

Representing biomass burning aerosols in models



Jeff Pierce
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Key contributors

Nicole June



Kim Sakamoto

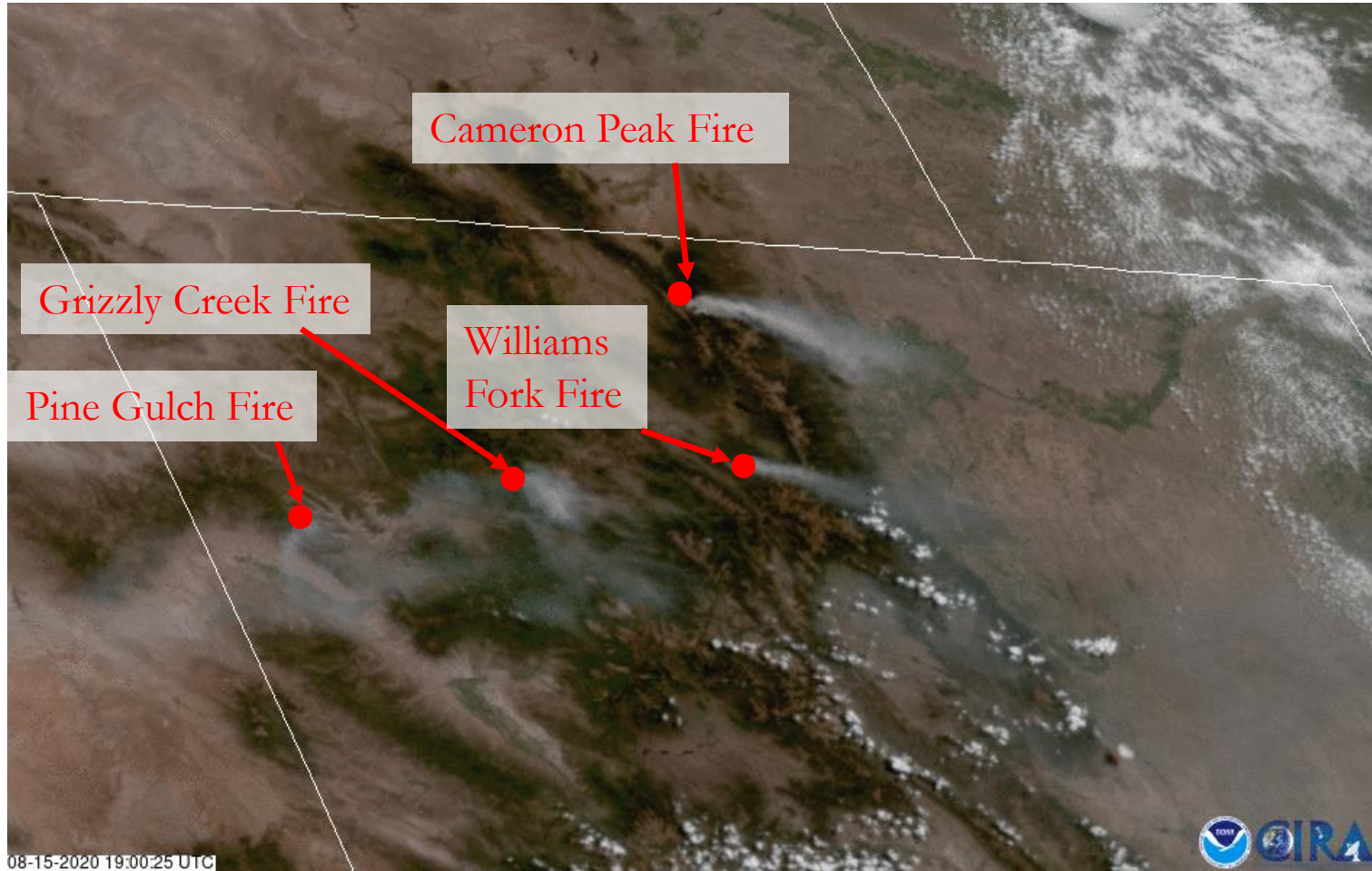


Emily Ramnarine



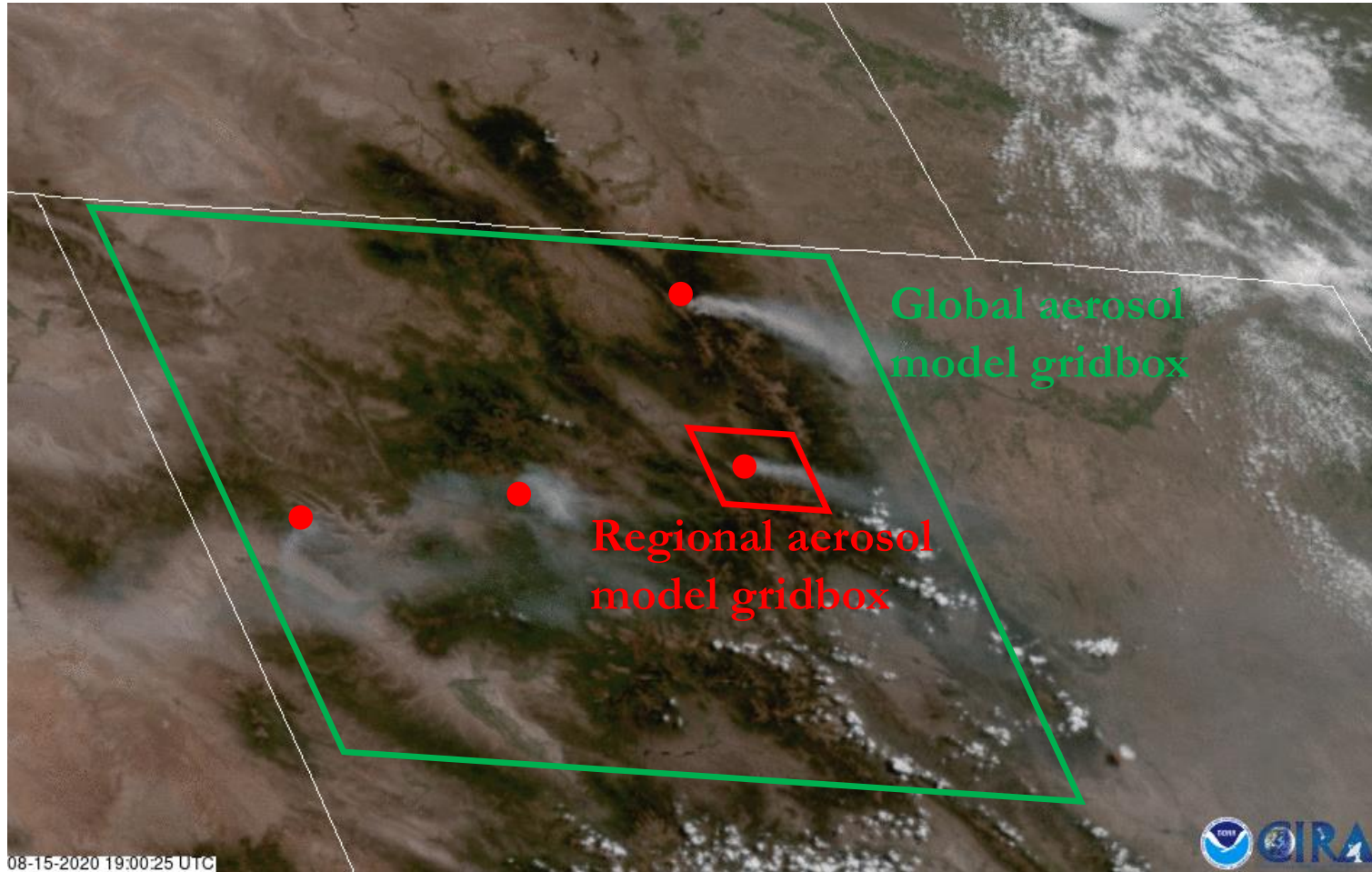
Lots of other amazing collaborators!

Smoke is super complicated...
spatially and temporally.



Colorado, Aug 15, 2020.
View from GOES-West.

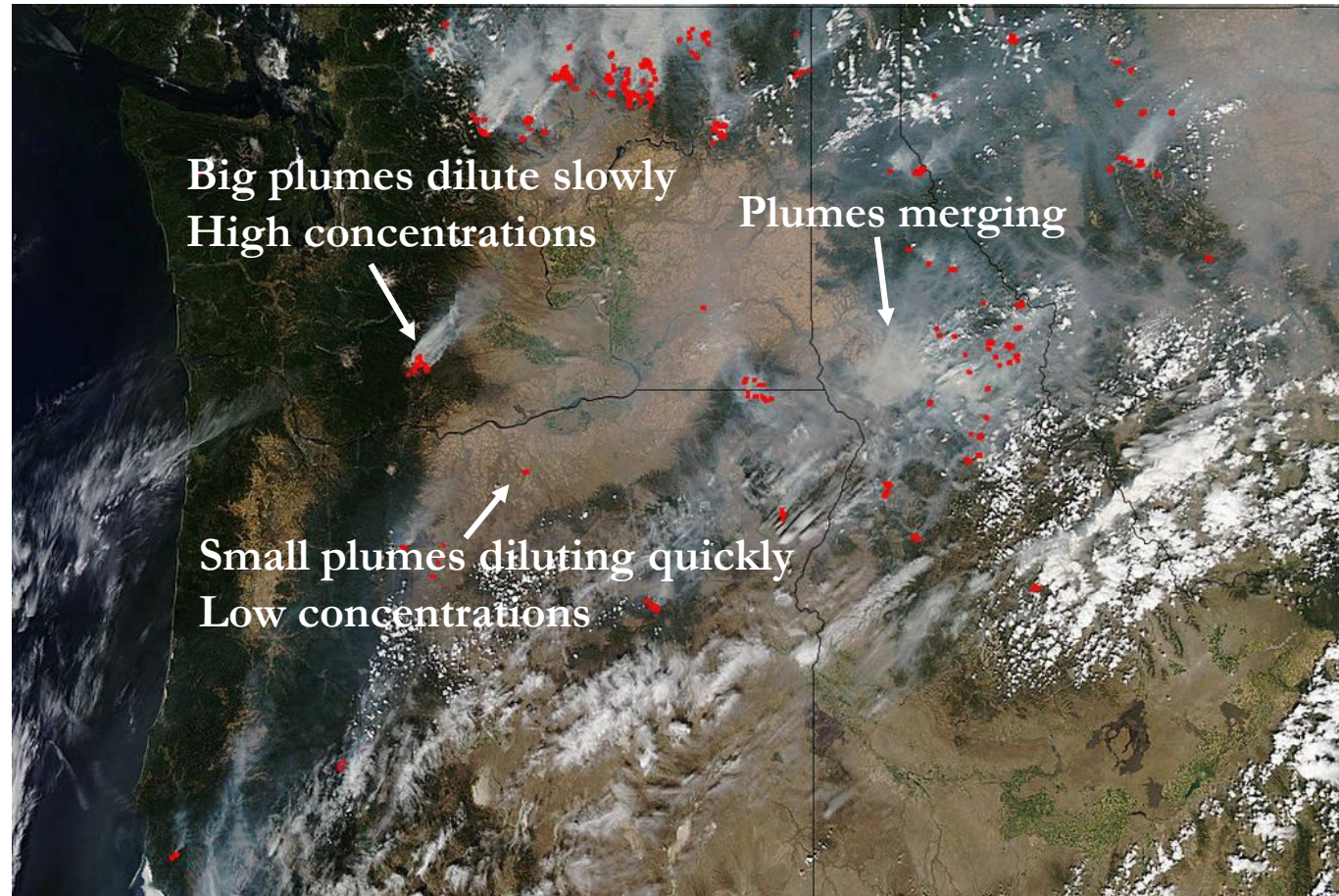
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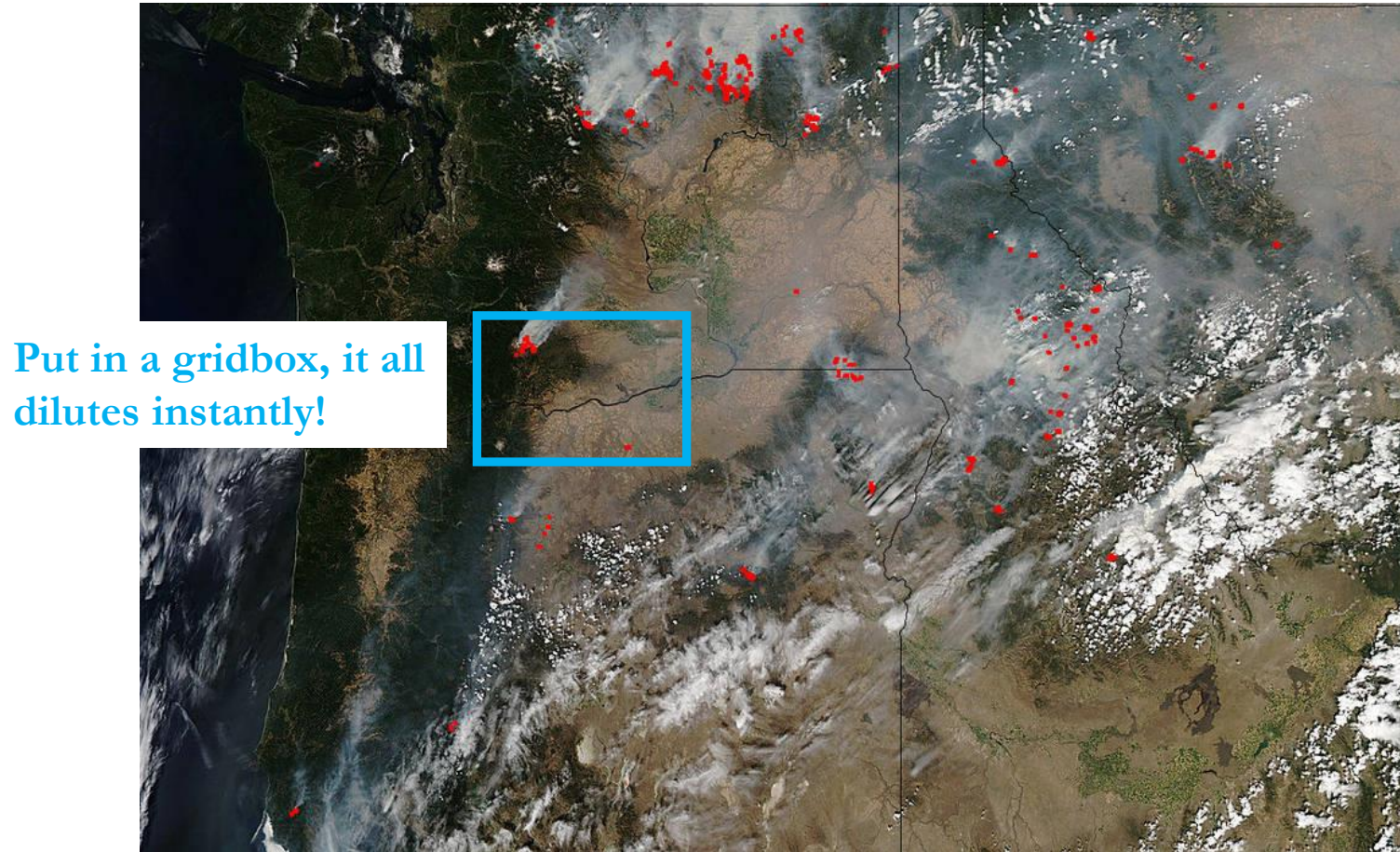
- Subgrid concentration variability
- Plume injection height
- Smoke color
- Temporal variability of emissions
- Different plume sizes and dilution rates

Plumes from small fires dilute faster than plumes from large fires.



NASA Aqua-MODIS, Pacific Northwest fires, Aug. 15, 2015.
NASA image courtesy Jeff Schmaltz, MODIS Rapid Response Team.

...but it all dilutes instantly when emitted into gridboxes.



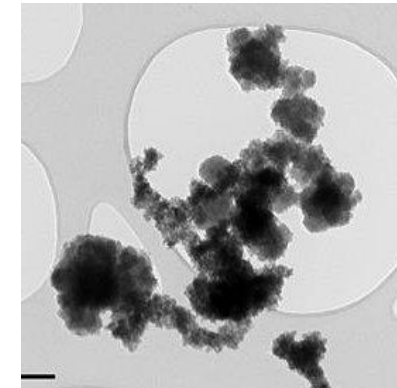
NASA Aqua-MODIS, Pacific Northwest fires, Aug. 15, 2015.
NASA image courtesy Jeff Schmaltz, MODIS Rapid Response Team.

Which details matter depends on your simulation goal?

