New insights into physical and chemical atmospheric transformations of biomass burning aerosol from wildfires in Siberia

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Motivation

- Wildfires in Siberia are a major source of aerosol in Northern Eurasia.

- Accurate knowledge of the atmospheric evolution of biomass burning (BB) aerosol is a key prerequisite for accurate estimates of the impact of fire emissions on climate.

- Field and laboratory studies indicate that atmospheric aging of BB aerosol is associated with profound and diverse chemical and physical transformations which, in most cases, are not adequately represented in the state-of-the-art atmospheric models.

This study builds upon our previous studies of BB aerosol using analysis of satellite observations and modeling: Konovalov et al., ACP (2017); (2018); (2019).

Methods: Analysis of satellite data

- Evaluation of the enhancement ratio (EnR) for AAOD and AOD, defined as the ratio of an enhancement of the actual AAOD (τ_{abs}) or AOD due to fire emissions to a corresponding enhancement of an inert aerosol tracer as follows (in the case of AAOD):

$$EnR_{abs} = \frac{\tau_{abs} - \tau_{abs}^{bgr}}{\tau_{abs}^{t}} \langle \frac{\tau_{abs} - \tau_{abs}^{bgr}}{\tau_{abs}^{t}} \rangle^{-1}$$

τ_{abs}^{t} : AAOD of the BB aerosol tracer composed of non-volatile material (subject to only dry and wet deposition and coagulation)

τ_{abs}^{bgr} : "Background" AAOD (in the absence of fires)

- Estimation of the BB aerosol photochemical age (the solar radiation exposure time):

$$t_e = k_T^{-1} \log([T_1]\,[T_2]^{-1})$$

: [T1] and [T2] are the columnar concentrations of the inert and reactive gaseous tracers of BB aerosol (from a chemistry transport model)

- Evaluation of the nonlinear trends in EnRs :

$$\boldsymbol{w} = argmin \left\{ \sum_{i=1}^{Nd} (EnR_i - y(t_{ei}))^2 \right\}$$

$$y(t_e) = \sum_{k=1}^{N} \frac{w_{k1}}{(1 + \exp(w_{k2} t_e + w_{k3}))} + w_0$$

Methods: BB aerosol evolution modeling

- The CHIMERE chemistry transport model combined with the OPTSIM module is used to simulate the BB aerosol sources, evolution and optical properties
- A simplified (mechanistic) adjustable VBS scheme (based on Ciarelli et al. (GMD, 2017) is used to represent organic aerosol (OA) oxidation processes

MV-POA + OH $\rightarrow$ 1.3 LV-SOA + OH
LV-POA + OH $\rightarrow$ 1.3 LV-SOA + OH

NTVOC + OH $\rightarrow$ ξ_1 *0.33 HV-SOA + (1-ξ_1) 0.30 LV-SOA + OH
HV-SOA + OH $\rightarrow$ MV-SOA + OH
MV-SOA+ OH $\rightarrow$ ξ_2 LV-SOA + OH

$E_{NTVOC} = \xi_3 E_{POA}$

C*= {10^0, 10^2, 10^3} (μg m^{-3}) for LV-, MV-, HV- SOA (or POA)

ξ_1 - ξ_3 are adjustable parameters

- Evaluation of the BB aerosol optical properties for a core/shell structure of the particles

Different constant values of the imaginary refractive index are assumed for the POA and SOA species; SOA is assumed to be much less absorbing than POA.

Satellite data

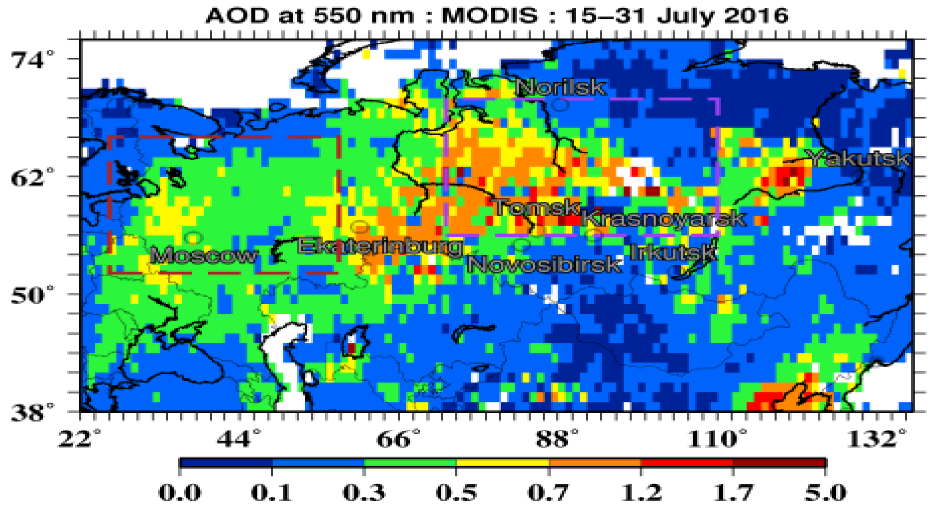
✓ Retrievals from the OMI observations (the OMAERUV Level-2 data product): AOD and SSA (quality flag “0”, very few data), AAOD (quality flag “1”, more abundant data) at the 388 nm wavelength for the “biomass burning” type of aerosol

