Effect of absorbing organic carbon on global oxidation

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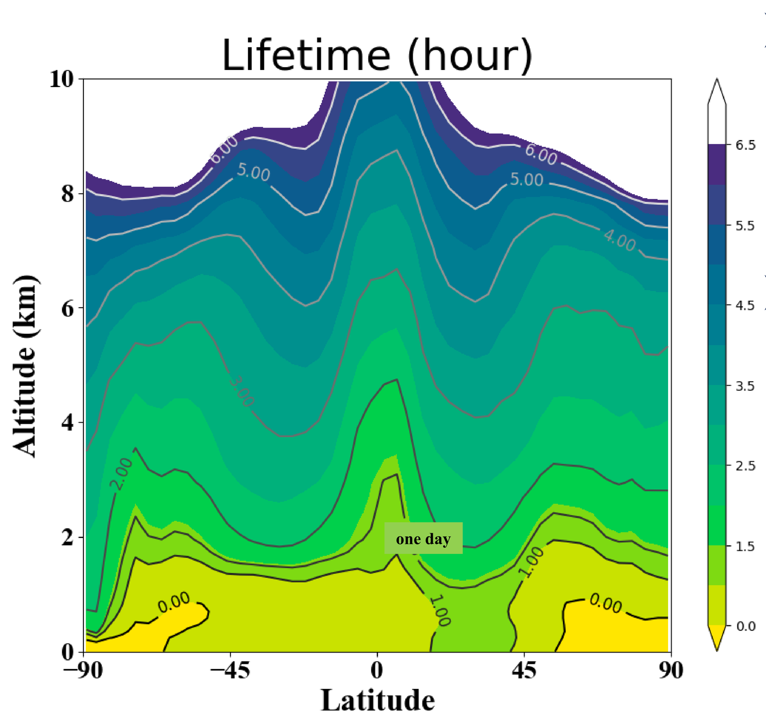
◆Background

- Brown carbon (BrC) is light-absorbing organic carbon, which has a significant global impact on climate, atmospheric chemistry, and health. Emissions from wildfires are a major source of BrC globally. Due to its absorption in the near ultraviolet and visible wavelengths, BrC affects the rates of photolysis reactions, thus perturbing the formation of ozone and OH, key oxidative species in the atmosphere.
- Photobleaching and oxidative whitening occur when BrC is a day or more removed from it's source. Thus accurate calculation of BrC lifetime is crucial for the simulation results.

◆Simulation Experiments

- We use the GEOS-Chem to evaluate the chemical effect of BrC on global oxidation with a horizontal resolution of 4° × 5° with 47 vertical levels. All biomass burning OC from GFED4s is simulated as BrC.
- We implement a temperature- and humidity- dependent chemical aging scheme based on recent publications, which leads to longer BrC whitening lifetime in the upper troposphere than the previous implementation of 1 days.

