Biomass Burning Dominates Brown Carbon Absorption in Switzerland
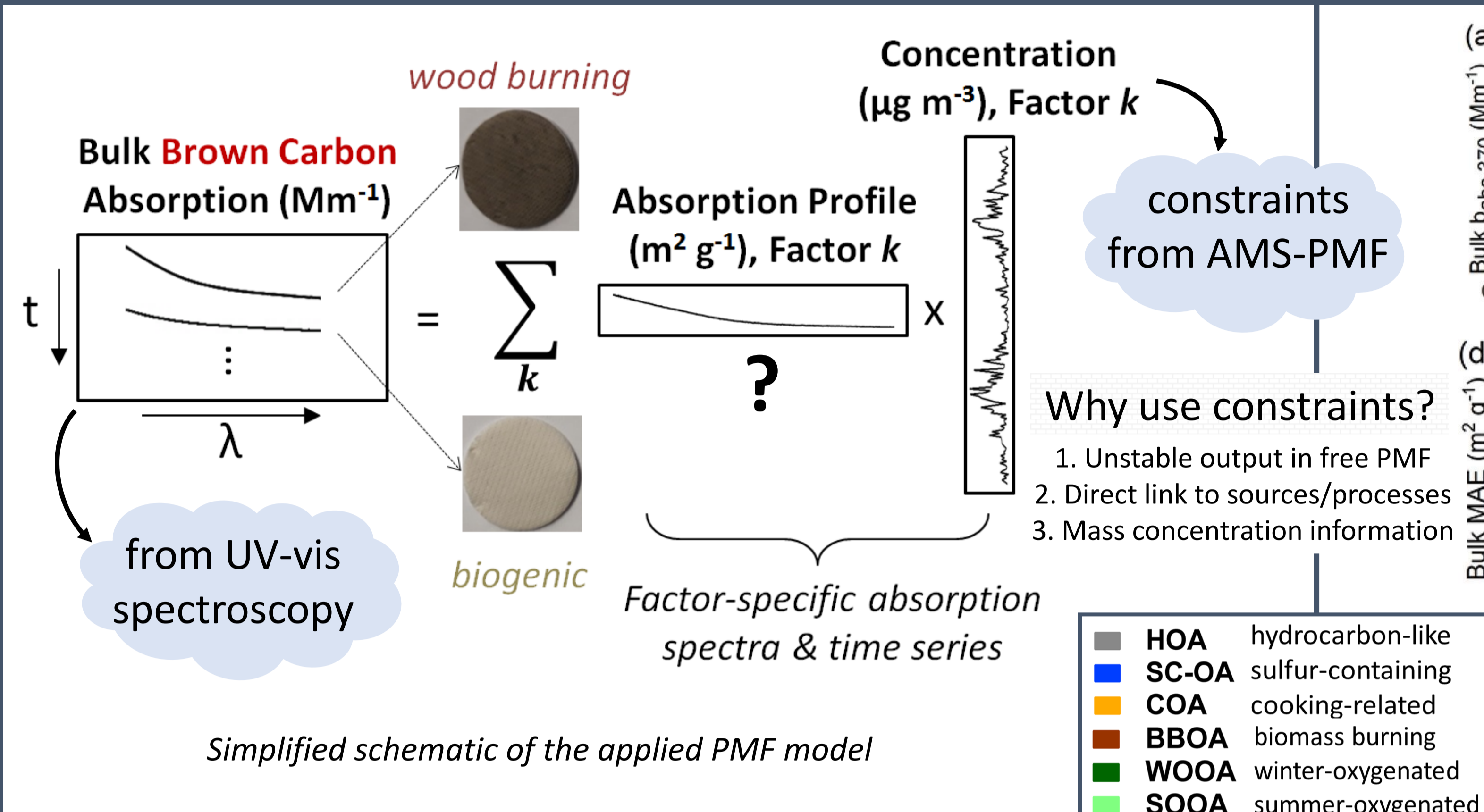
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