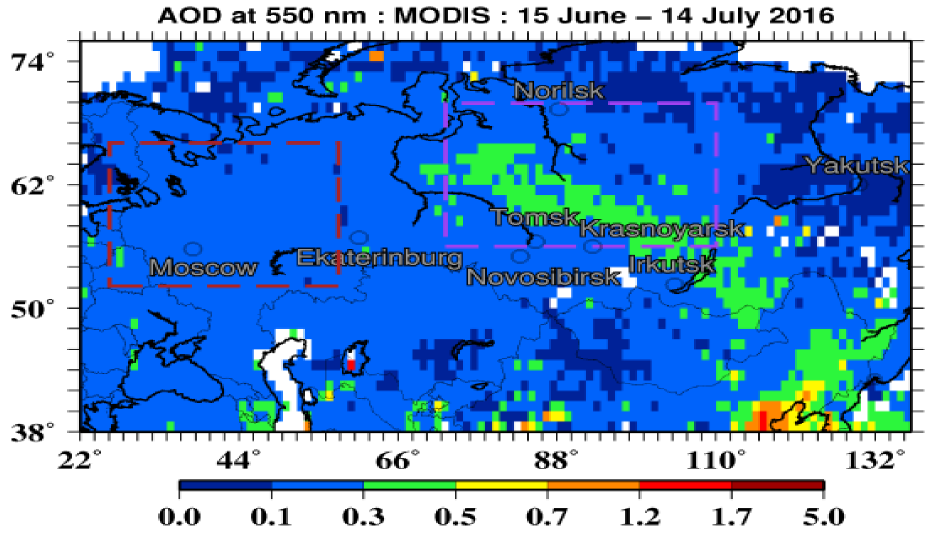
✓ The 550-nm AOD retrievals from the MODIS observations onboard the AQUA и TERRA satellites (MOD04/MYD04 Level-2 data products)

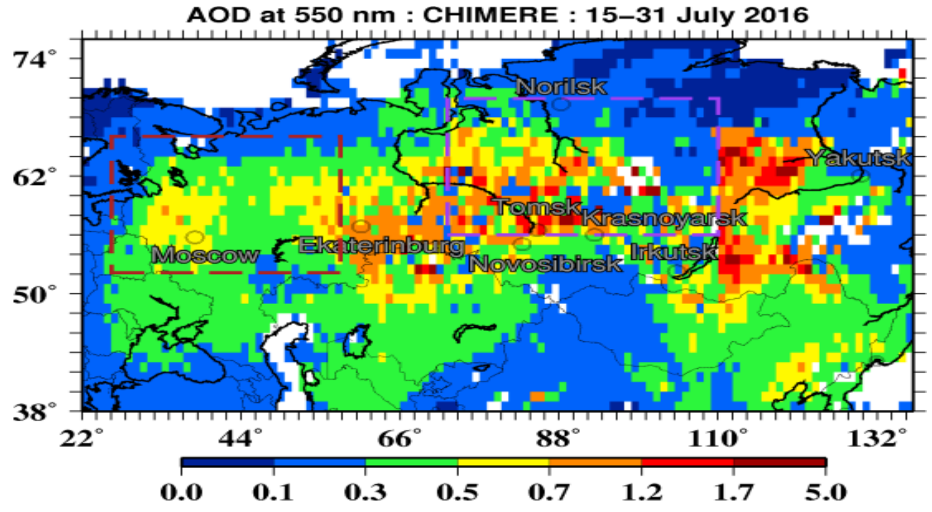
✓ Fire Radiative Power retrieved from the MODIS observations (MOD14/MYD14 Level-2 data products)