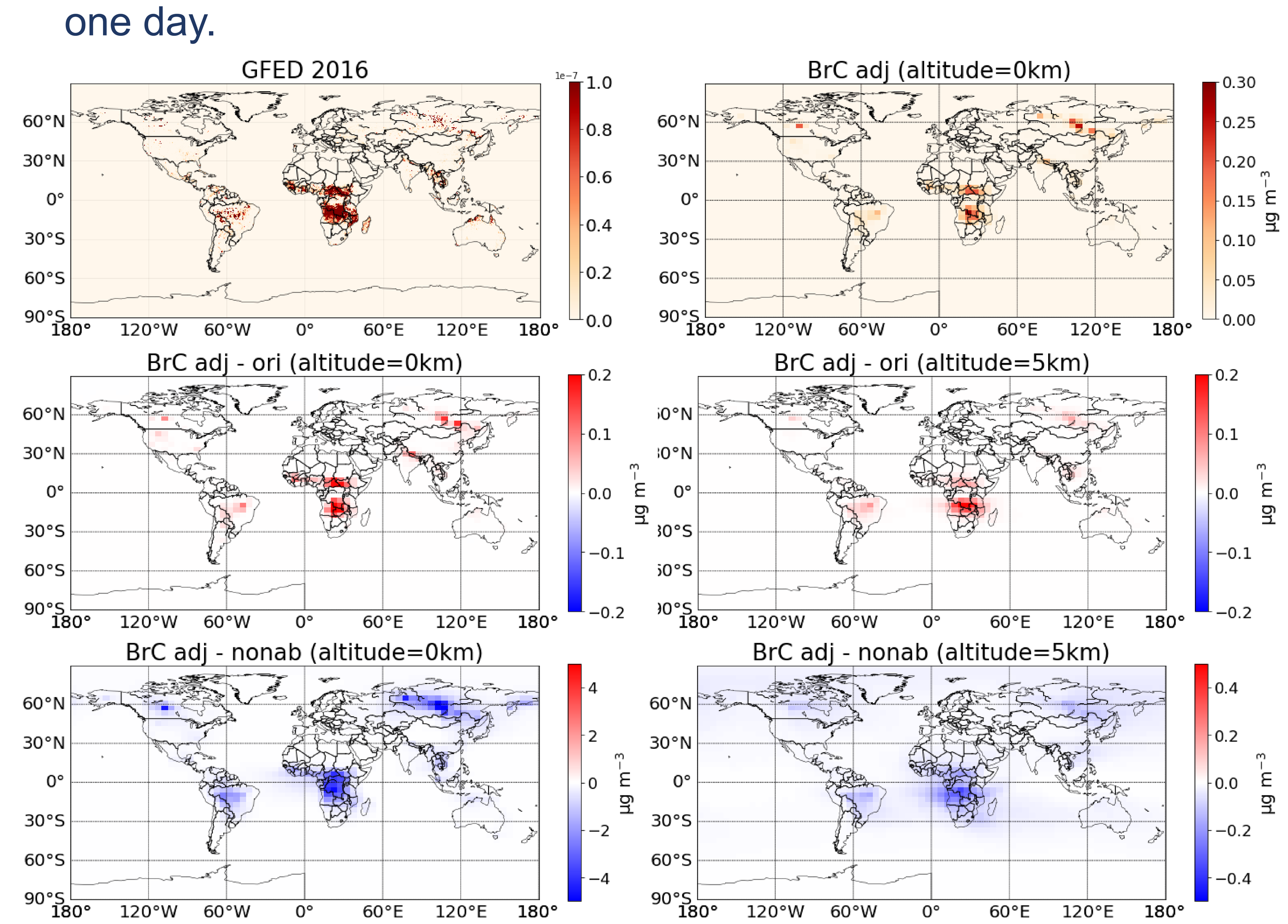


- Plume rise has a significant impact on vertical distribution of BrC, so, the GFED 3D has been taken on.
- For now, we simulated three experiments, two of them have different lifetime, and the other assumes that BrC does not absorb light, i.e. there is no bleaching process (like OC).

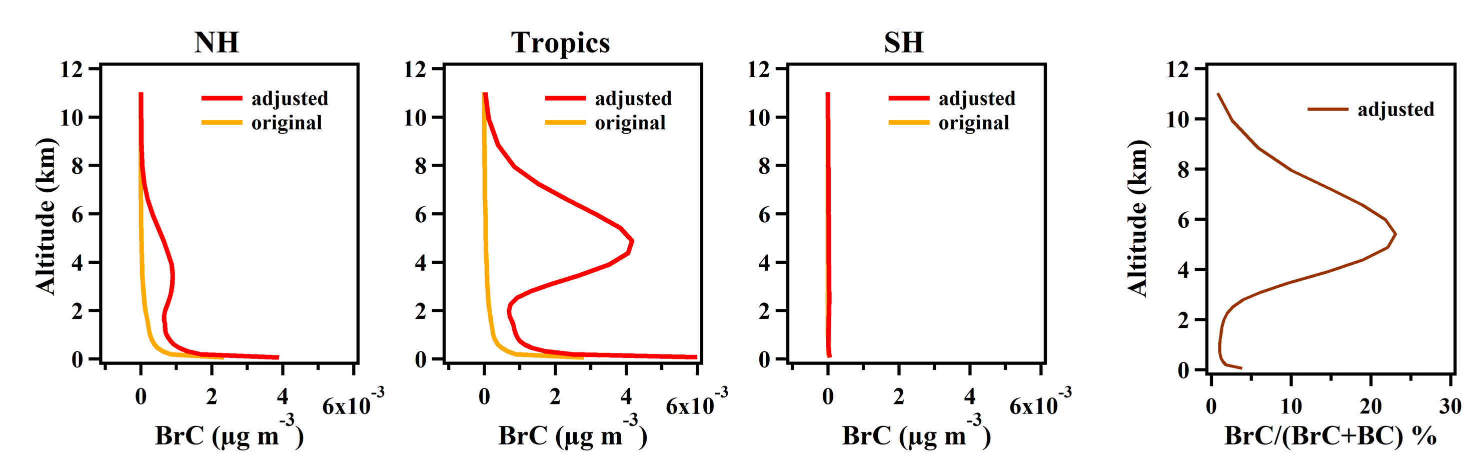
simulations	Description (year of 2016)
Original Simu	BrC lifetime ≈ 1 day.
Adjusted Simu	BrC lifetime adjusted by RH and temperature (min = 1 day).
Nonab Simu	BrC but non absorbing.