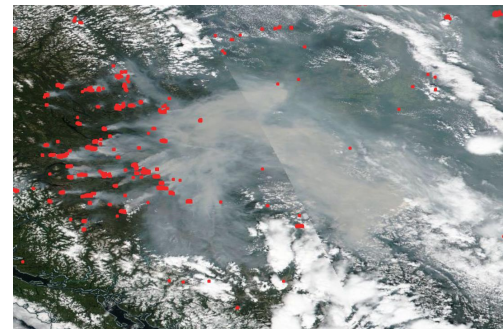
- Do you care about...
 - Surface PM concentrations/forecasts



- Detailed aerosol physical and chemical properties

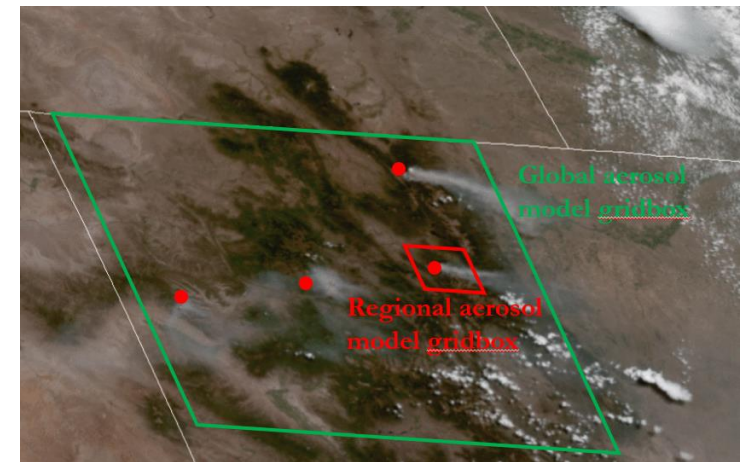


- Direct and indirect radiative forcings



Some of the challenges in simulating smoke:

- Plume injection height (PIH)
 - How much is at the surface?
 - Does this affect aerosol lifetime?
 - Are particles above/below clouds?
- Subgrid-scale plumes
 - Non-linear physics and chemistry
 - Coagulation
 - Primary Organic Aerosol (POA) evaporation, Secondary Organic Aerosol (SOA) formation
 - Errors in surface PM predictions at subgrid locations
- Emissions (daily cycle, amount)
- Composition
 - Absorption
 - Health/toxicity

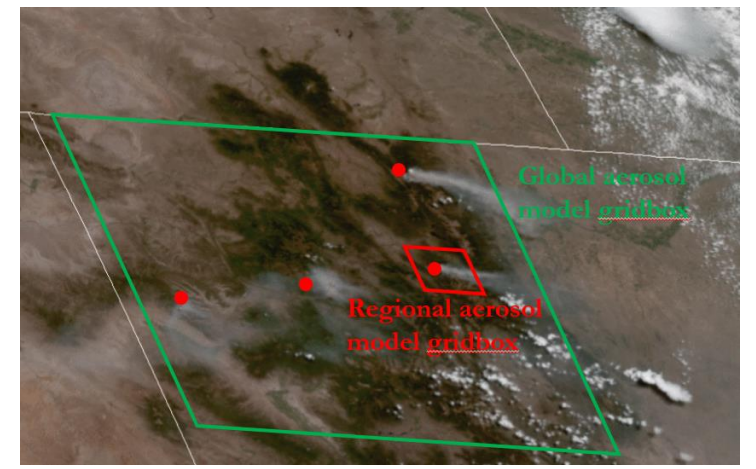


What I'll cover today:

- Plume injection height (PIH)
 - How much is at the surface?
 - Does this affect aerosol lifetime?
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Maegan DeLessio talk on Friday

Manabu Shiraiwa talk later today and
Havala Pye poster



Simulating Plume Injection Height

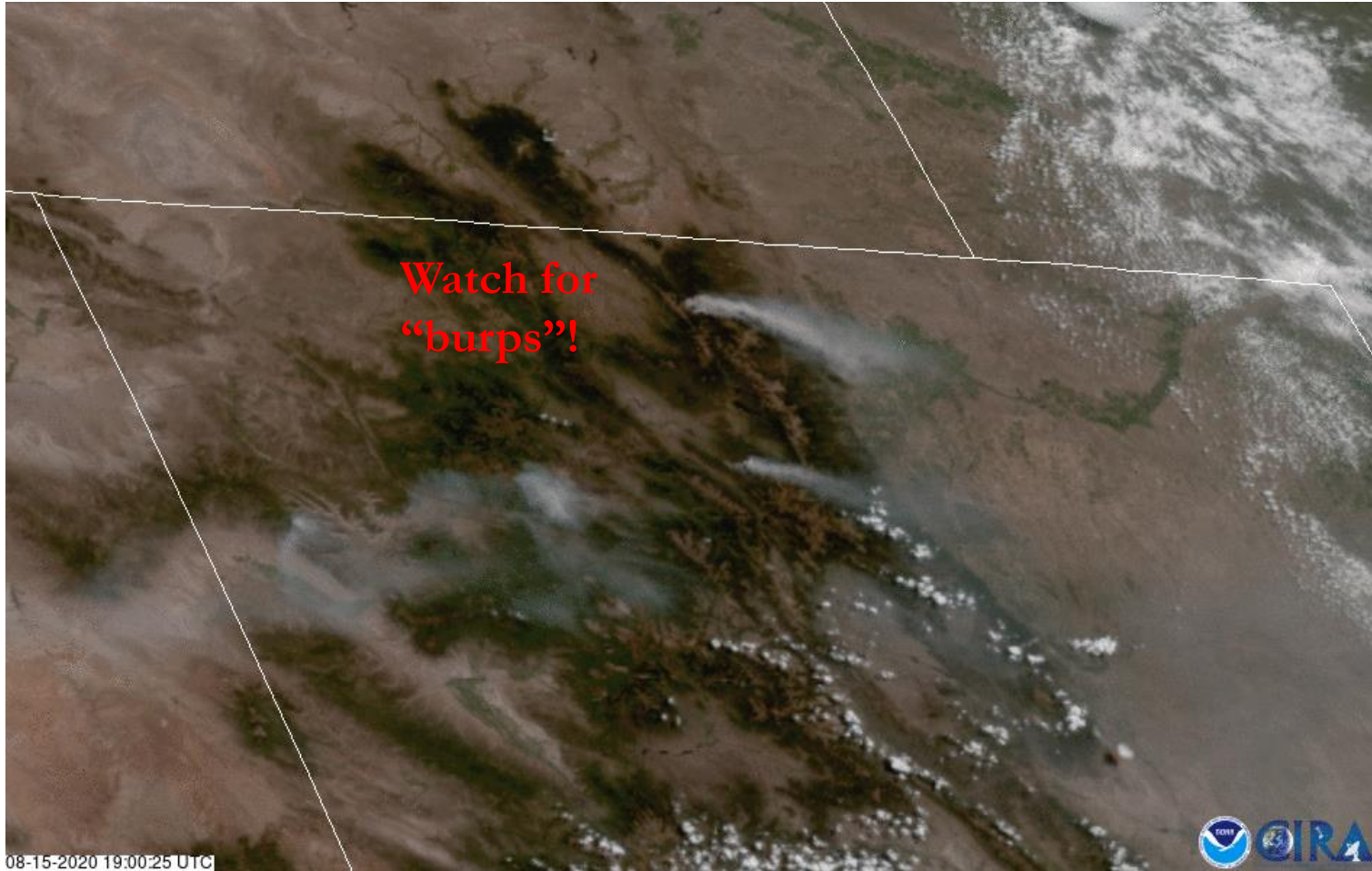
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Also see Jun Wang's talk on Friday

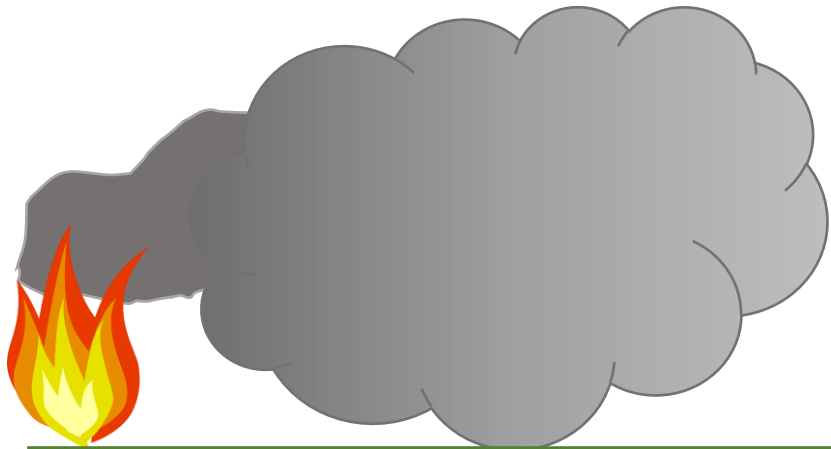


Plume injection height is challenging to predict.

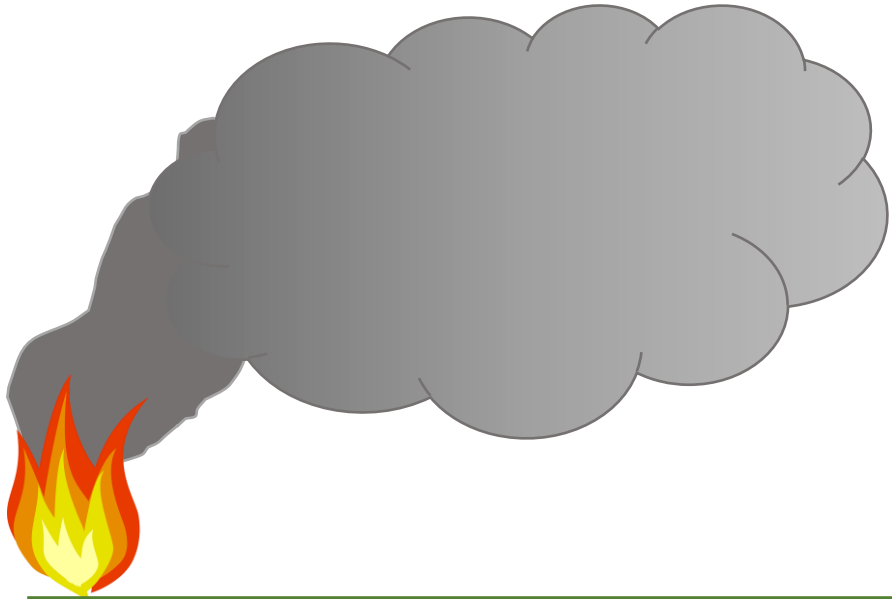


Colorado, Aug 15, 2020.

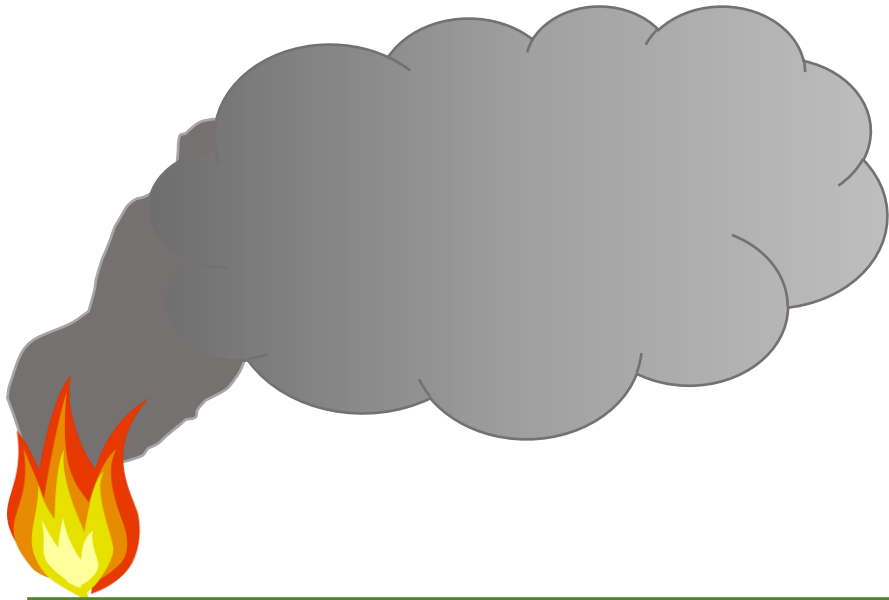
Compare smoke emitted near surface
to smoke injected into free troposphere.



Compare smoke emitted near surface
to smoke injected into free troposphere.

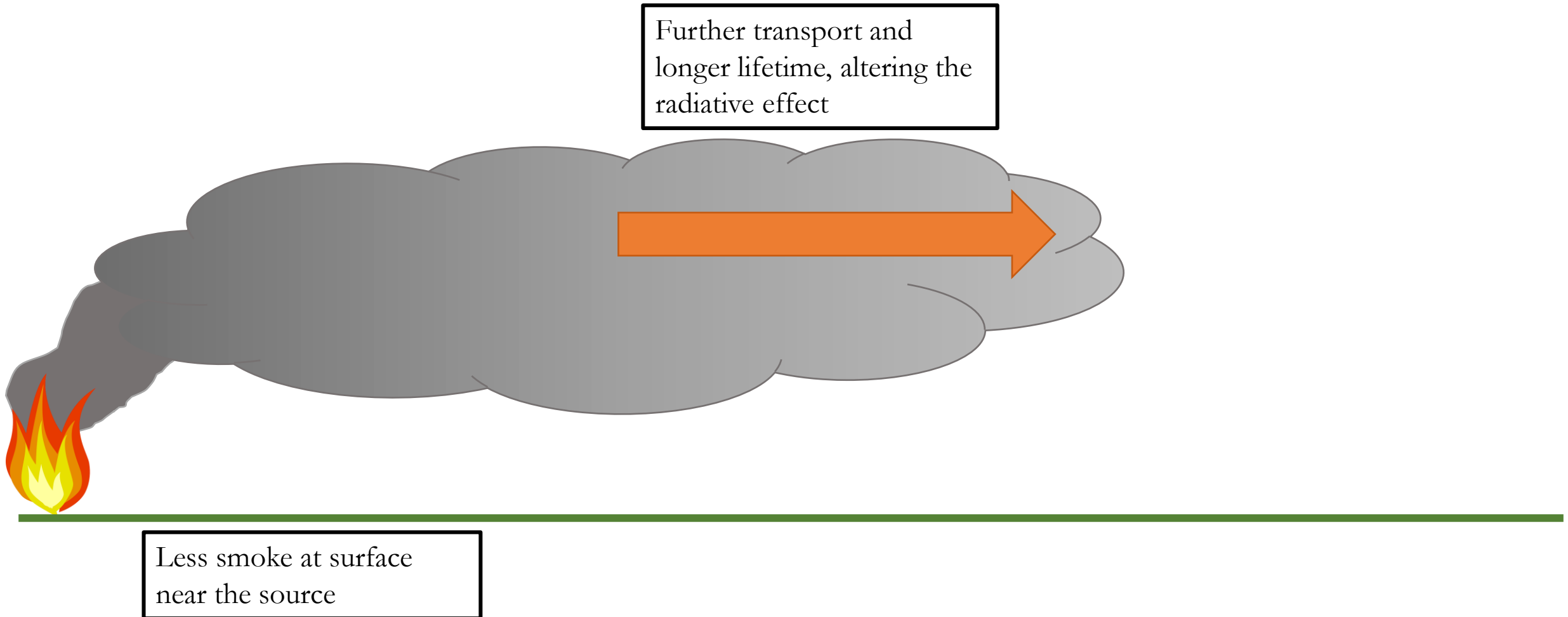


The plume injection height of biomass burning aerosols may have impacts on climate and air quality.