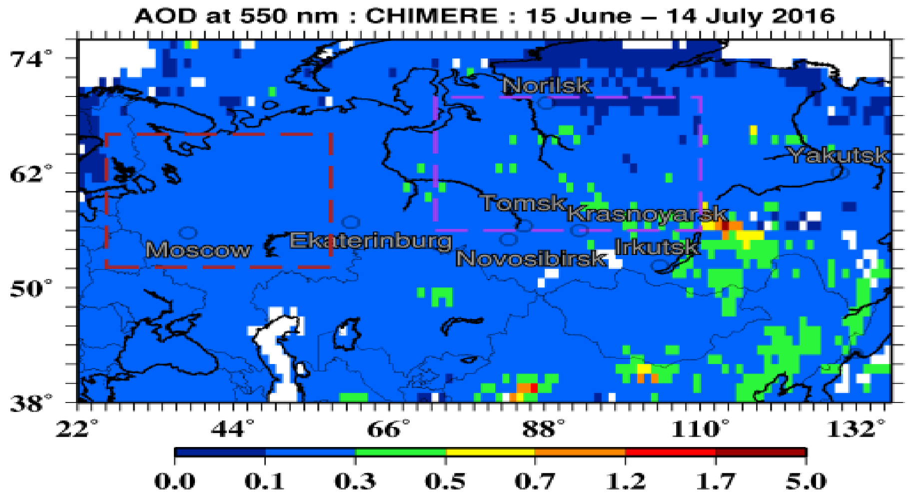
✓ Auxiliary data set: CO column amounts retrieved from the IASI observations onboard the METOP satellite (LATMOS/CNRS and ULB Level-2 data)

Overview of the input data









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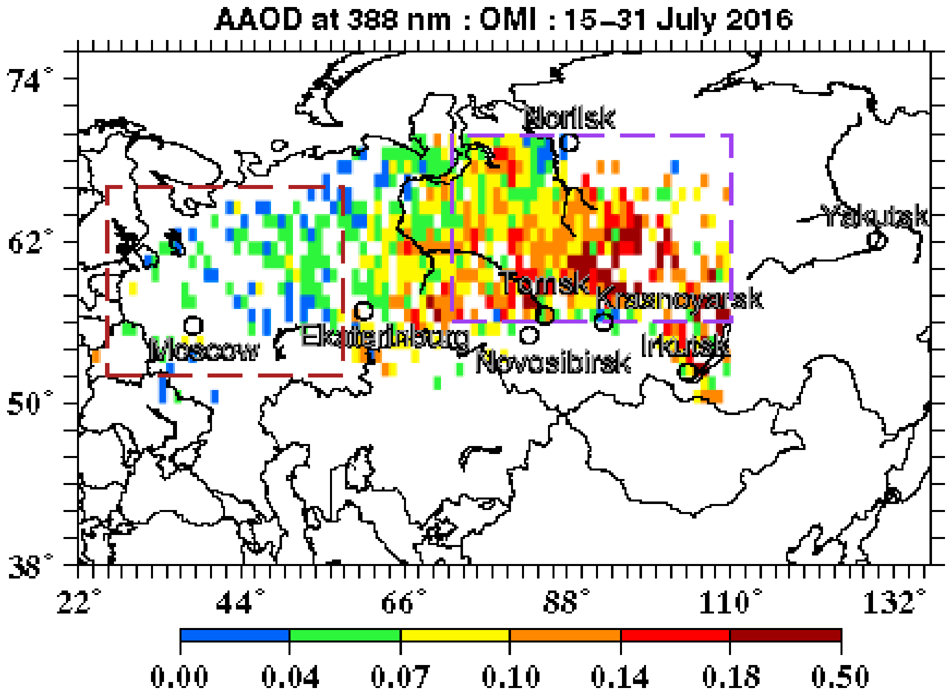