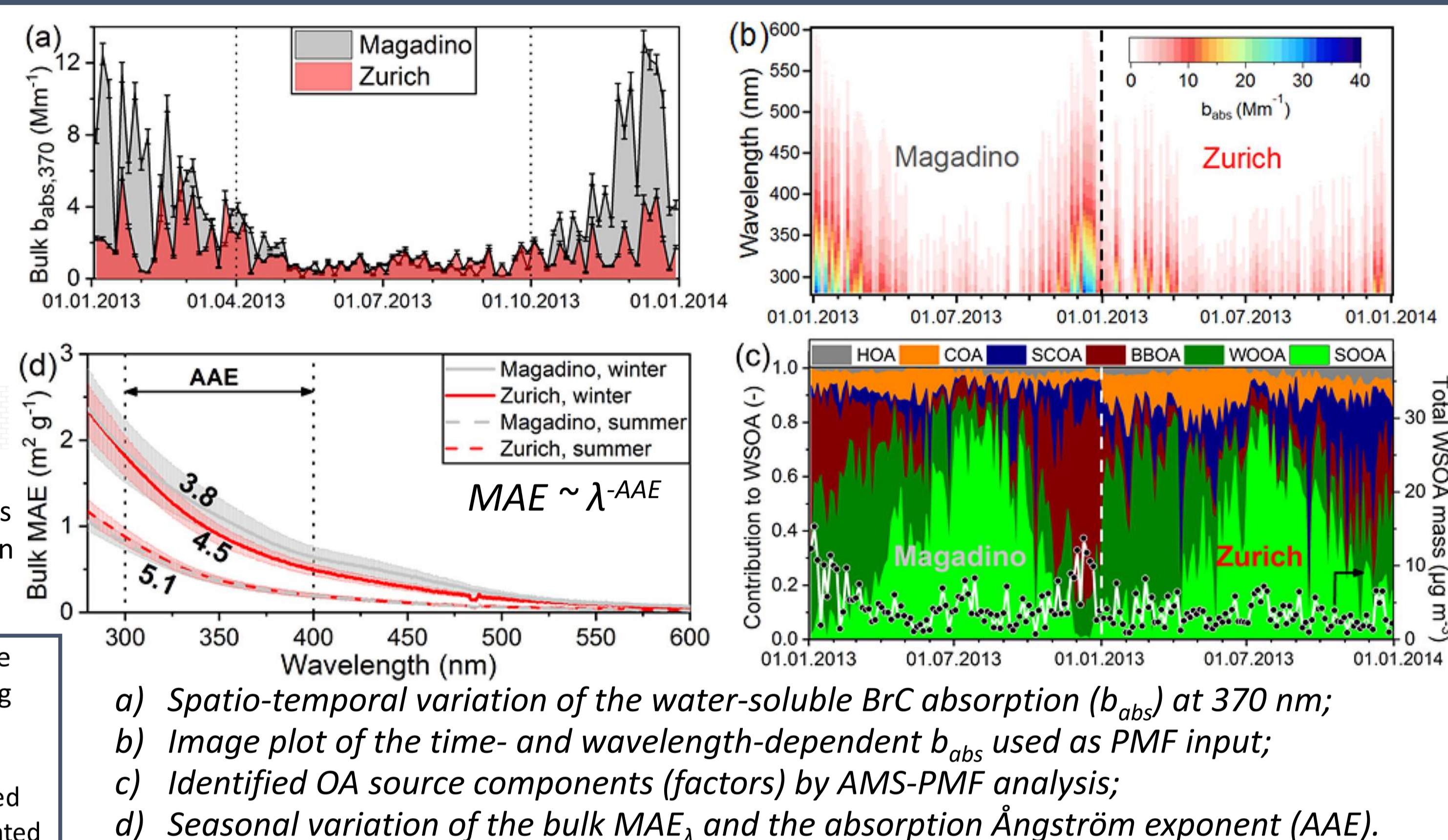
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Introduction