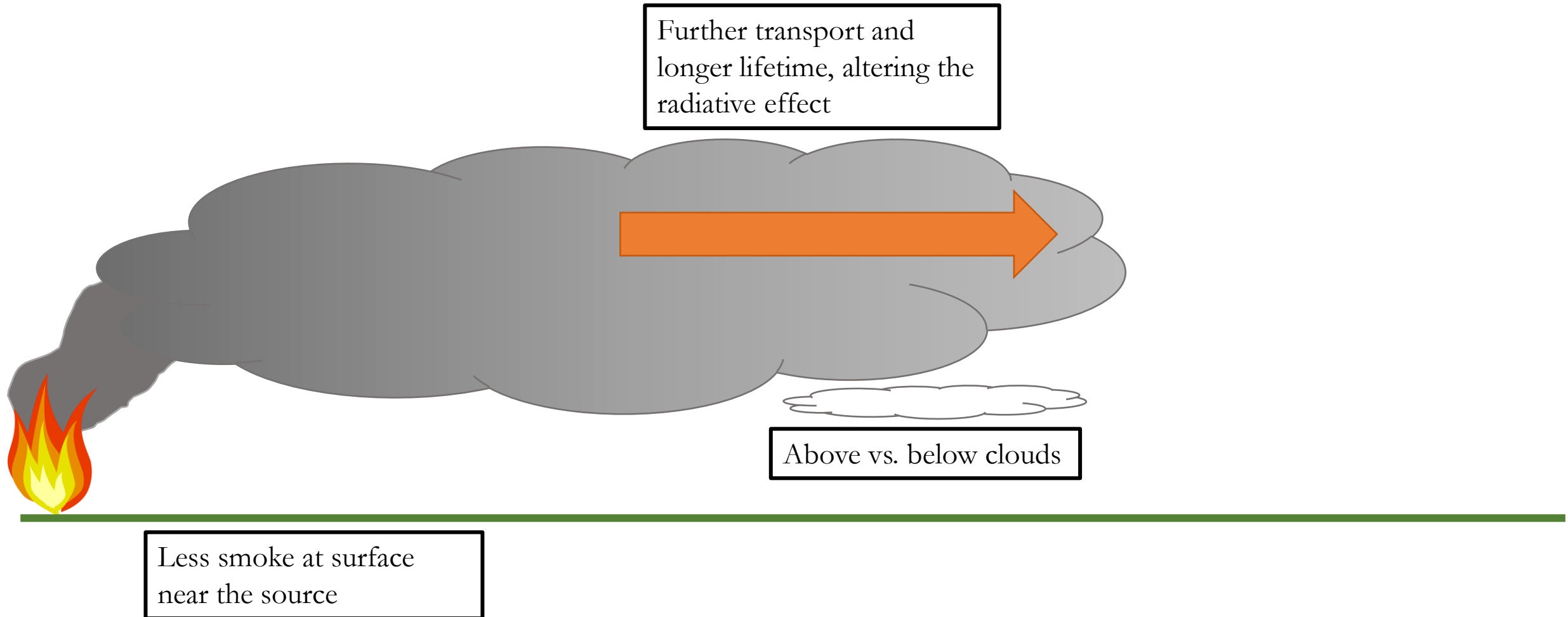


Less smoke at surface
near the source

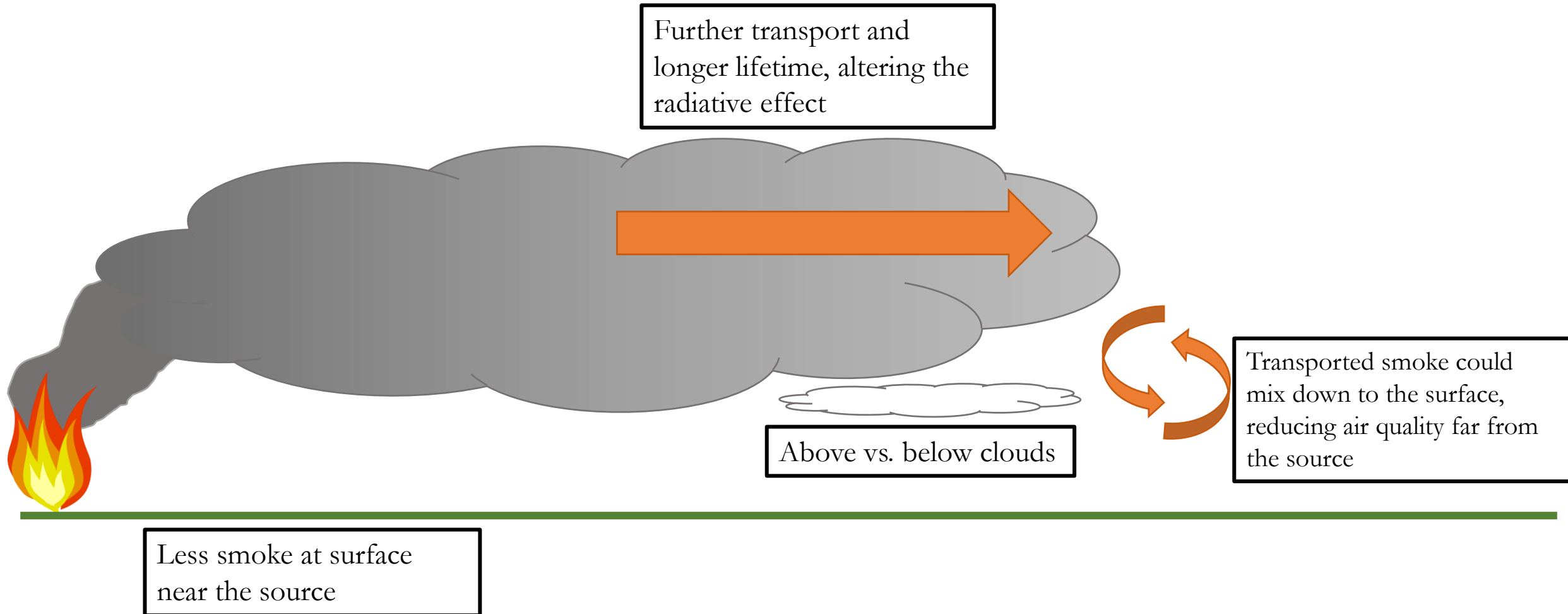
The plume injection height of biomass burning aerosols may have impacts on climate and air quality.



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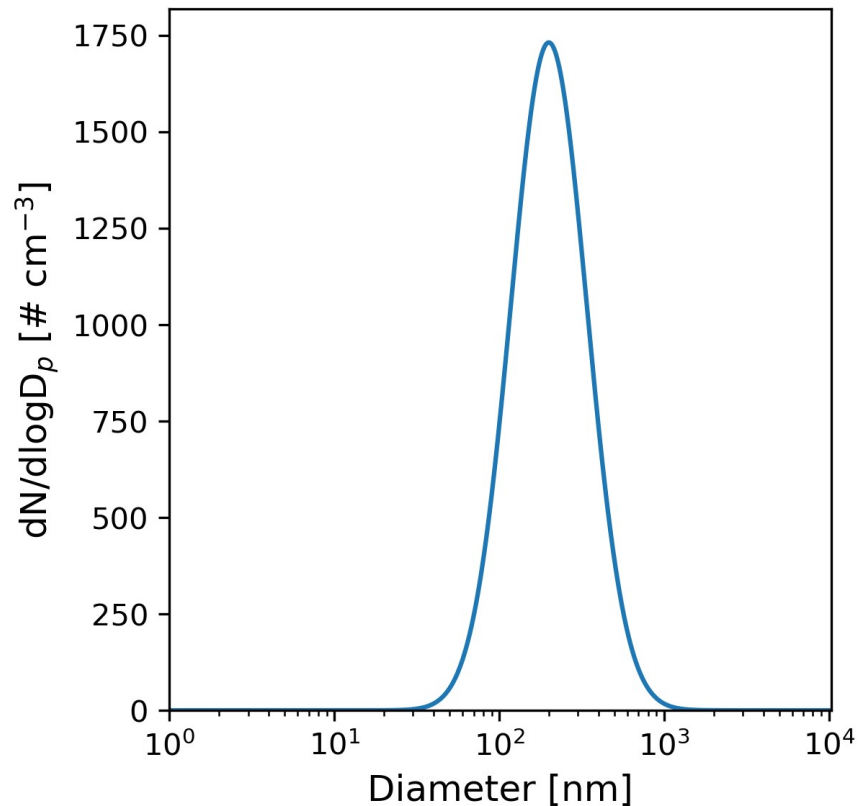


The plume injection height of biomass burning aerosols may have impacts on climate and air quality.



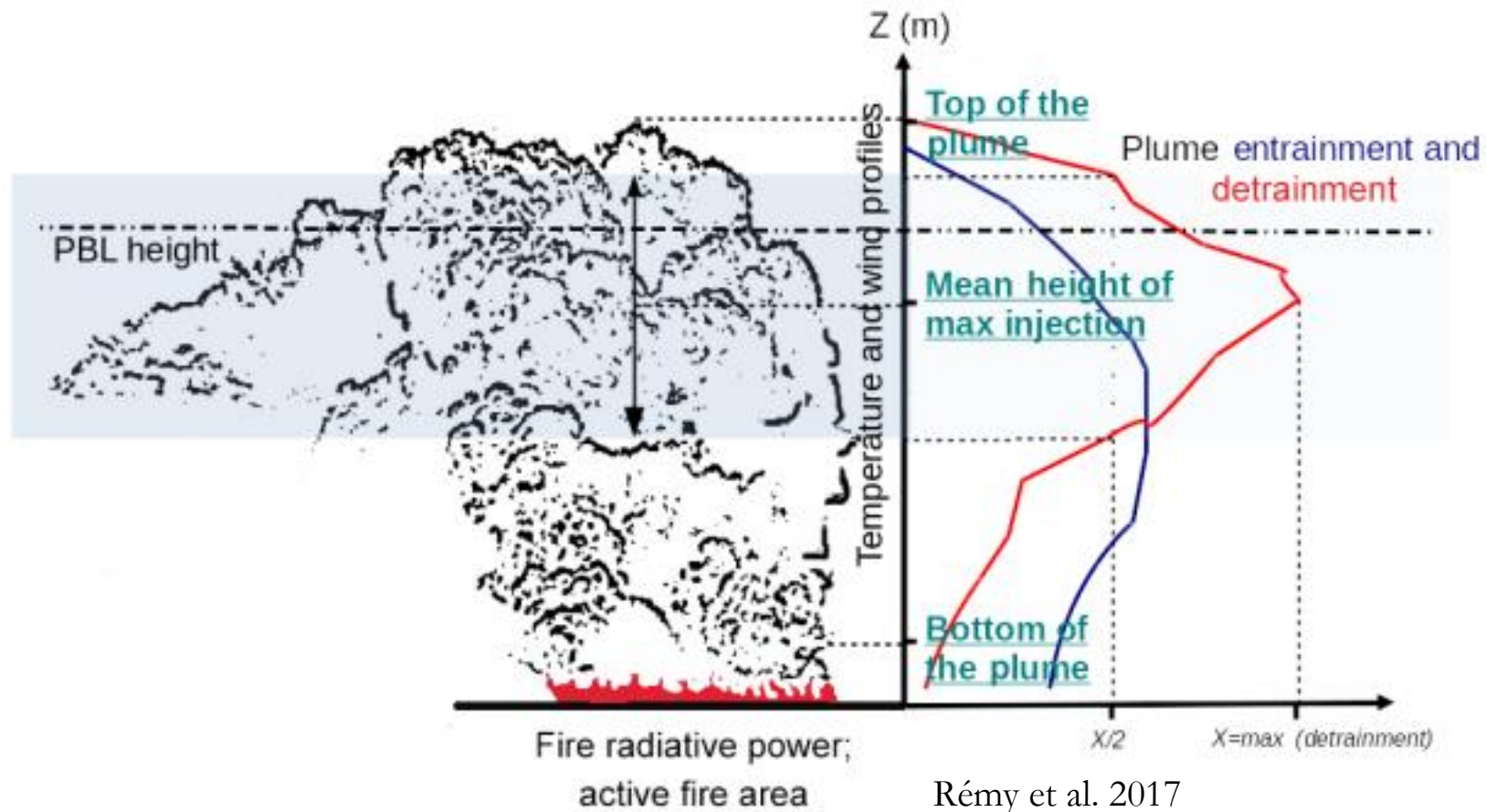
We ran GEOS-Chem-TOMAS for 2019 and 2020.

- $4^\circ \times 5^\circ$ horizontal resolution
- 47 vertical layers
- 15 size bins (3 nm to $10 \mu\text{m}$)



Nicole June

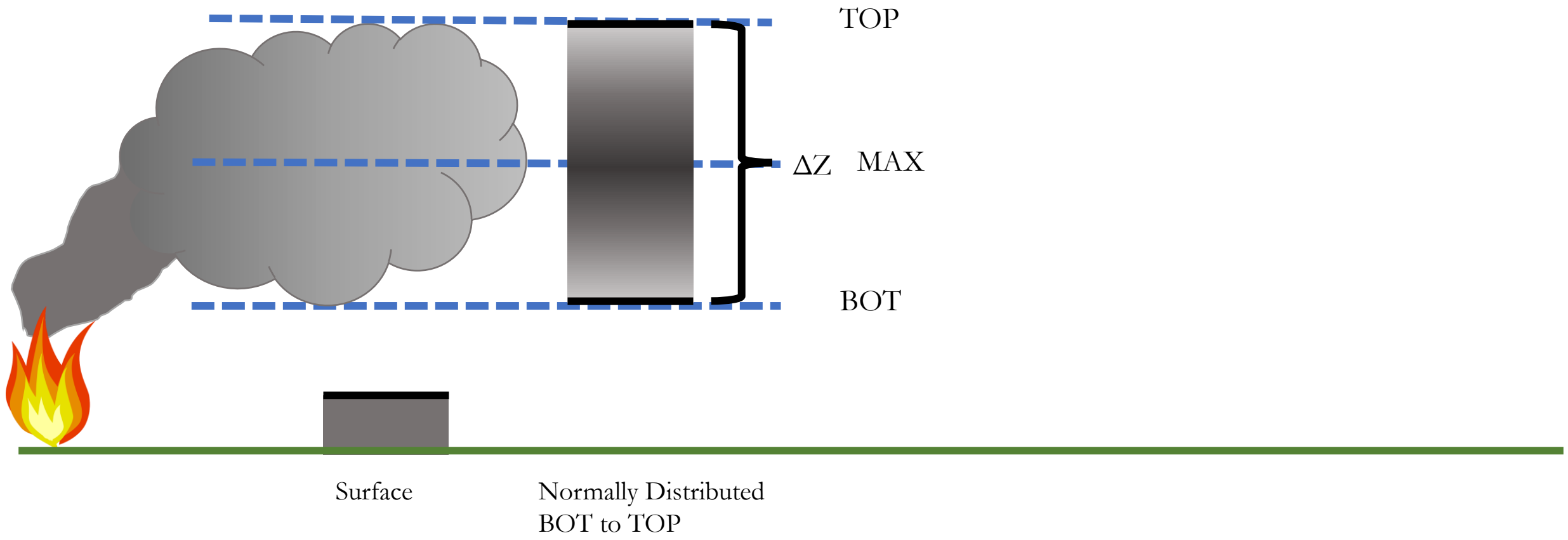
Biomass burning emissions and plume heights are provided in the GFAS emissions inventory.



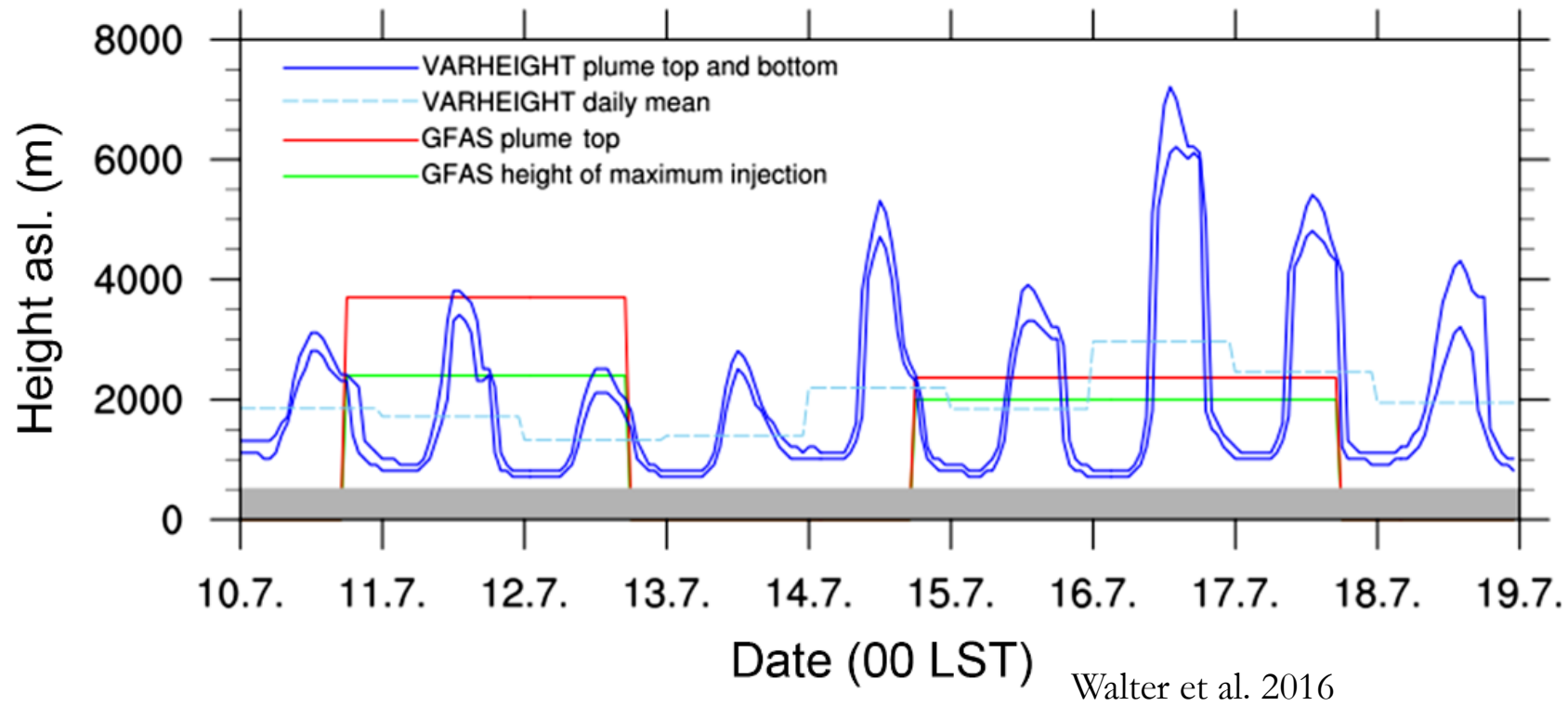
In the base case GFAS BB emissions are put into the surface layer.