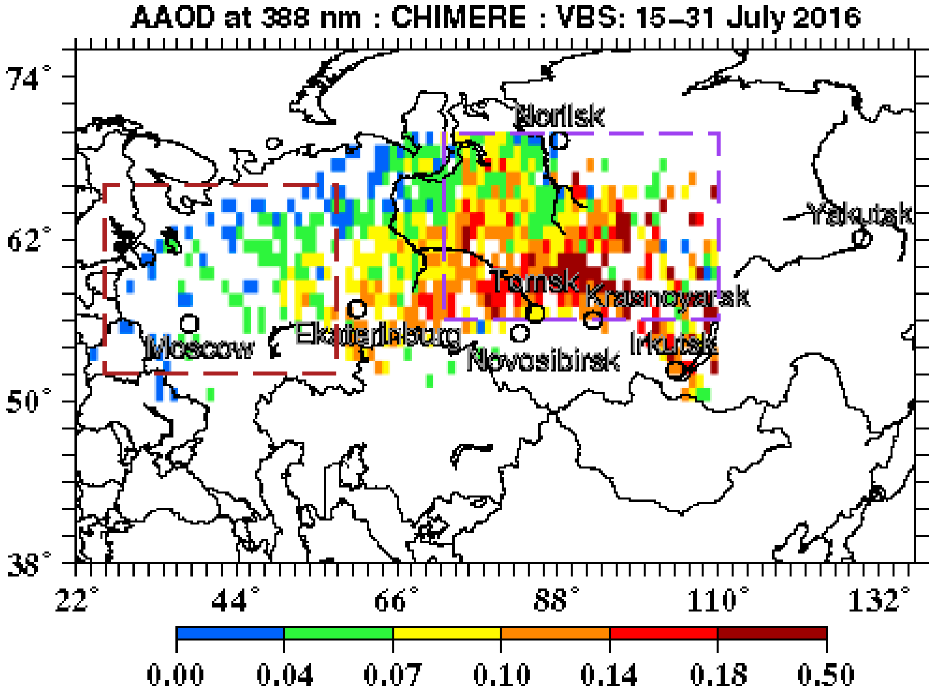

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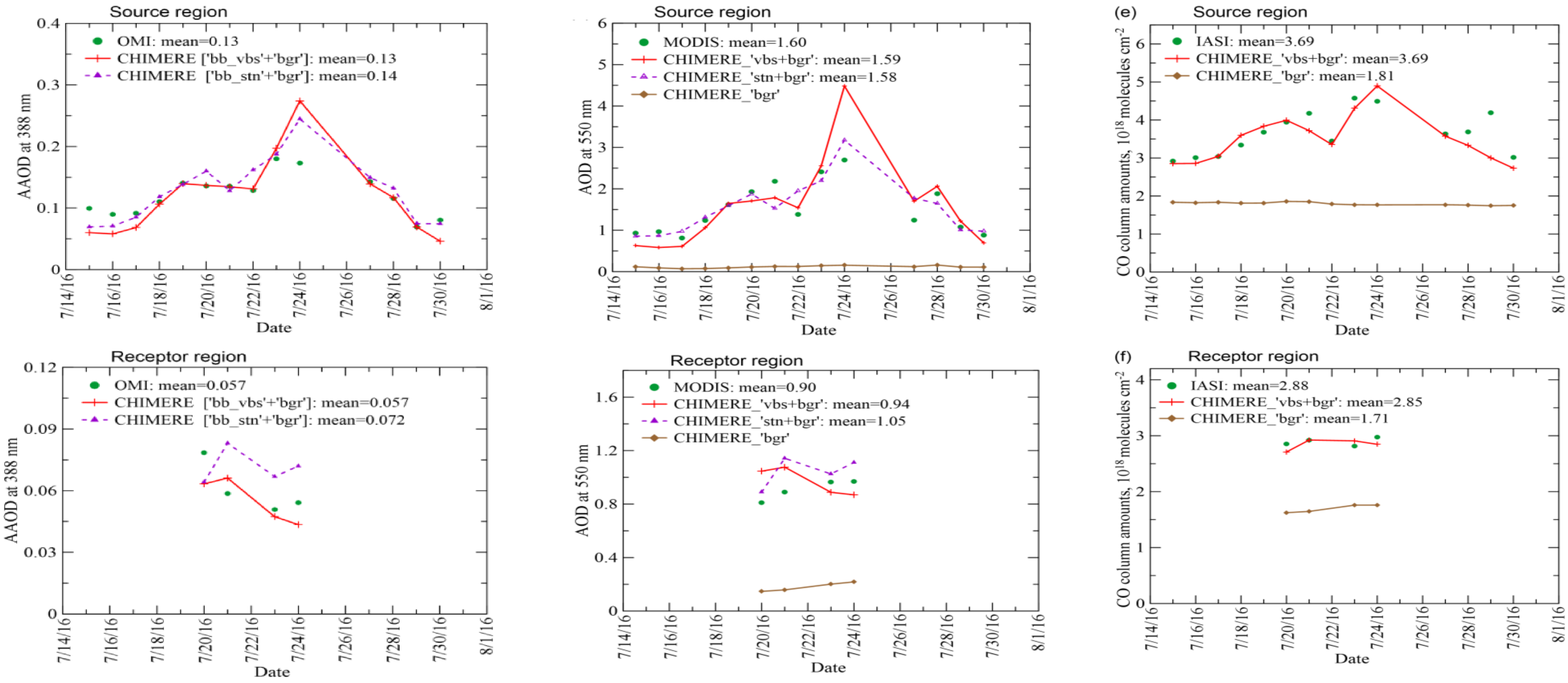