The optical properties and sources of atmospheric aerosols are of prime importance in the context of a changing climate.[1] Organic aerosol (OA) emissions consist of light-absorbing compounds (brown carbon, BrC) that may exert, along with soot carbon, a significant positive direct radiative forcing on the climate system.[2] Current ambient studies of BrC are limited to correlations of the measured absorption with seasonal patterns or marker species specific to certain sources and/or processes (factors).[3] Here, we combine long-term aerosol mass spectrometry (AMS) and ultraviolet-visible (UV-vis) spectroscopy measurements within a positive matrix factorization (PMF) model, to determine factor-specific BrC absorption properties (e.g., mass absorption efficiency, MAE) for a case study in Switzerland.
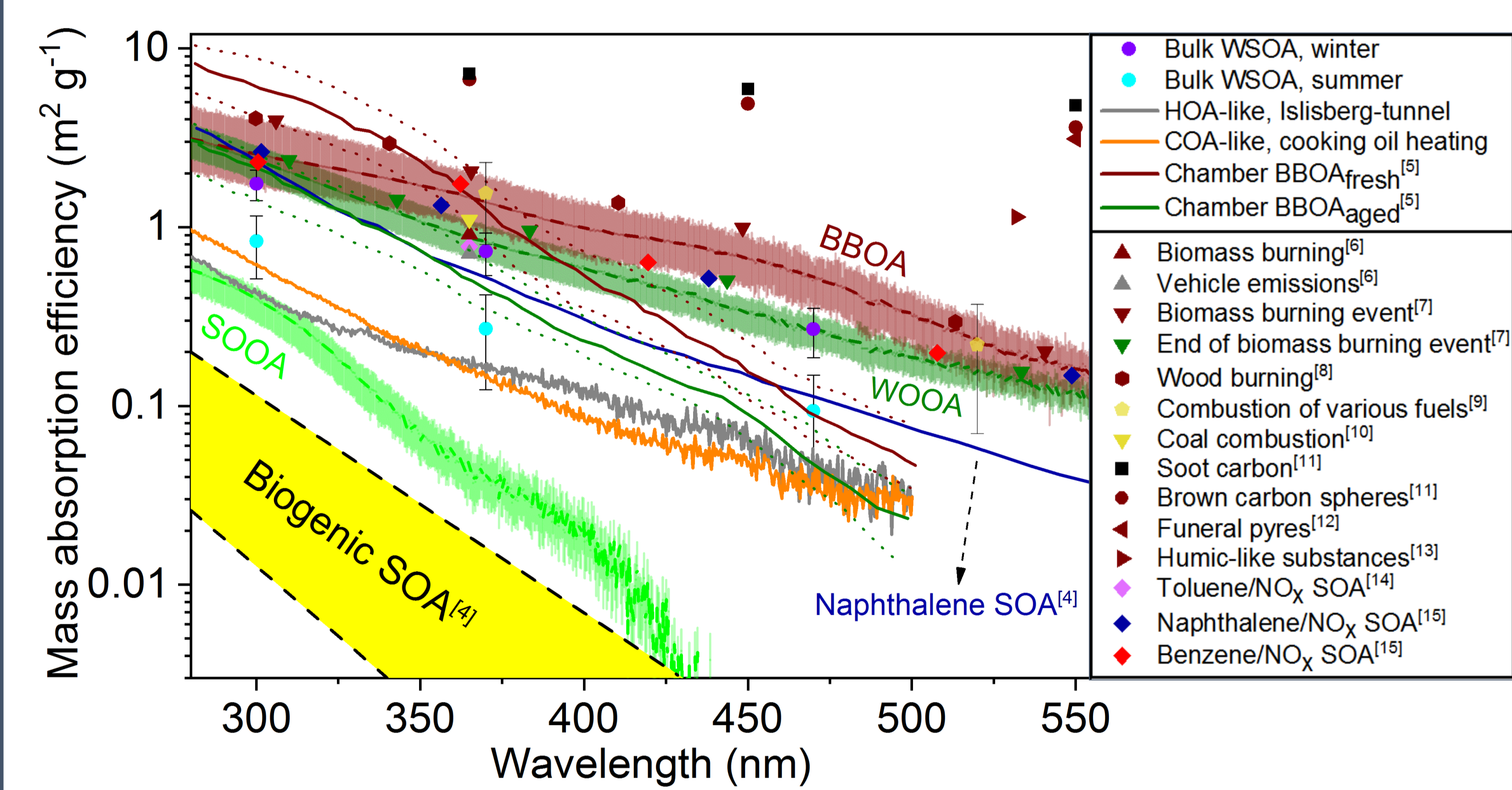
Methodology