◆Emissions and concentration of BrC

- The results show that when the lifetime is considered, the concentration of BrC that really has an effect on atmospheric radiation is significantly reduced compared to the non-absorbent condition. After lifetime adjustment, the concentration of BrC increased at both ground and altitude compared to the lifetime of just one day.

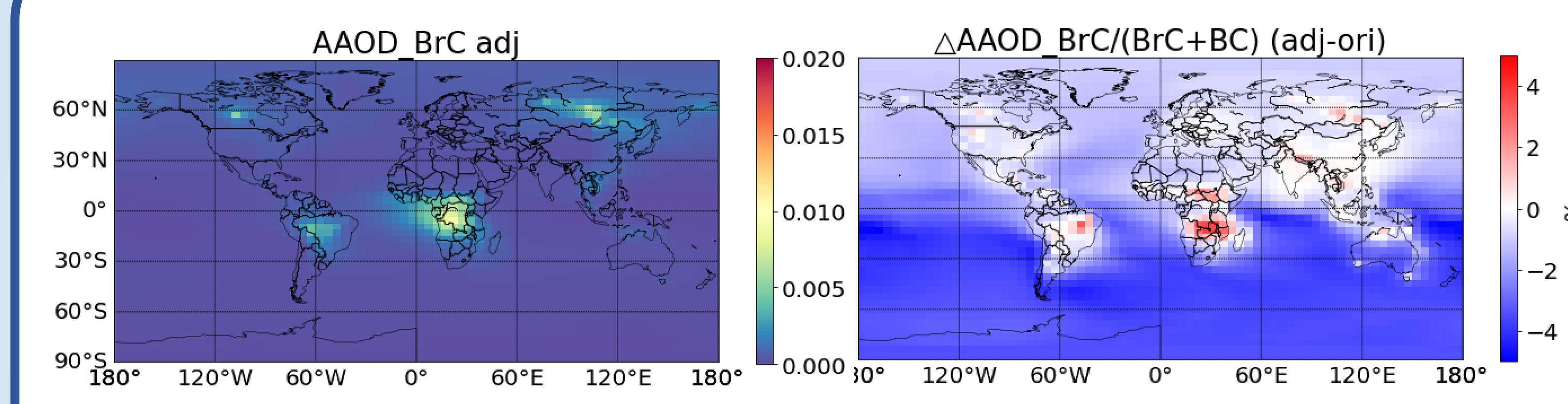


- As can be seen from the figure below, the concentration difference caused by lifetime is most significant at an altitude of about 5km. And the BrC concentration can reach more than 20% of BC.