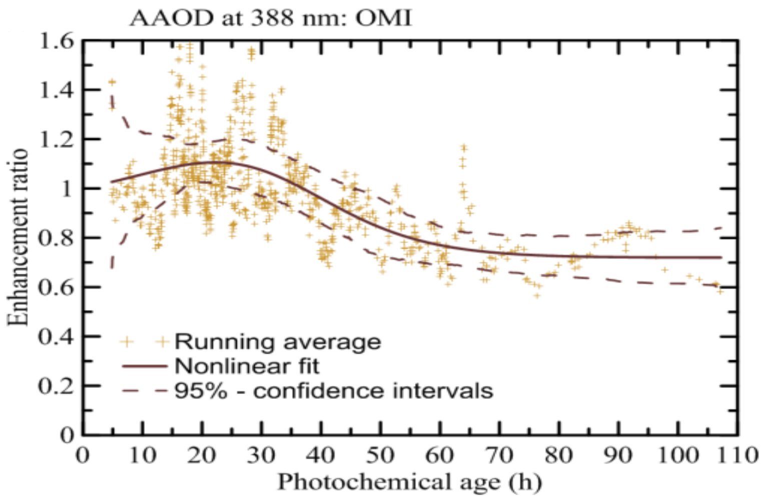
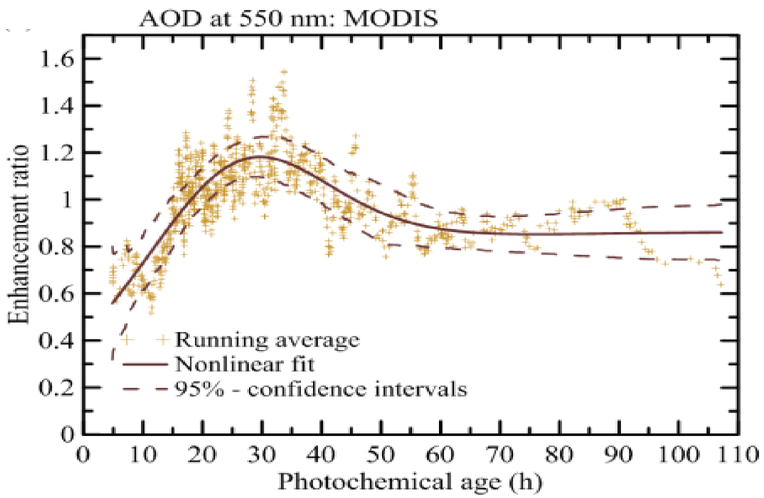
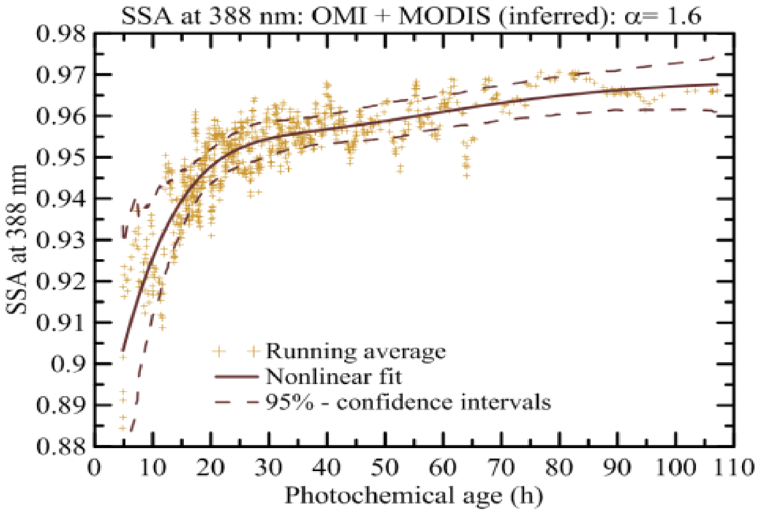
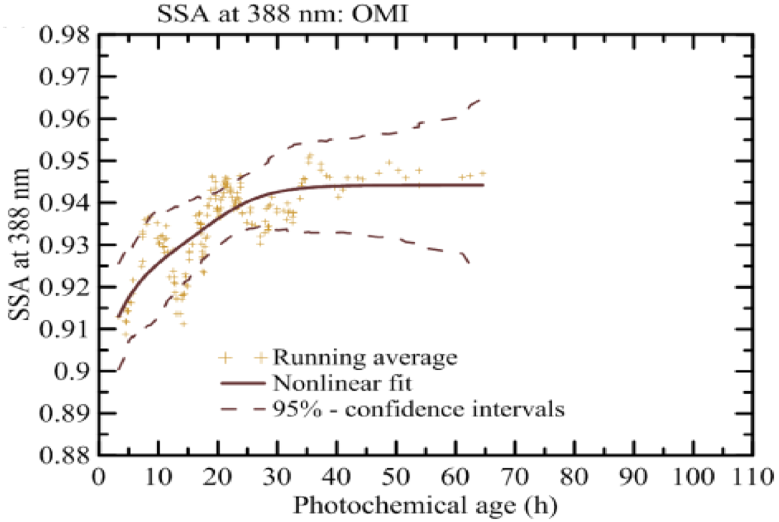


