The radiative impact assessments focus on clear-sky conditions.
- The libRadtran model employs the SDISORT scheme for solving the radiative transfer equation, ensuring accurate calculations of RF.
- Specific atmospheric and surface conditions are defined based on established climatological standards and empirical data.

- The chosen wavelength range and solar flux input contribute to the precision of the RF estimates.

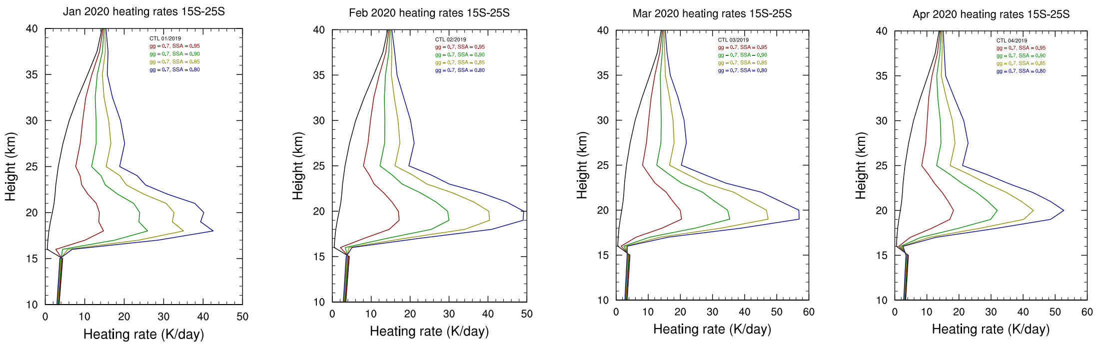

Figure 2. Monthly mean heating rate profiles for the latitude band 15°–25° South for the first 4 months of 2019 and 2020 wildfire season calculated by libRadtran radiative transfer model showcasing the amount of radiative warming caused by the aerosol injection into the stratosphere.