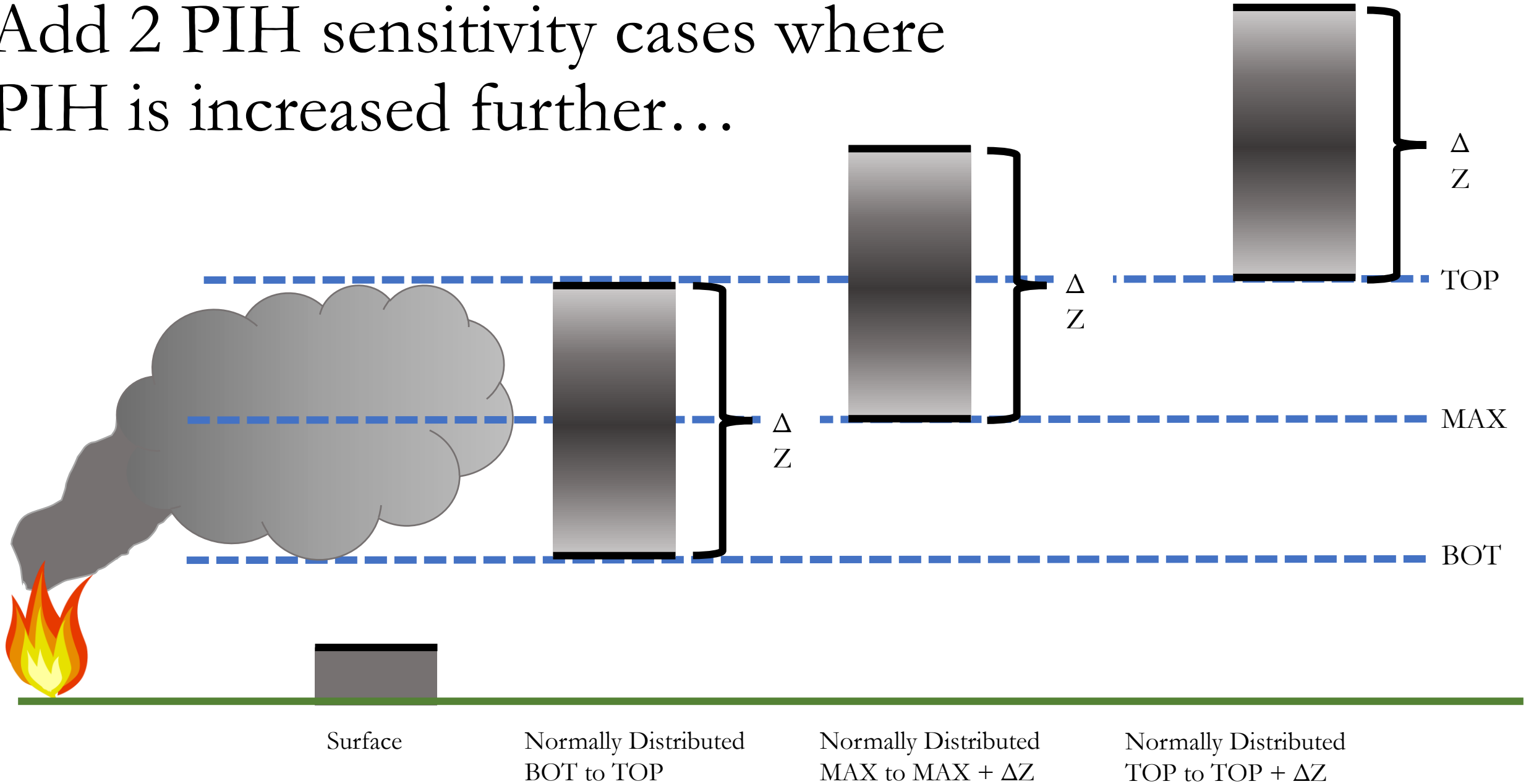
Use GFAS-provided plume injection heights.



Some evidence that GFAS plume heights may be biased low.

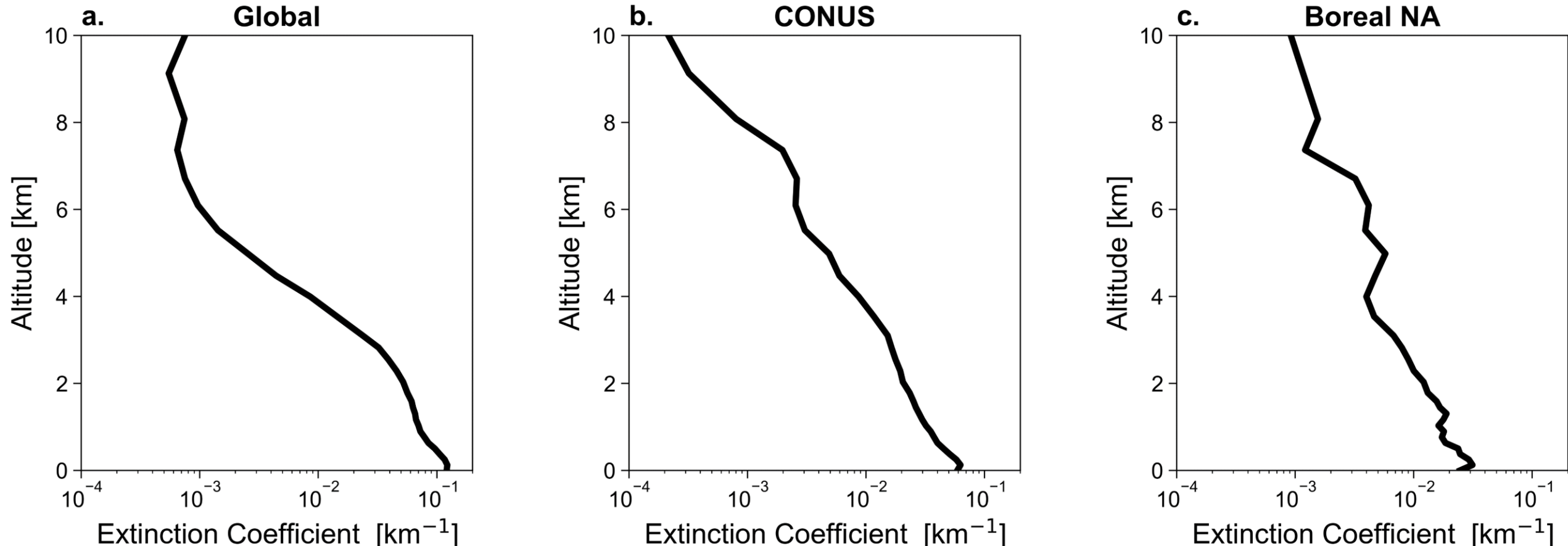


Add 2 PIH sensitivity cases where PIH is increased further...

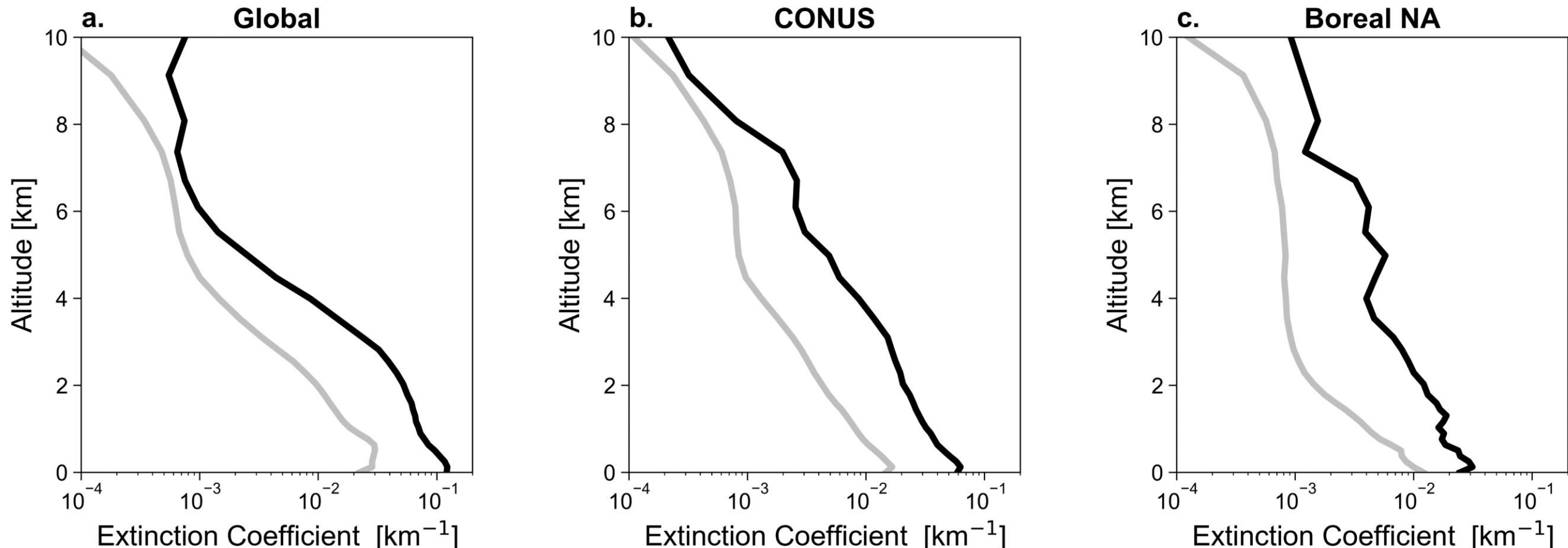


Evaluate simulated extinction coefficient vertical profiles against Calipso lidar observations.

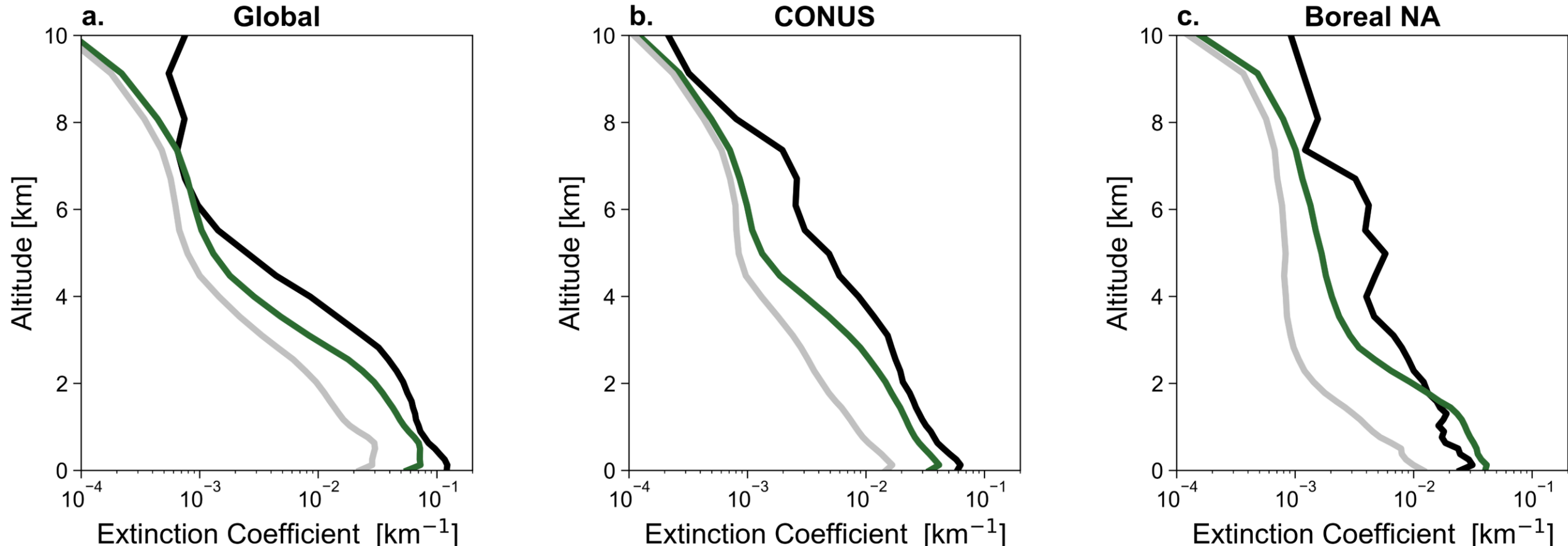
(Limited to time periods impacted by biomass burning smoke.)



Simulation with no biomass burning underestimates extinction at all altitudes.



Adding biomass burning emissions at surface underestimates extinction in free troposphere.

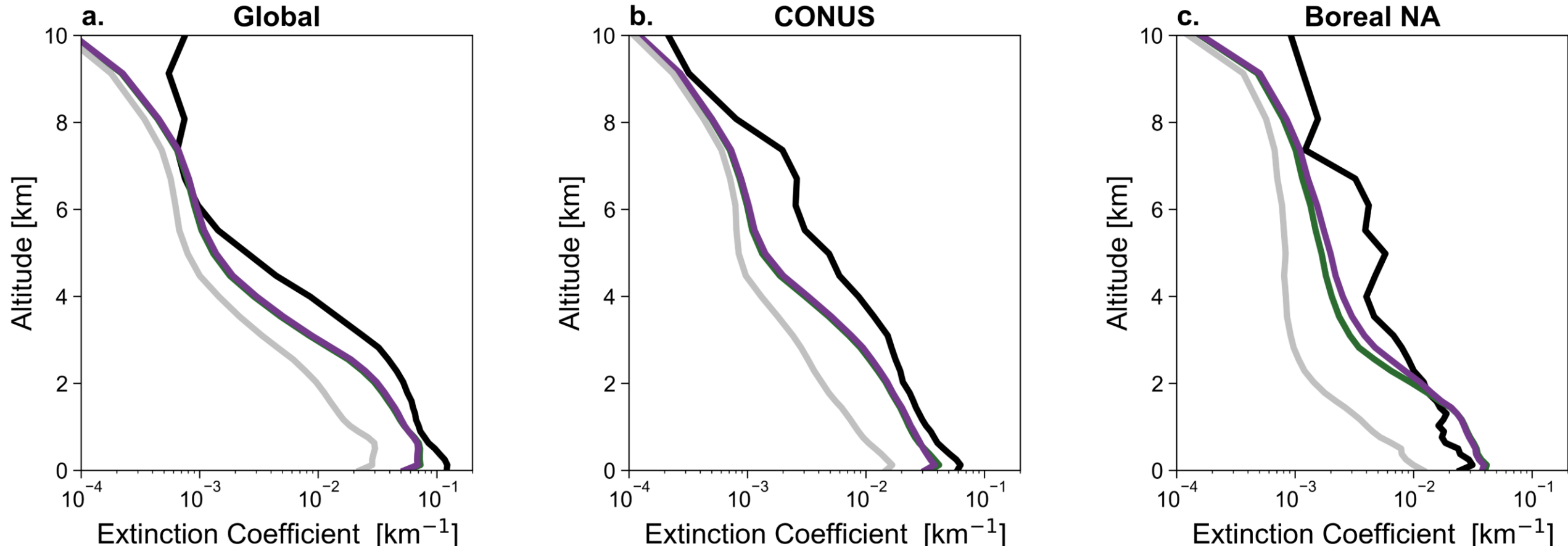


Using GFAS injection heights improves free tropospheric extinction in Boreal North America.