Results: Analysis of the satellite data

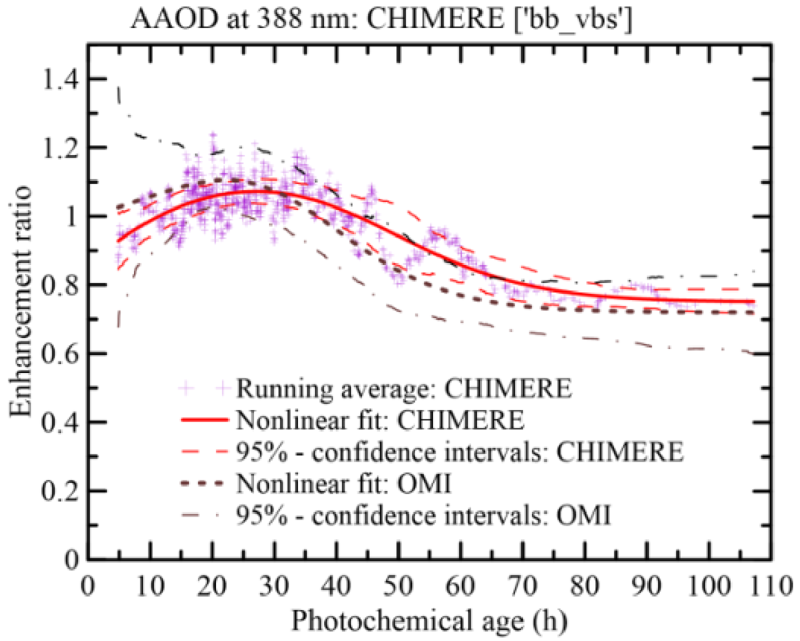


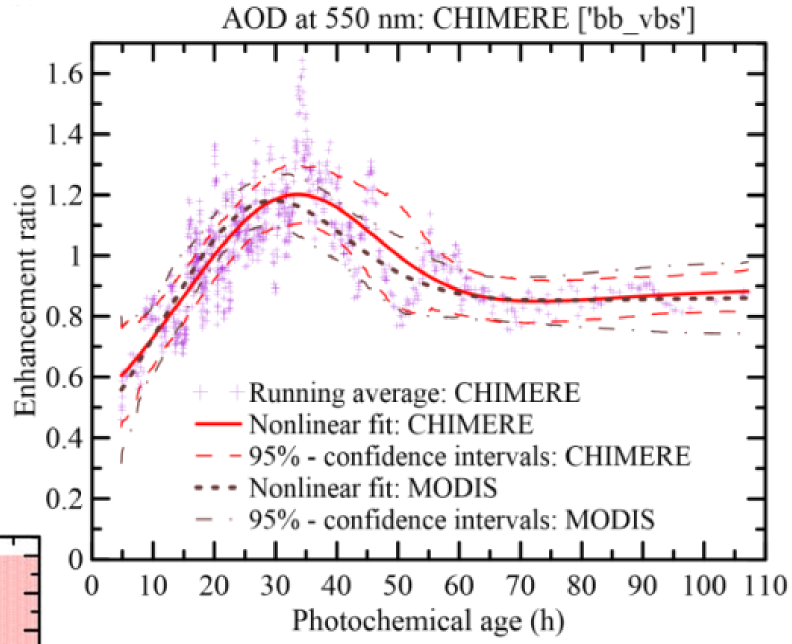


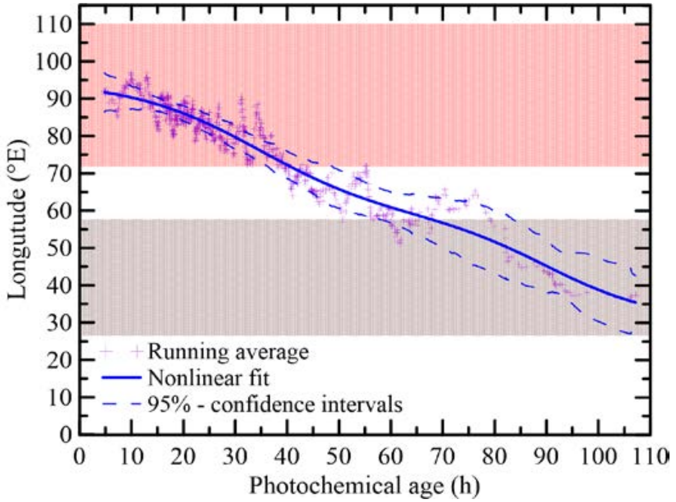




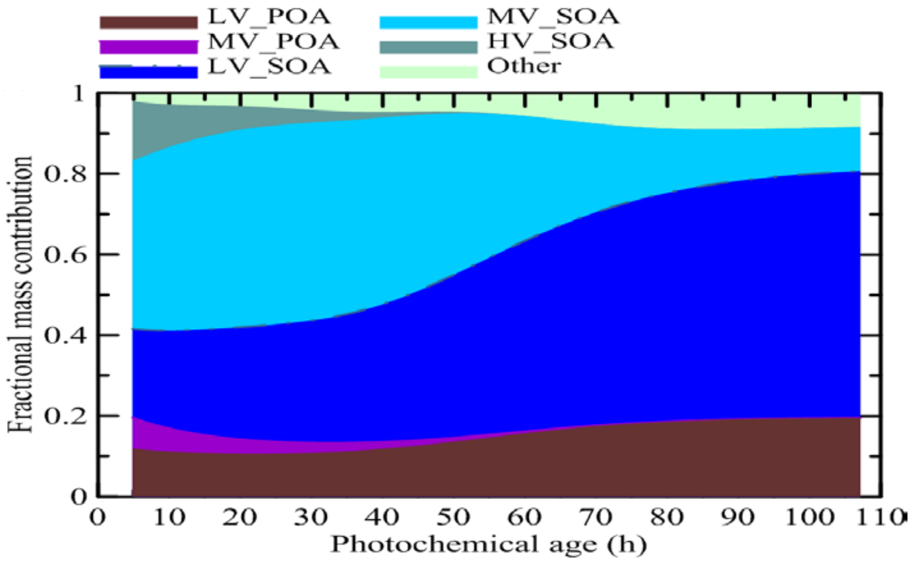
Model results: Trends in AAOD and AOD

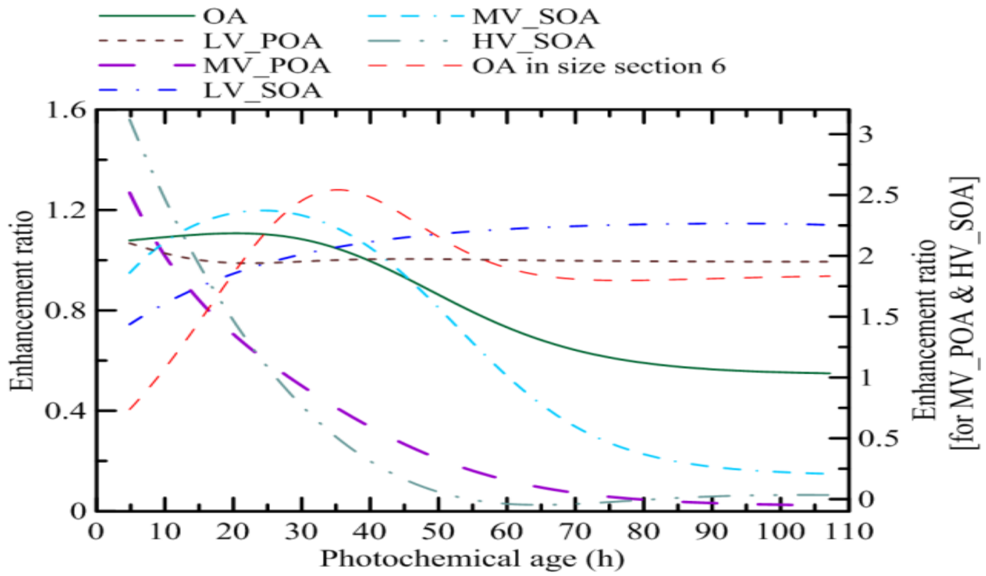


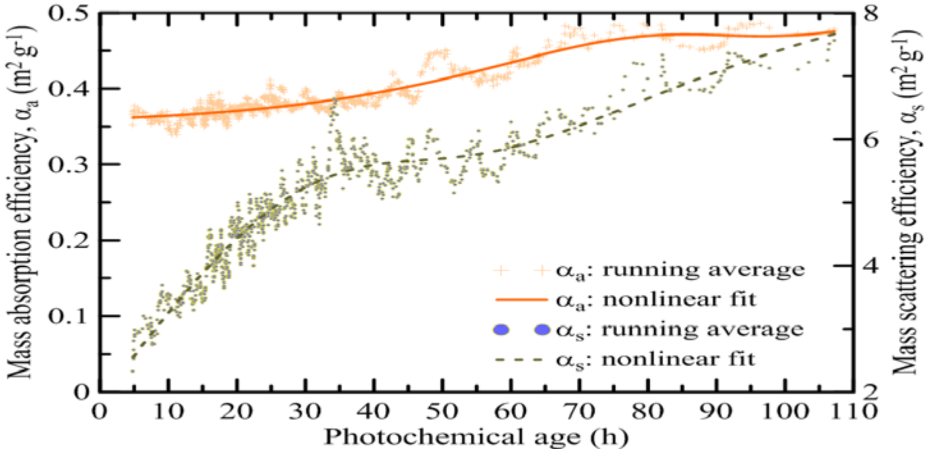


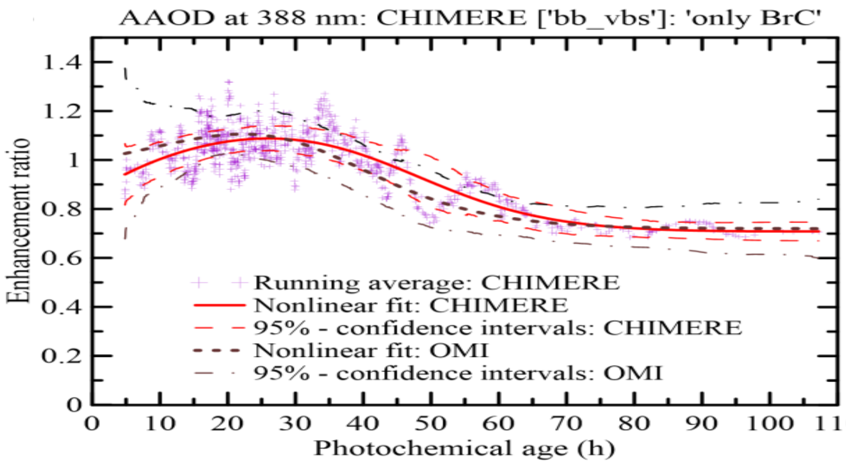


Model results: BB aerosol evolution









Conclusions

❖ We developed a methodology to analyze the atmospheric evolution of BB aerosol by using satellite observations and a chemistry-transport model involving an adjustable VBS scheme.

❖ The dilution-corrected AOD at 550 nm and SSA show significant enhancements while the photochemical age of BB aerosol increased from about 5 until 30 h. Both the AAOD and AOD values corrected for dilution and deposition significantly decline afterwards.

❖ Our simulations indicate that the changes in the BB aerosol optical properties are driven by the OA oxidation processes replacing POA and high-volatility SOA by low-volatility SOA and modifying the particle size distribution. Ensuring adequate representations of these processes in chemistry-transport and climate models is a challenging task.

❖ Further in-situ and aerosol chamber studies of BB aerosol evolution are needed to provide stronger constraints to the simulations.

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