wood burning

Bulk Brown Carbon Absorption (Mm⁻¹)

$$\text{Bulk Brown Carbon Absorption} = \sum_k \text{Absorption Profile (m}^2\text{ g}^{-1}\text{), Factor } k \times \text{Concentration (µg m}^{-3}\text{), Factor } k$$

biogenic

from UV-vis spectroscopy

constraints from AMS-PMF

Why use constraints?
1. Unstable output in free PMF
2. Direct link to sources/processes
3. Mass concentration information

Factor-specific absorption spectra & time series

Simplified schematic of the applied PMF model

HOA hydrocarbon-like
SC-OA sulfur-containing
COA cooking-related
BBOA biomass burning
WOOA winter-oxygenated
SOOA summer-oxygenated

Bulk absorption and OA sources

$$MAE \sim \lambda^{-AAE}$$

a) Spatio-temporal variation of the water-soluble BrC absorption (b_{abs}) at 370 nm;
b) Image plot of the time- and wavelength-dependent b_{abs} used as PMF input;
c) Identified OA source components (factors) by AMS-PMF analysis;
d) Seasonal variation of the bulk MAE_λ and the absorption Ångström exponent (AAE).

Source apportionment of BrC absorption

Legend:
- Bulk WSOA, winter
- Bulk WSOA, summer
- HOA-like, Islisberg-tunnel
- COA-like, cooking oil heating
- Chamber BBOA_fresh [5]
- Chamber BBOA_aged [5]
- Biomass burning [6]
- Vehicle emissions [6]
- Biomass burning event [7]
- End of biomass burning event [7]
- Wood burning [8]
- Combustion of various fuels [9]
- Coal combustion [10]
- Soot carbon [11]
- Brown carbon spheres [11]
- Funeral pyres [12]
- Humic-like substances [13]
- Toluene/NOₓ SOA [14]
- Naphthalene/NOₓ SOA [15]
- Benzene/NOₓ SOA [15]
- Naphthalene SOA [4]
- Biogenic SOA [4]