Increasing Plume Injection Height

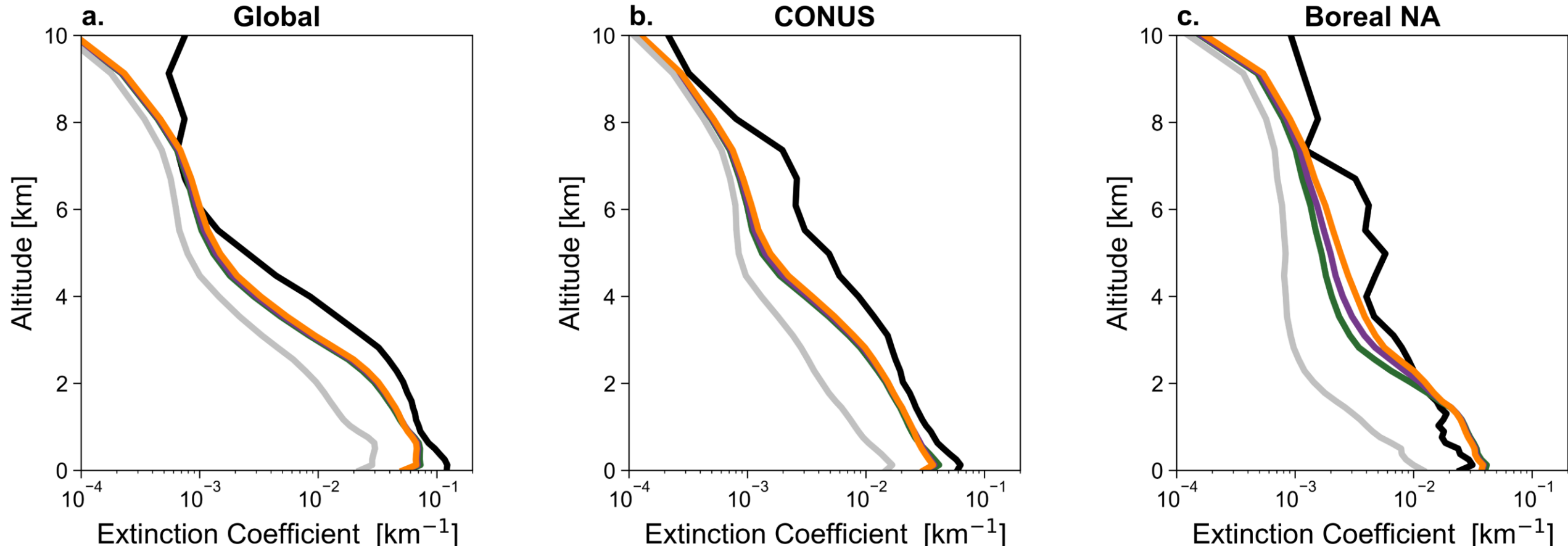
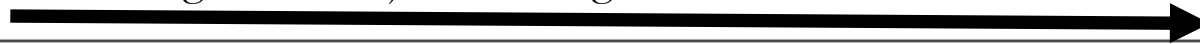


— Calipso lidar — No BB — Surface — BOTtoTOP



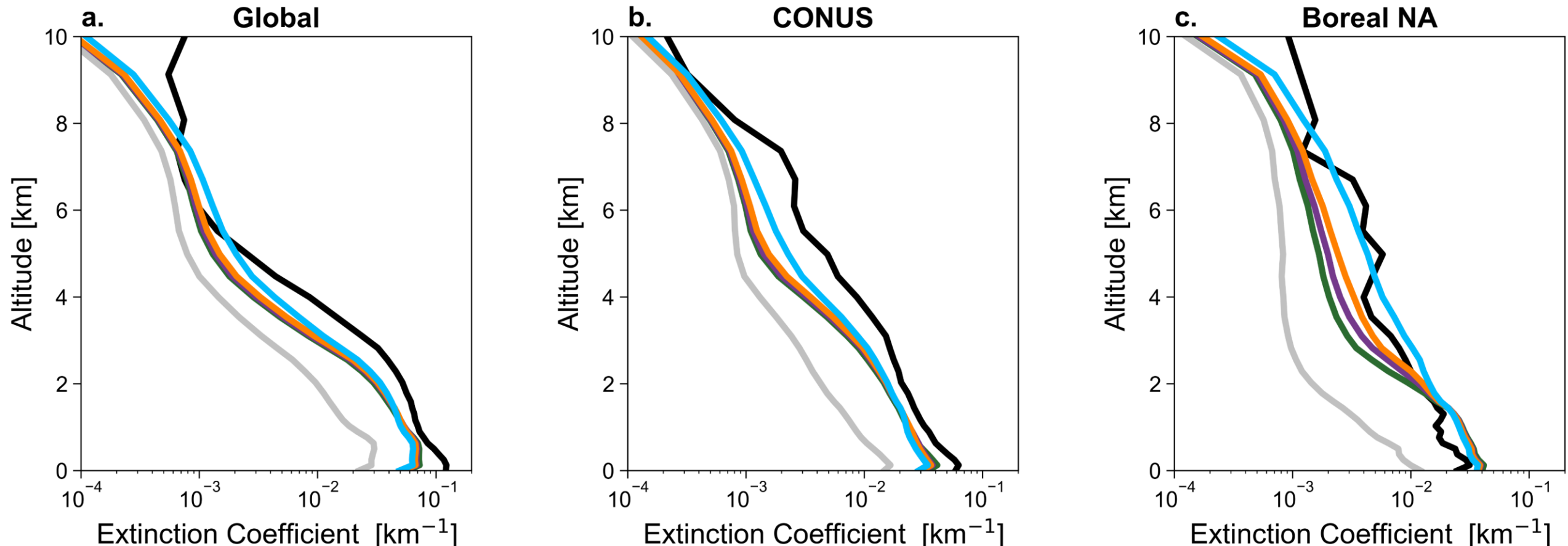
Increasing GFAS injection heights leads to further improvements.

Increasing Plume Injection Height

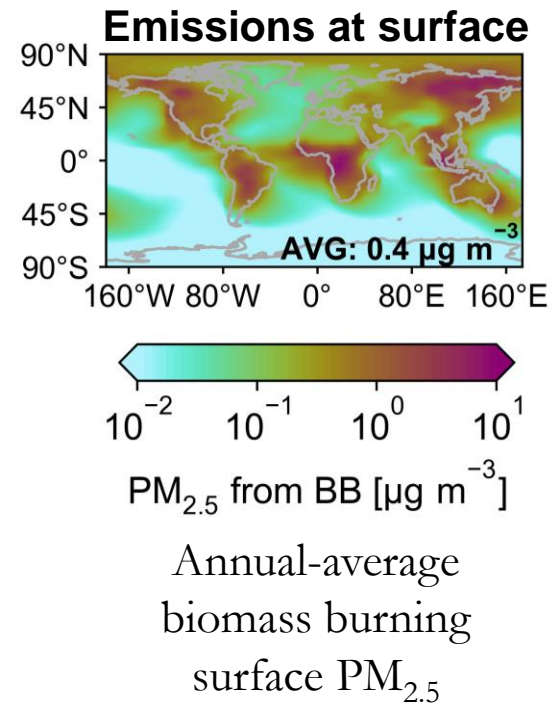


Increasing GFAS injection height more leads to general improvement in all regions.

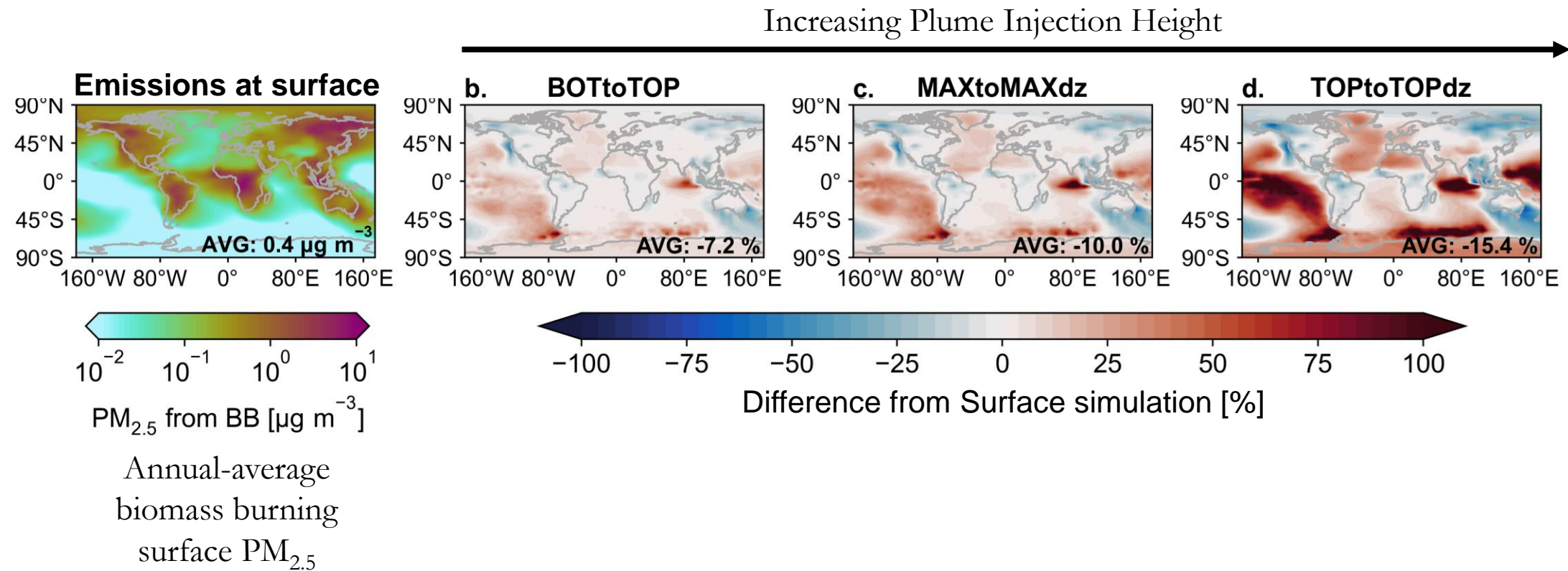
Increasing Plume Injection Height



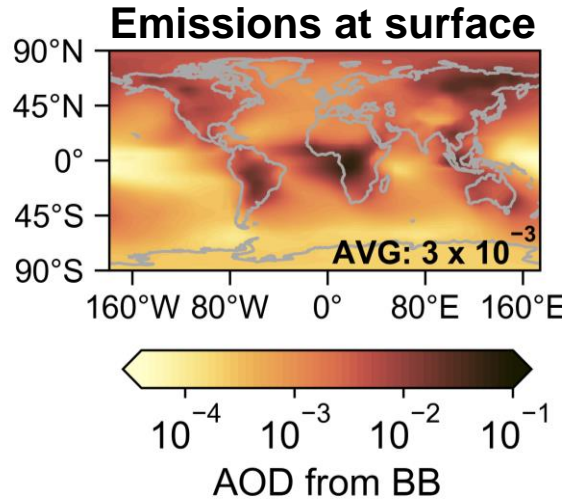
Surface PM_{2.5} from biomass burning in base case (surface emissions).



Increasing PIH decreases surface $\text{PM}_{2.5}$ in source regions and increases in marine areas.

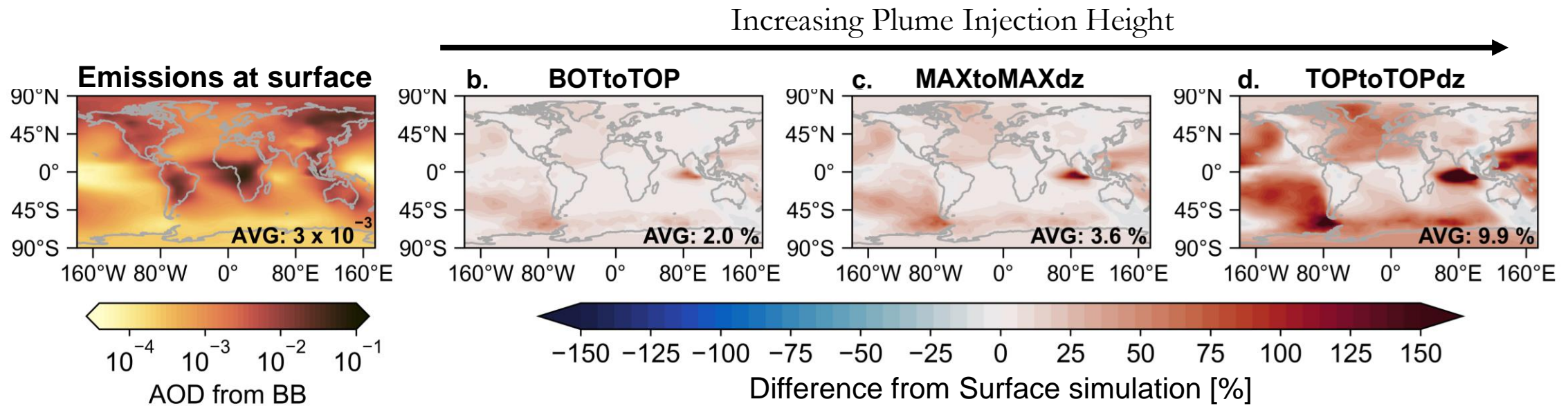


Aerosol optical depth (AOD) from biomass burning in base case (surface emissions).



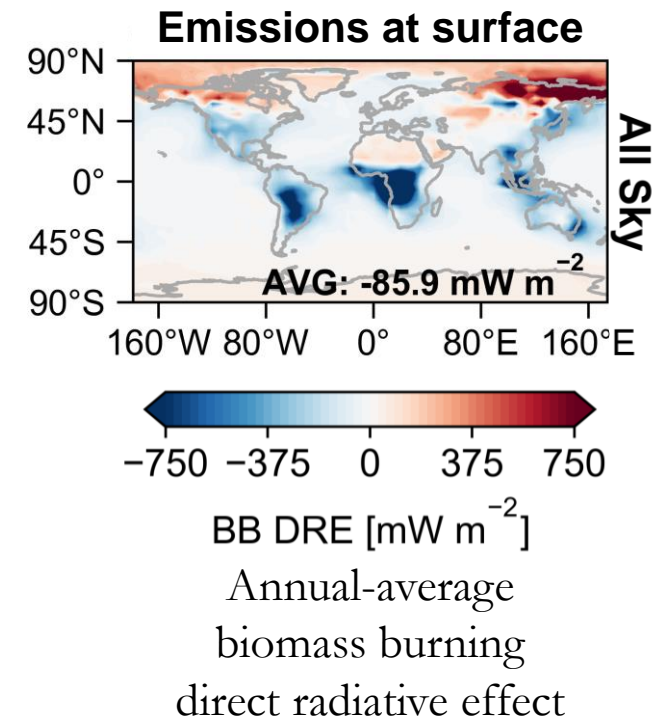
Annual-average
biomass burning AOD

Increasing PIH increases aerosol lifetime,
so aerosol optical depth increases in most regions.

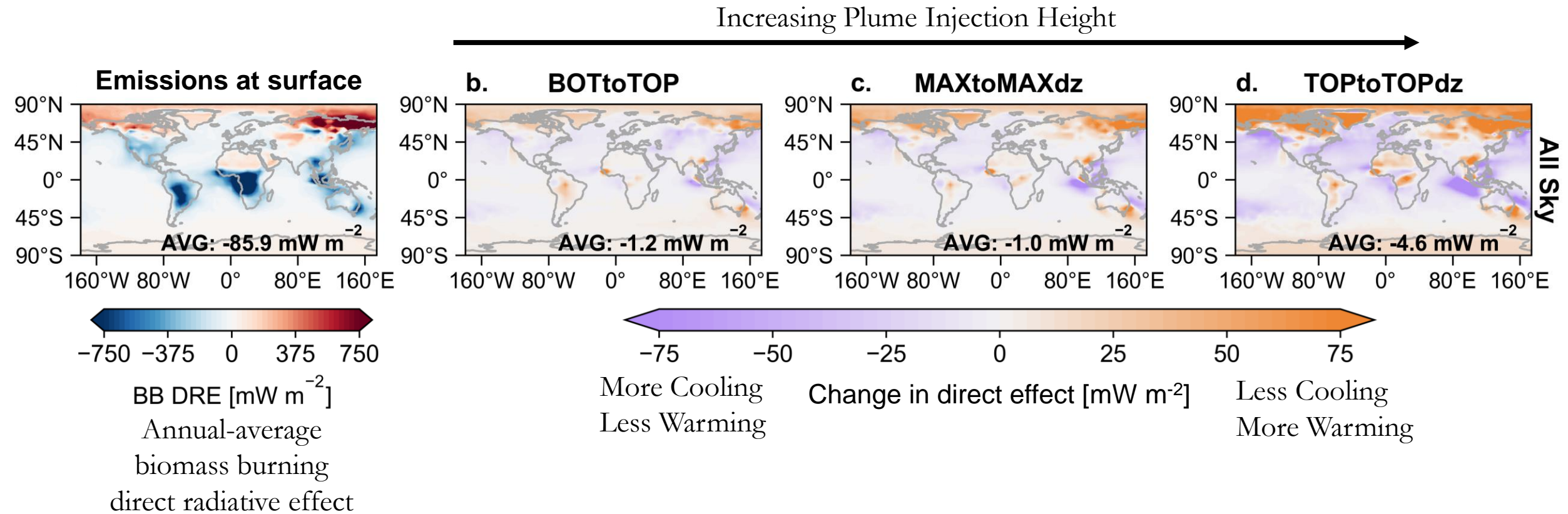


Annual-average
biomass burning AOD

Biomass burning has a cooling direct radiative effect.