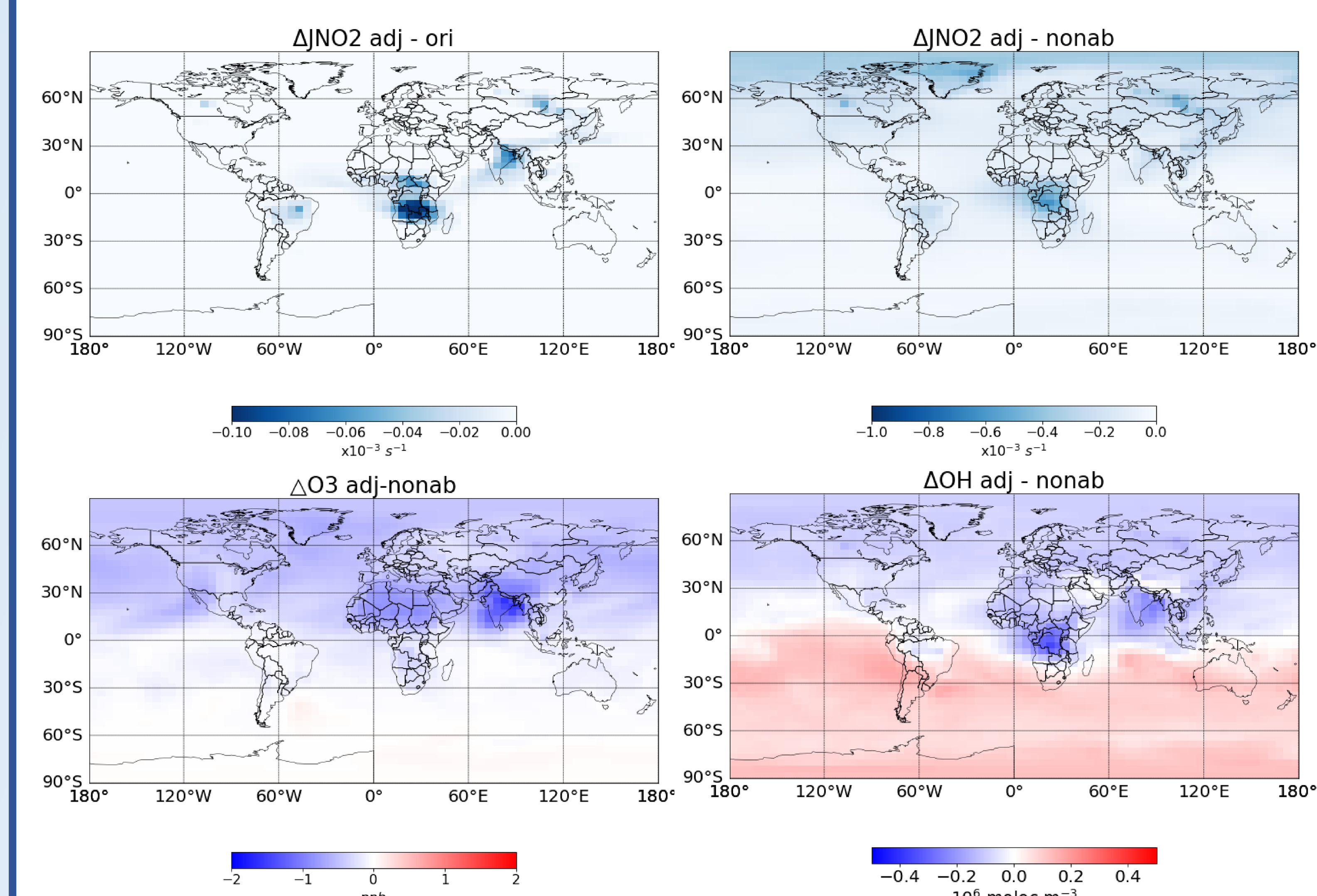
◆Light absorption of BrC

- The absorption AOD (AAOD) distribution of BrC is consistent with its concentration distribution. According to △AAOD, it can be found that in the hot spots of BrC, the proportion of absorption from BrC increases with the adjustment of lifetime.



◆Impacts on atmospheric oxidation from BrC

- The increase of the BrC concentration especially in the upper troposphere can lead to the decreasing of photolysis rate like JNO_2, which consist with BrC concentration distribution. In addition, due to the effect of light absorption properties of BrC on the photolysis rate, the ozone concentration and OH in the northern Hemisphere are reduced to a certain extent.



◆Further works

- Firstly, Observational data needs to be collected to constrain the model (e.g. satellite data such as OMI that includes AAOD or observational data that includes absorption coefficient).
- Secondly, the process and mechanism of wildfire development affecting atmospheric oxidation through BrC need to be further studied.

References

1. Wang, X.; Heald, C. L.; Liu, J.; Weber, R. J.; Campuzano-Jost, P.; Jimenez, J. L.; Schwarz, J. P.; Perring, A. E. Exploring the observational constraints on the simulation of brown carbon. Atmos Chem Phys 2018, 18 (2), 635-653.
2. Zhang, Y.; Forrister, H.; Liu, J.; Dibb, J.; Anderson, B.; Schwarz, J. P.; Perring, A. E.; Jimenez, J. L.; Campuzano-Jost, P.; Wang, Y.; Nenes, A.; Weber, R. J. Top-of-atmosphere radiative forcing affected by brown carbon in the upper troposphere. Nature Geoscience 2017, 10 (7), 486-489.
3. Schnitzler, E. G.; Gerrebos, N. G. A.; Carter, T. S.; Huang, Y.; Heald, C. L.; Bertram, A. K.; Abbatt, J. P. D. Rate of atmospheric brown carbon whitening governed by environmental conditions. Proc Natl Acad Sci U S A 2022, 119 (38), e2205610119.

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