Comparison of the modeled MAE_k spectra to those of chamber-generated fresh and aged wood burning BrC, mixed-source ambient samples and laboratory experiments employing anthropogenic and biogenic SOA precursors.
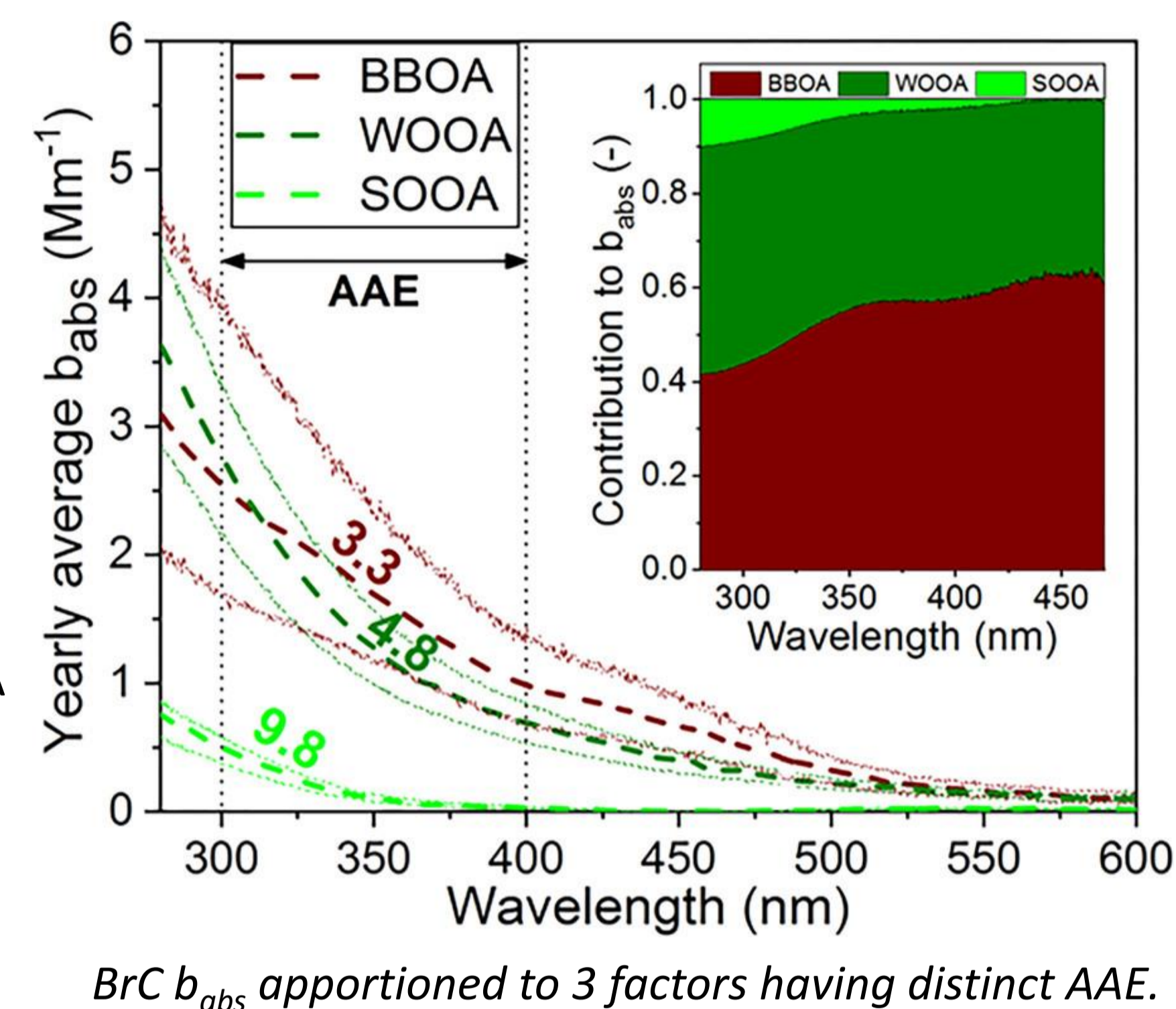
Absorbing factors:

BBOA
polar aromatics from wood burning, absorption hump in the visible range

WOOA
highly oxidized, non-fossil, long-range transported SOA or aged BBOA emissions