Increasing PIH hardly changes biomass burning direct effect even though lifetime and AOD increase... why?

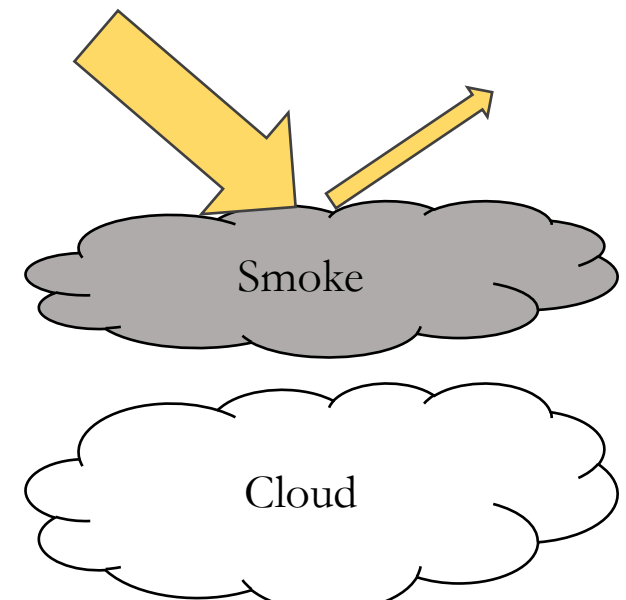
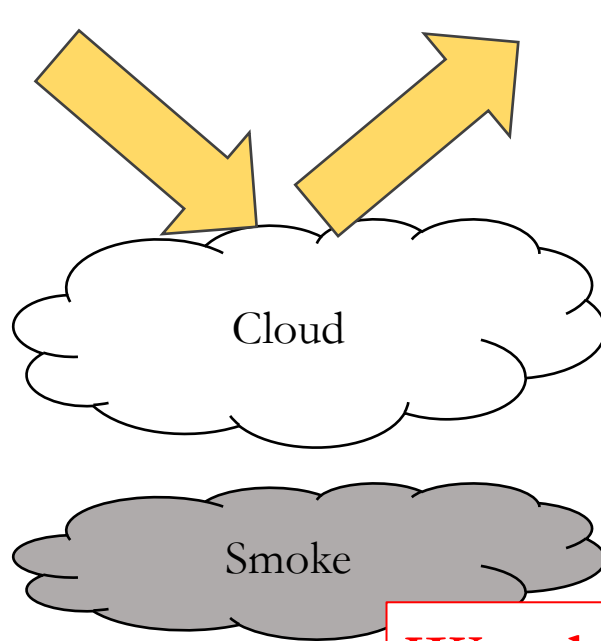
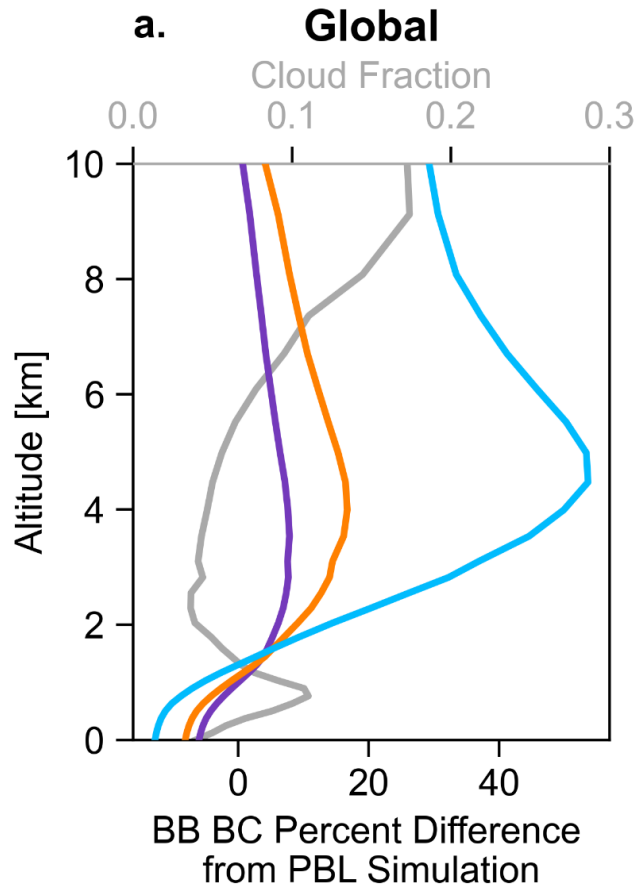
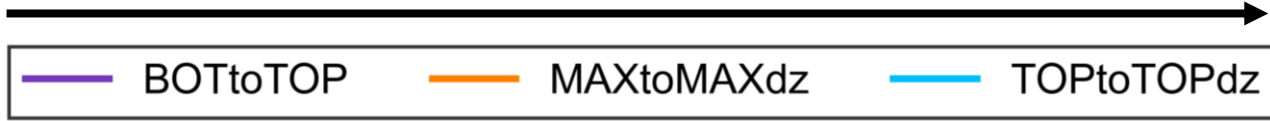


Smoke cools over dark surfaces, warms over clouds.



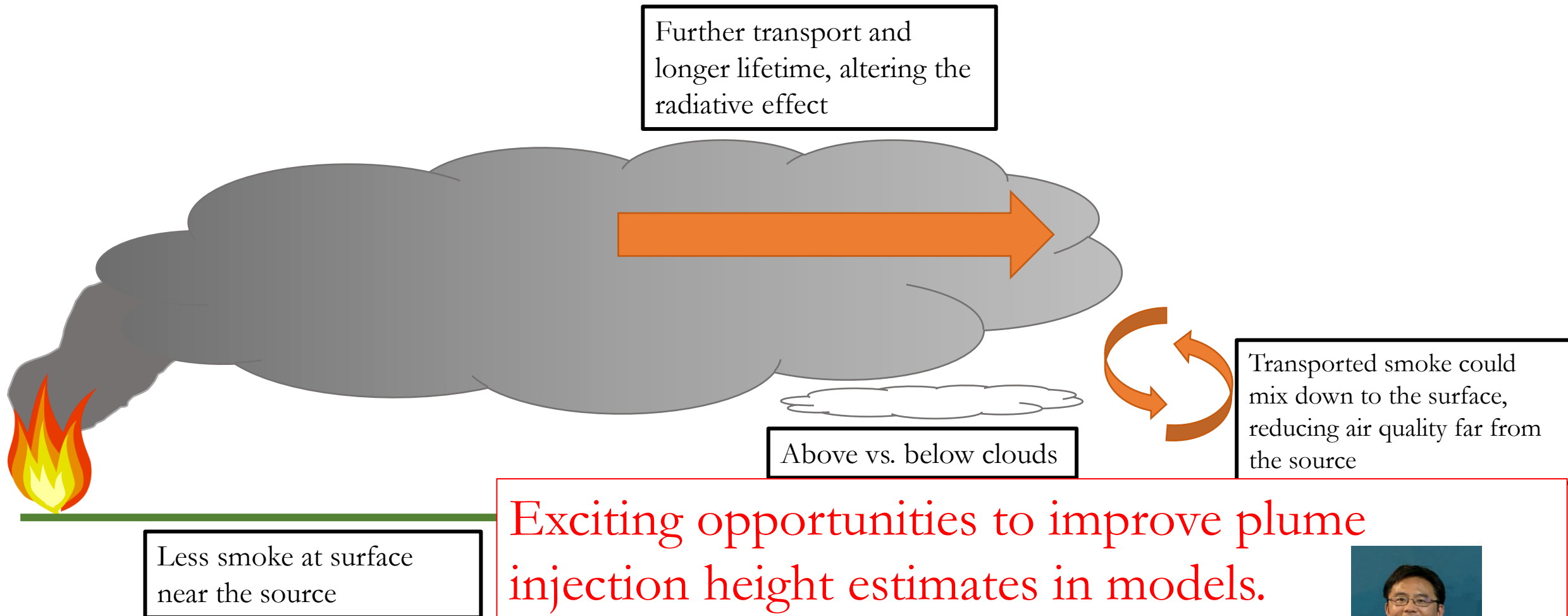
Some of the changes in the all-sky DRE are due to the impact of smoke above clouds.

Increasing Plume Injection Height →



We also assessed indirect effects...
skipping today for time!

Accurate plume injection heights are critical for estimating health and climate affects of biomass burning.



Exciting opportunities to improve plume injection height estimates in models.

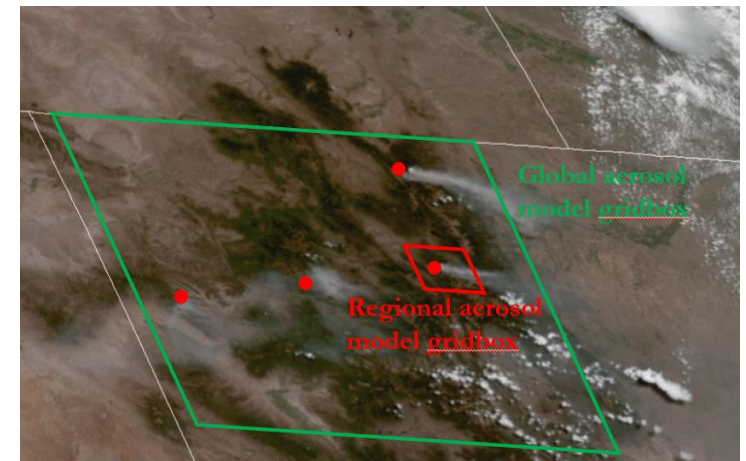
Jun Wang's talk on Friday!



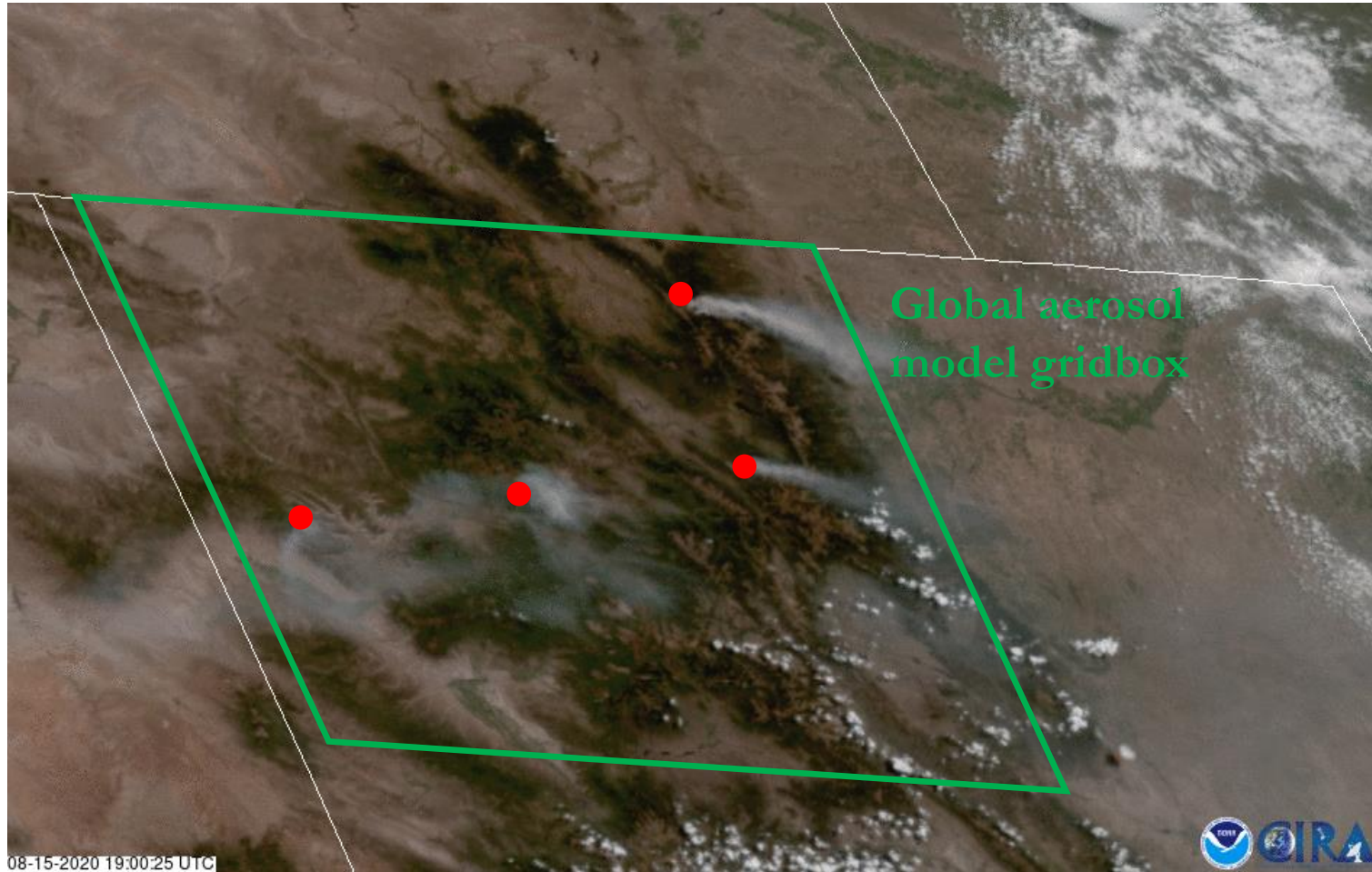
Non-linear sub-grid plume physics:

- Plume injection height (PIH)
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No time to discuss today, but lots of work with Shantanu Jathar and by others!



The sub-grid problem: Smoke is rarely well-mixed in a gridbox.

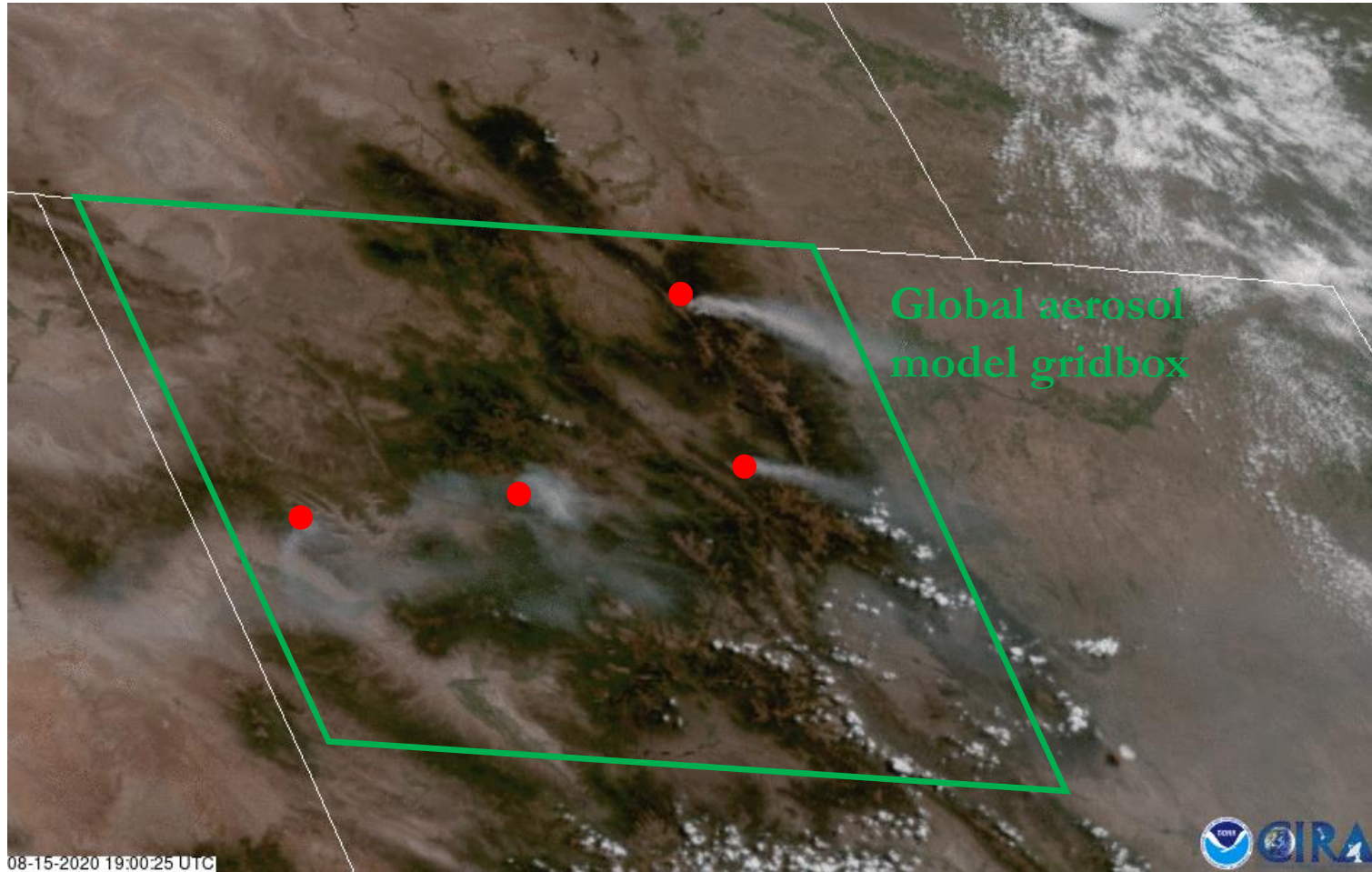


Colorado, Aug 15, 2020.

Two related issues:

1. Smoke mixes instantly into model gridboxes.
2. Real smoke plumes dilute at different rates.

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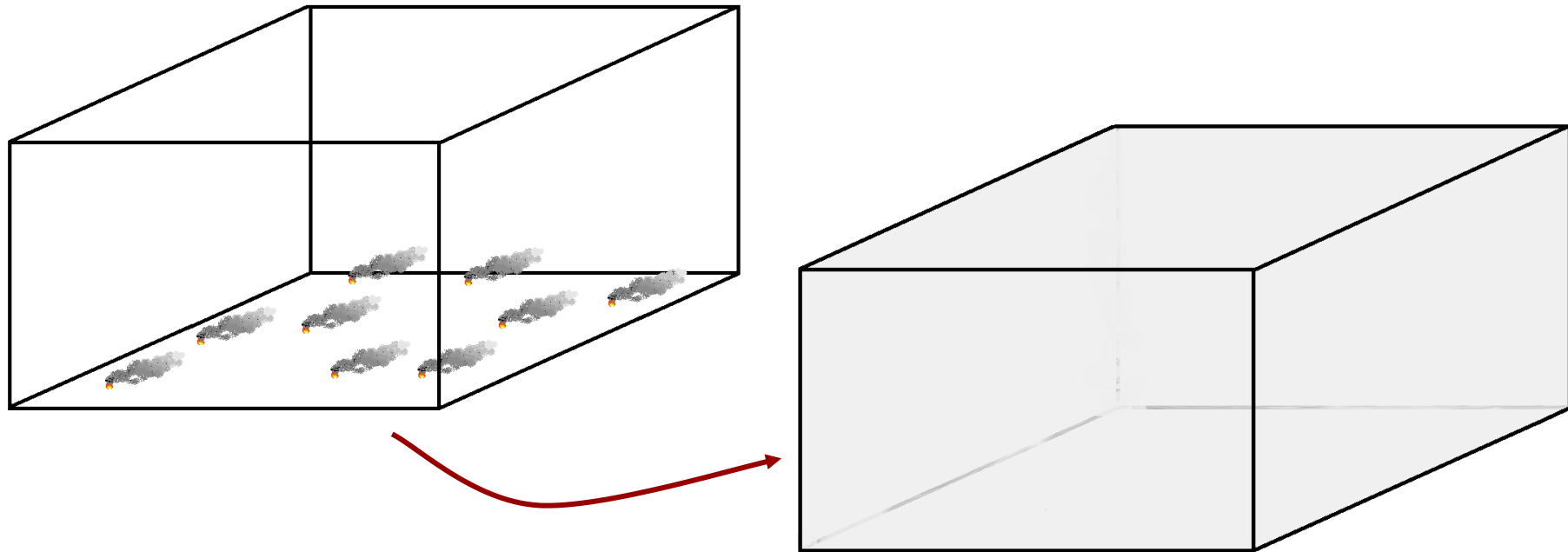


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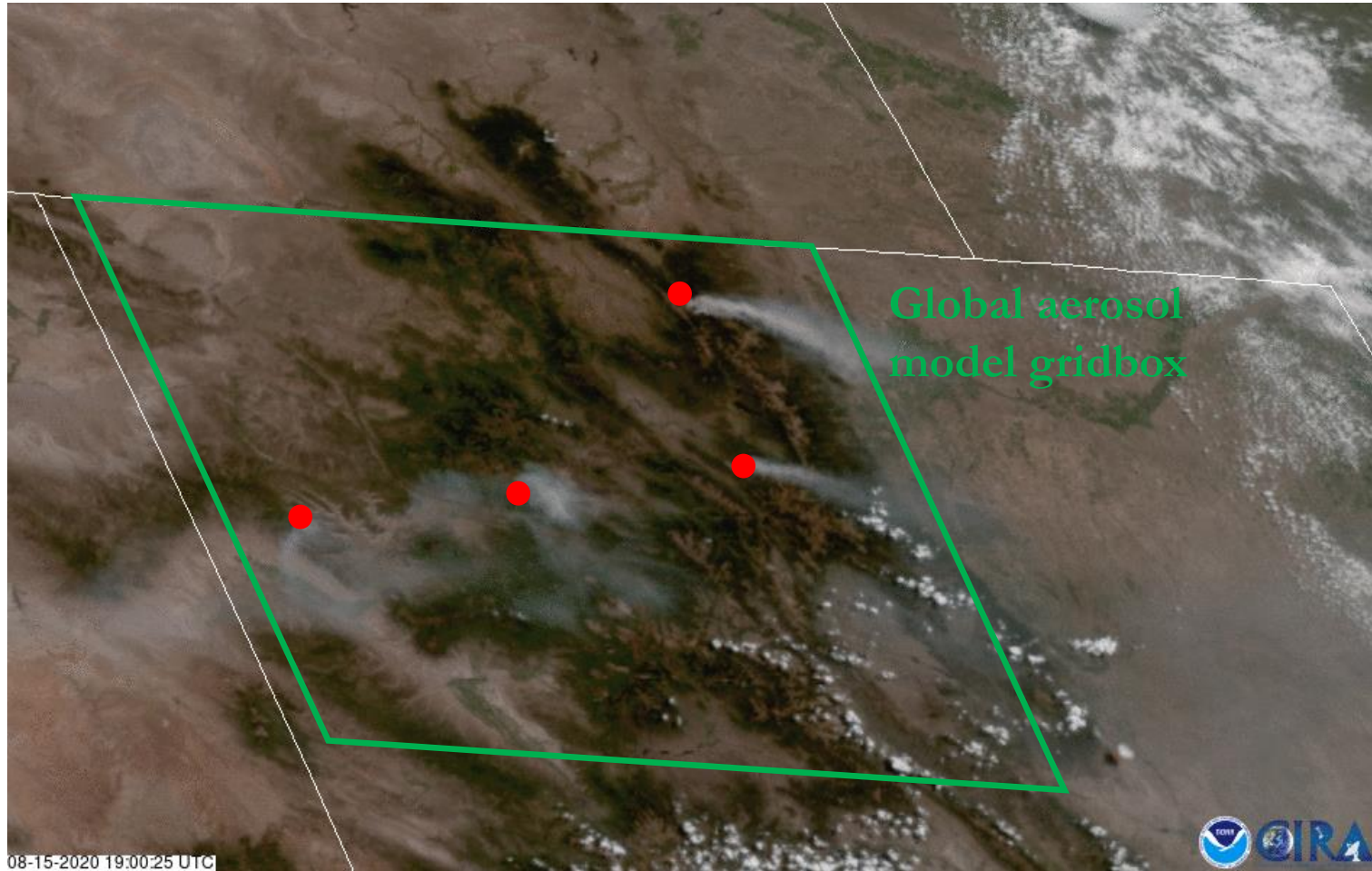
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Concentrated, sub-grid plumes instantly mix throughout the gridbox.



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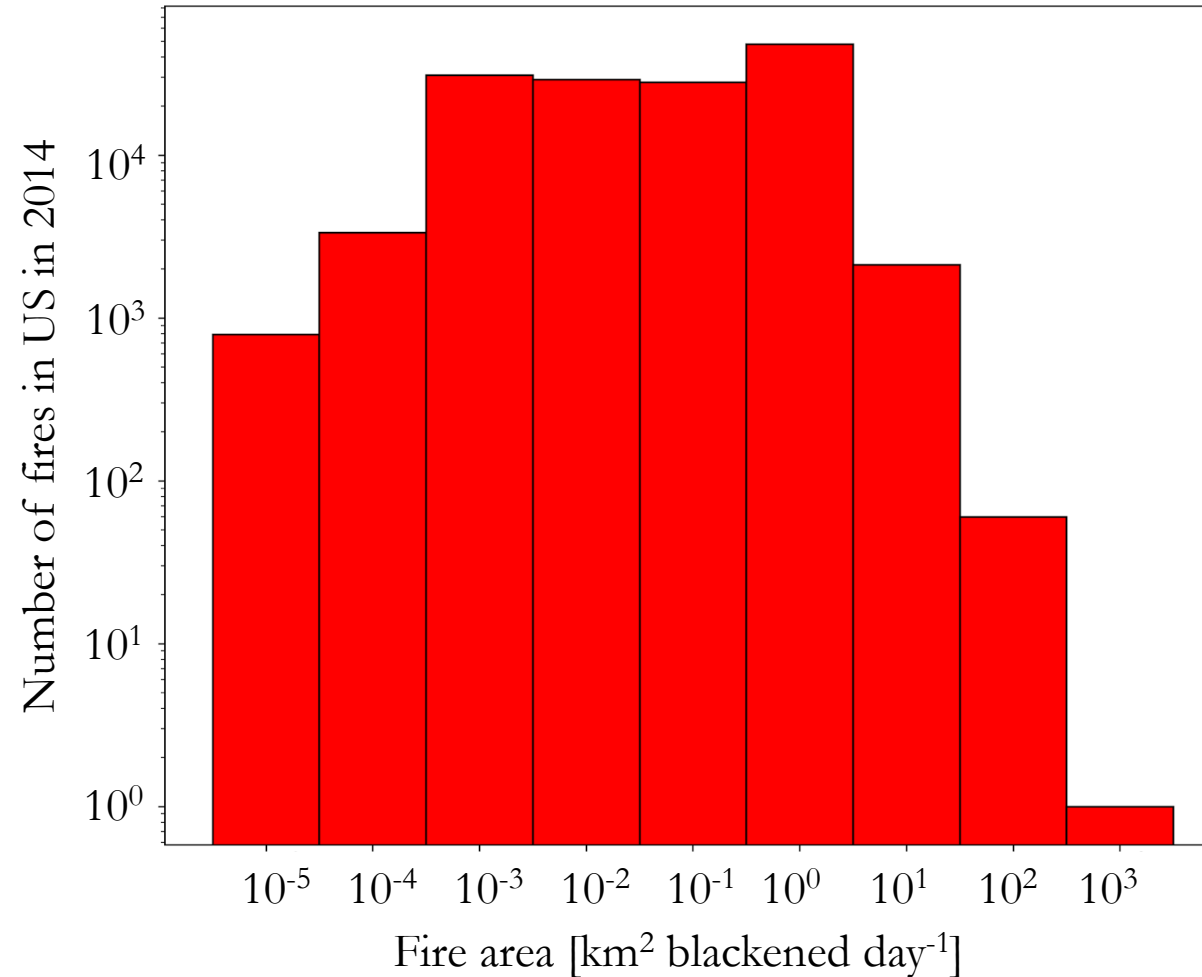


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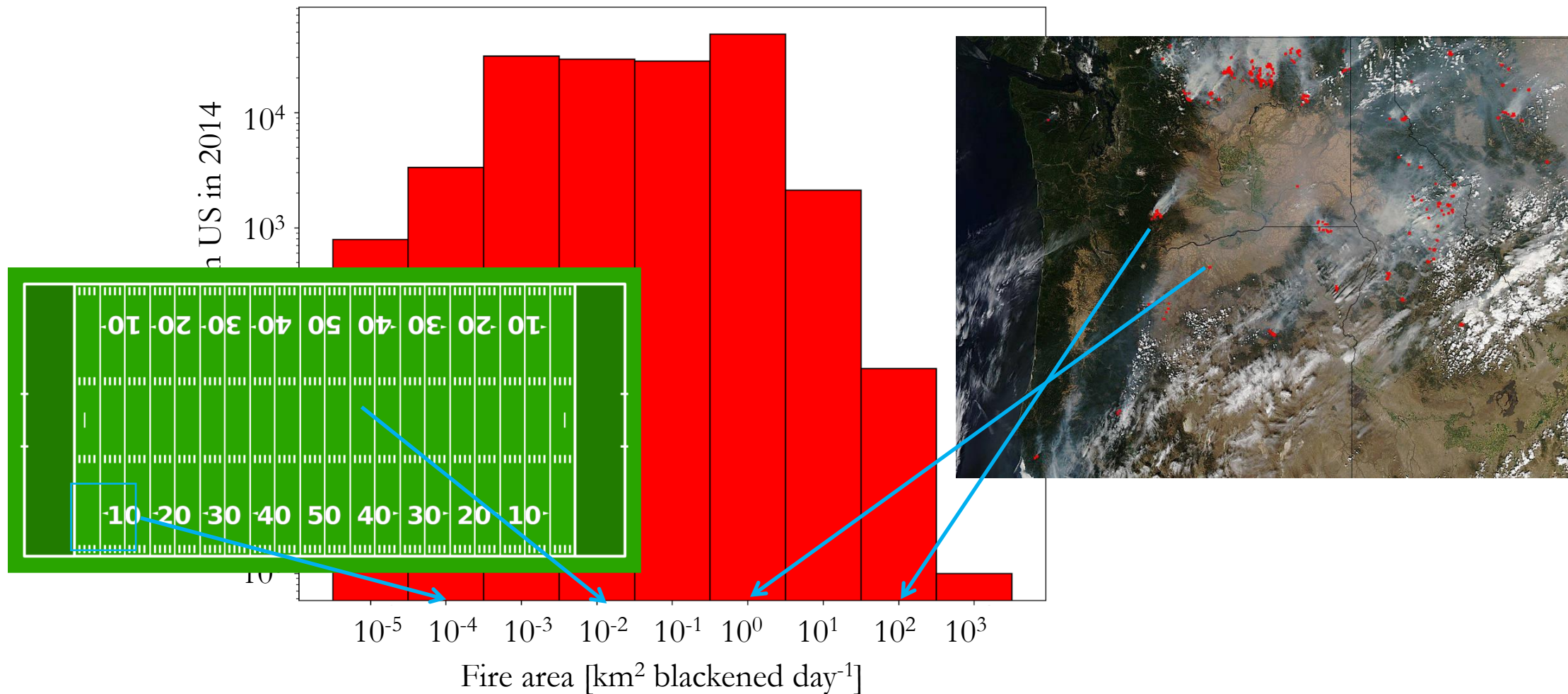
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HUGE range in fire sizes