SOOA
temperature-driven, biogenic-dominated, higher MAE than in chambers
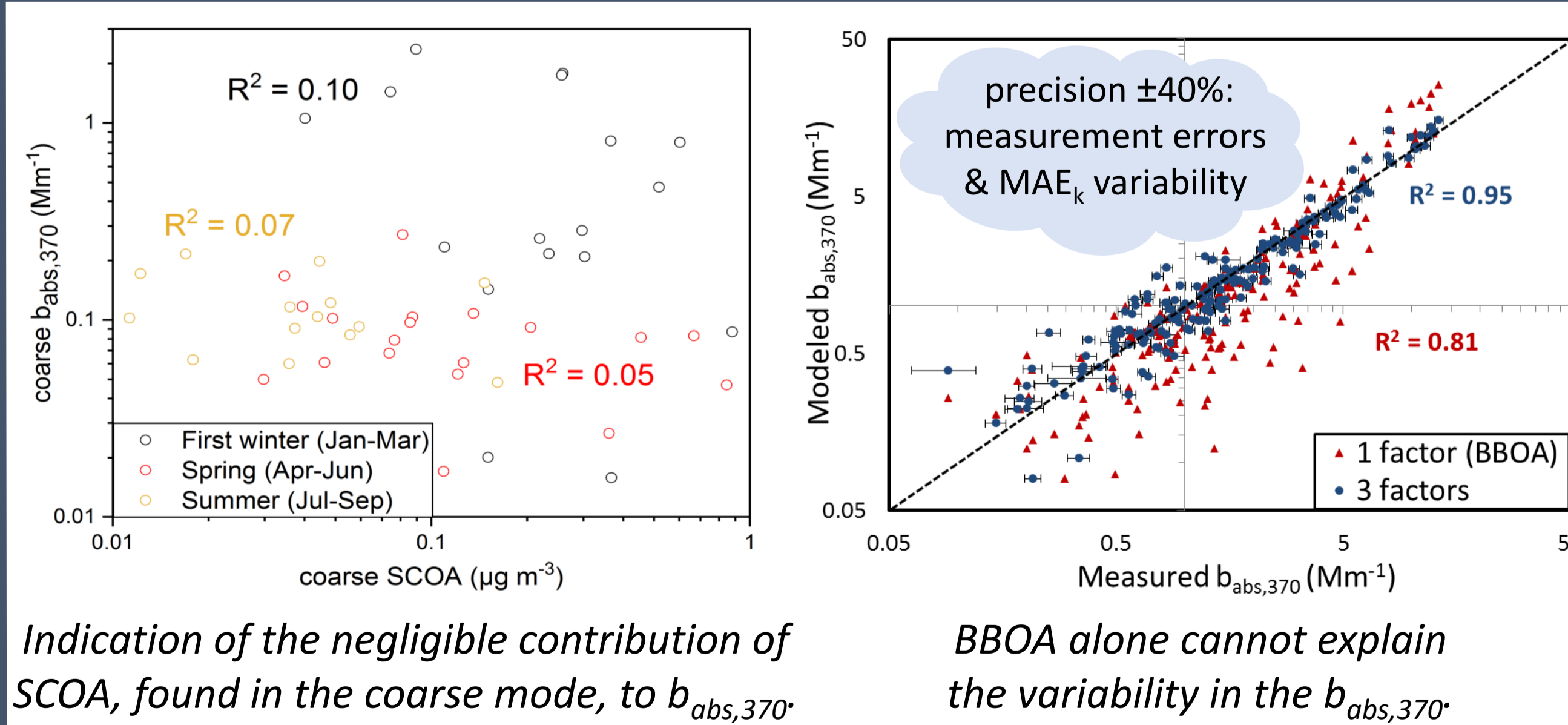

BrC b_{abs} apportioned to 3 factors having distinct AAE.

References

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Model validation

precision ±40%: measurement errors & MAE_k variability

Indication of the negligible contribution of SCOA, found in the coarse mode, to $b_{abs,370}$.

BBOA alone cannot explain the variability in the $b_{abs,370}$.

Relative factor contributions to bulk BrC b_{abs} in Magadino and Zurich during 2013, calculated by post-PMF analysis.

Acknowledgements

Conclusions

1. Novel framework that provides a direct link between chemical composition and absorptivity for individual OA classes.
2. Long-term predominance of anthropogenic over biogenic emissions in the water-soluble BrC absorption in Switzerland.
3. Importance of BrC versus soot carbon absorption should be examined to deepen our understanding of their climate effects.