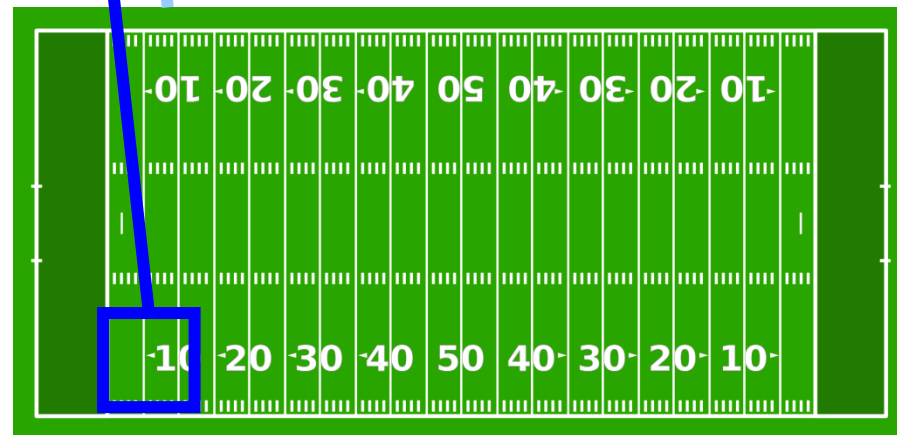
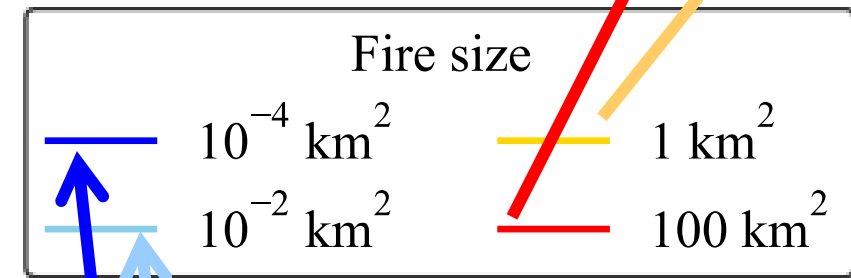
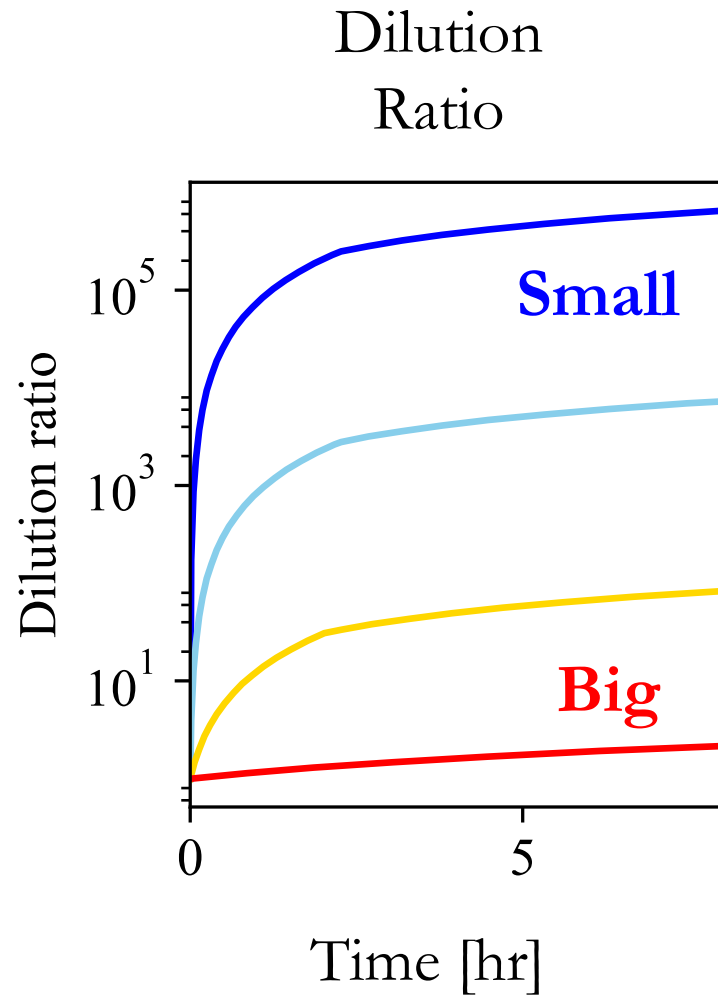
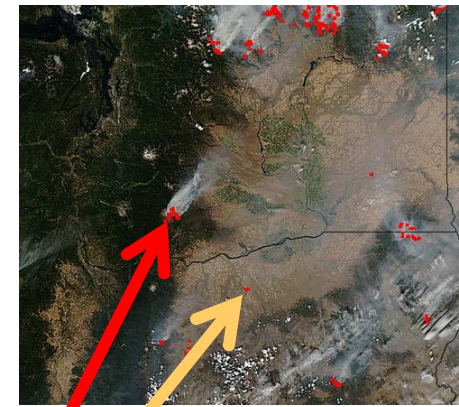
US Fires for 2014 from EPA NEI

HUGE range in fire sizes

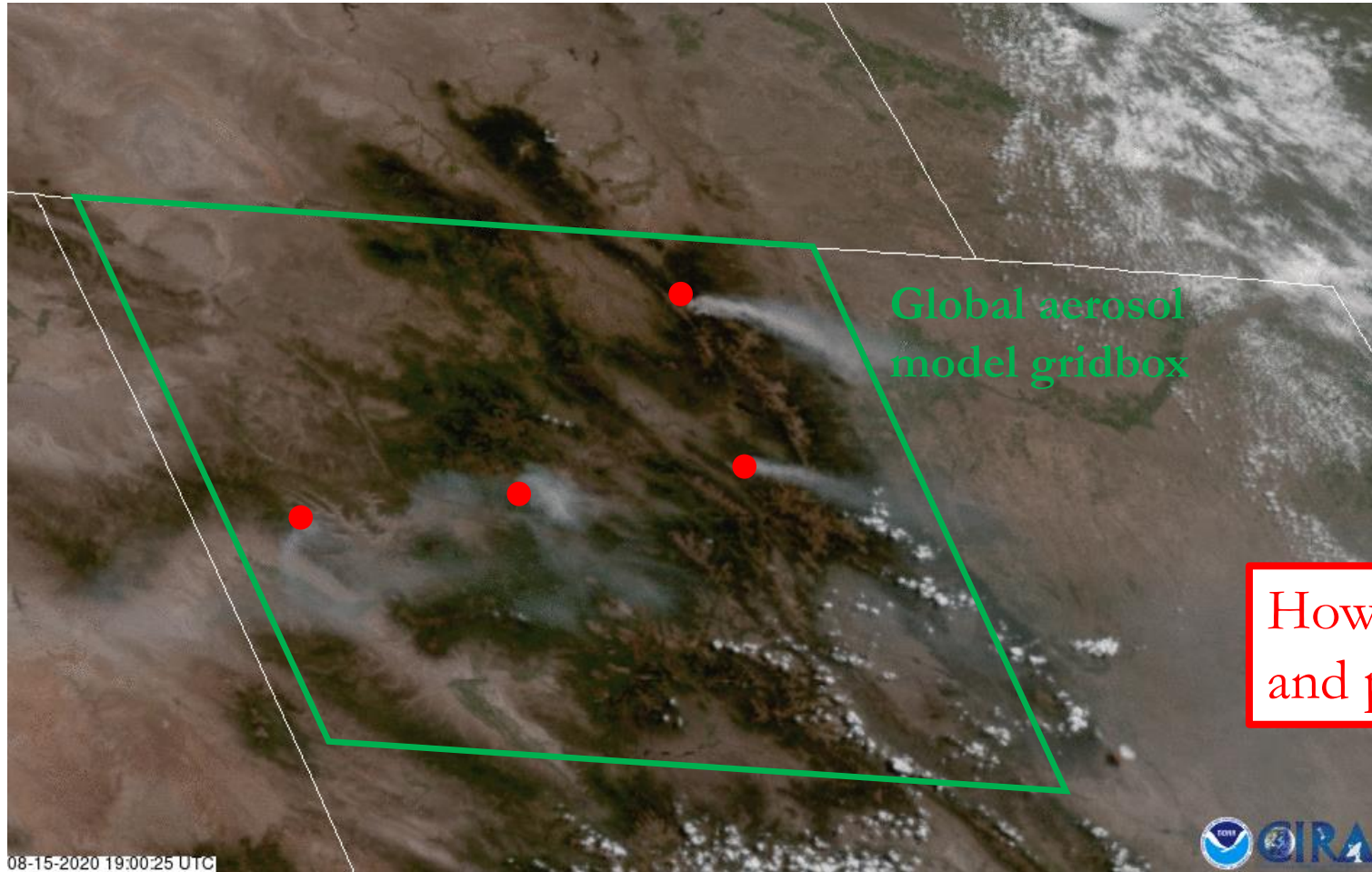


US Fires for 2014 from EPA NEI

Dilution rates span orders of magnitude with fire size.



The sub-grid problem: Smoke is rarely well-mixed in a gridbox.



Colorado, Aug 15, 2020.

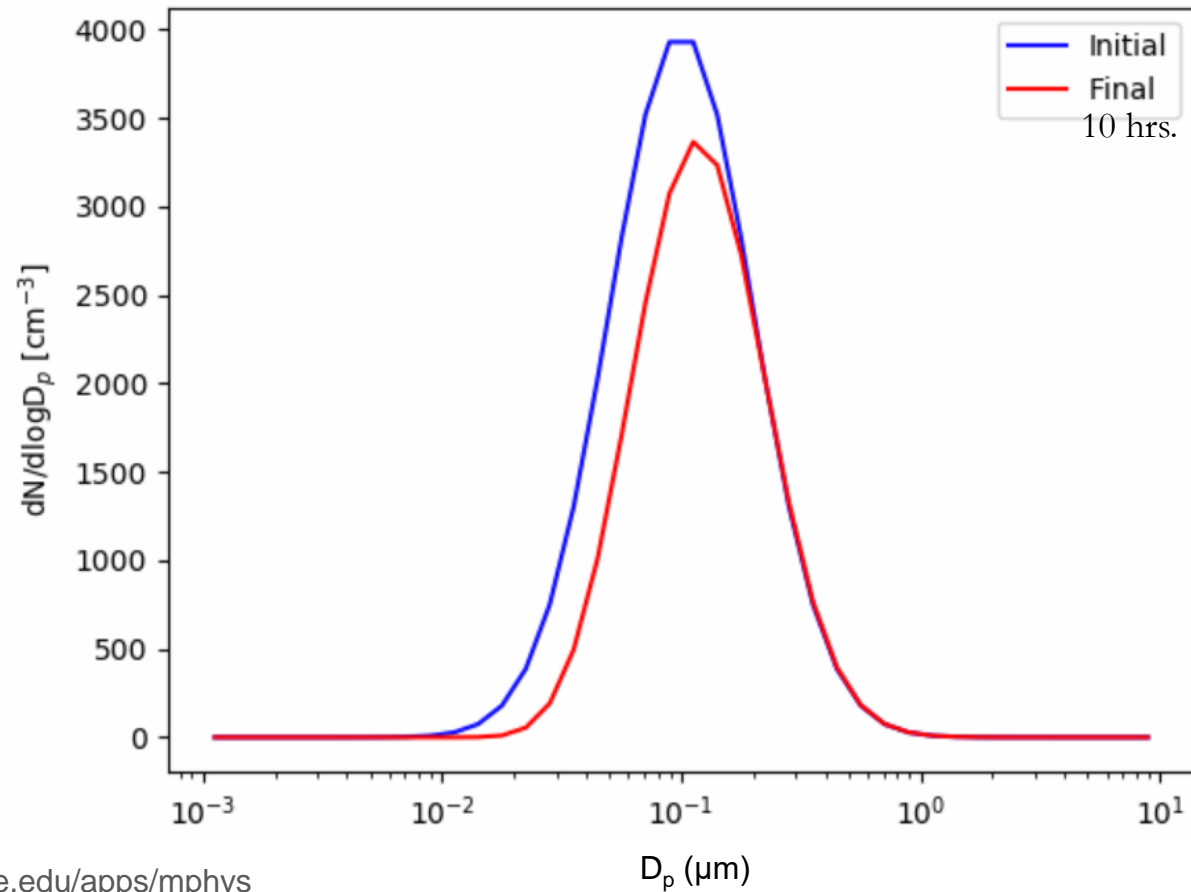
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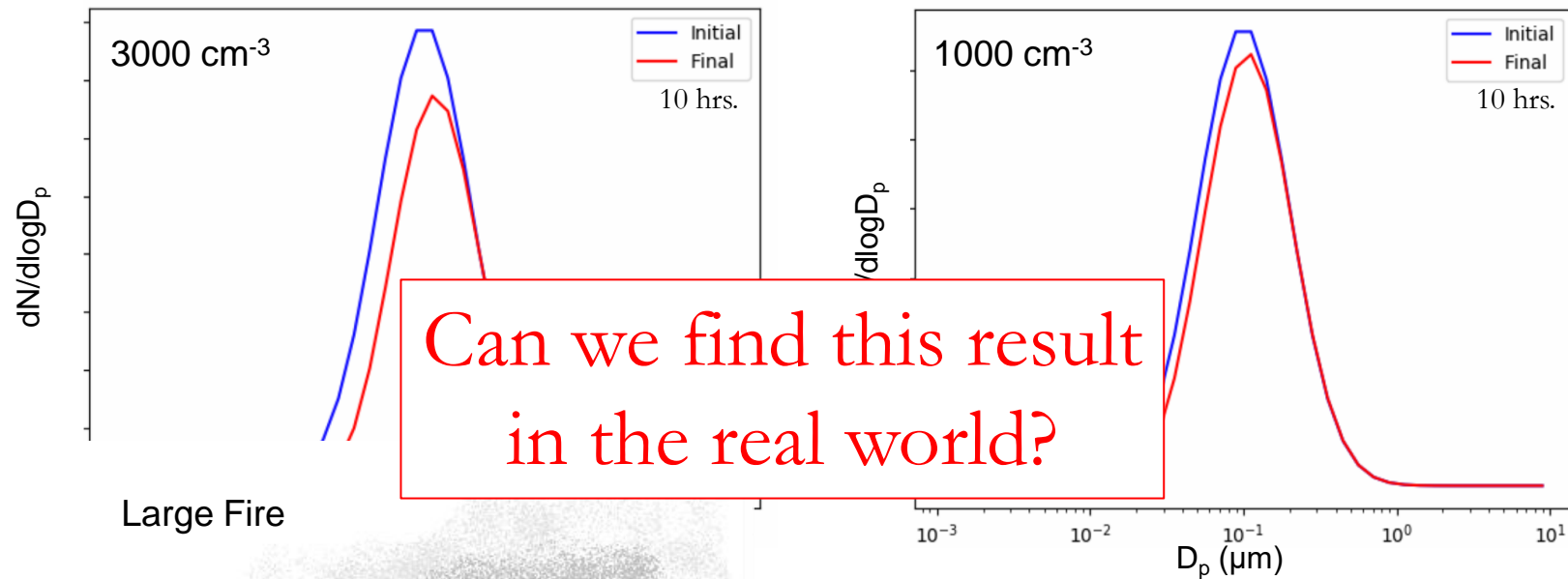
How do these issues affect coagulation and particle size in plumes?

Coagulation:

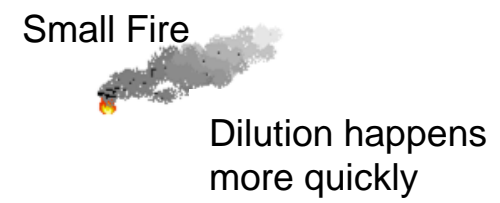
- (1) removes number
- (2) increases median diameter (D_{pg})
- (3) decreases modal width (σ_g)



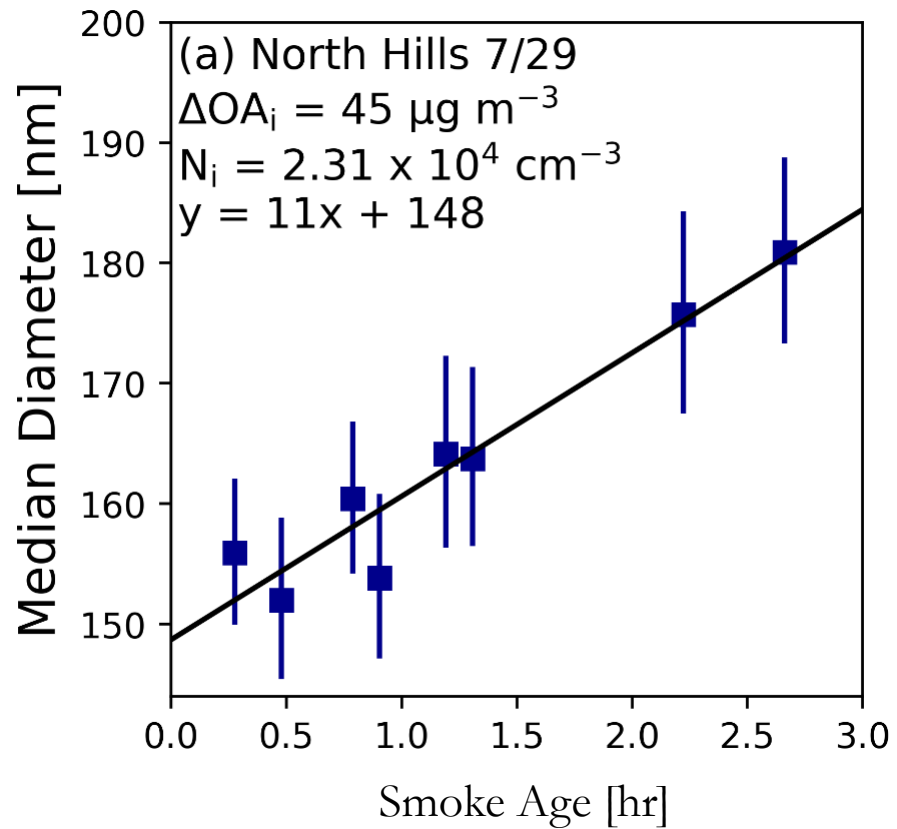
Higher number concentrations cause more coagulation.



Can we find this result in the real world?

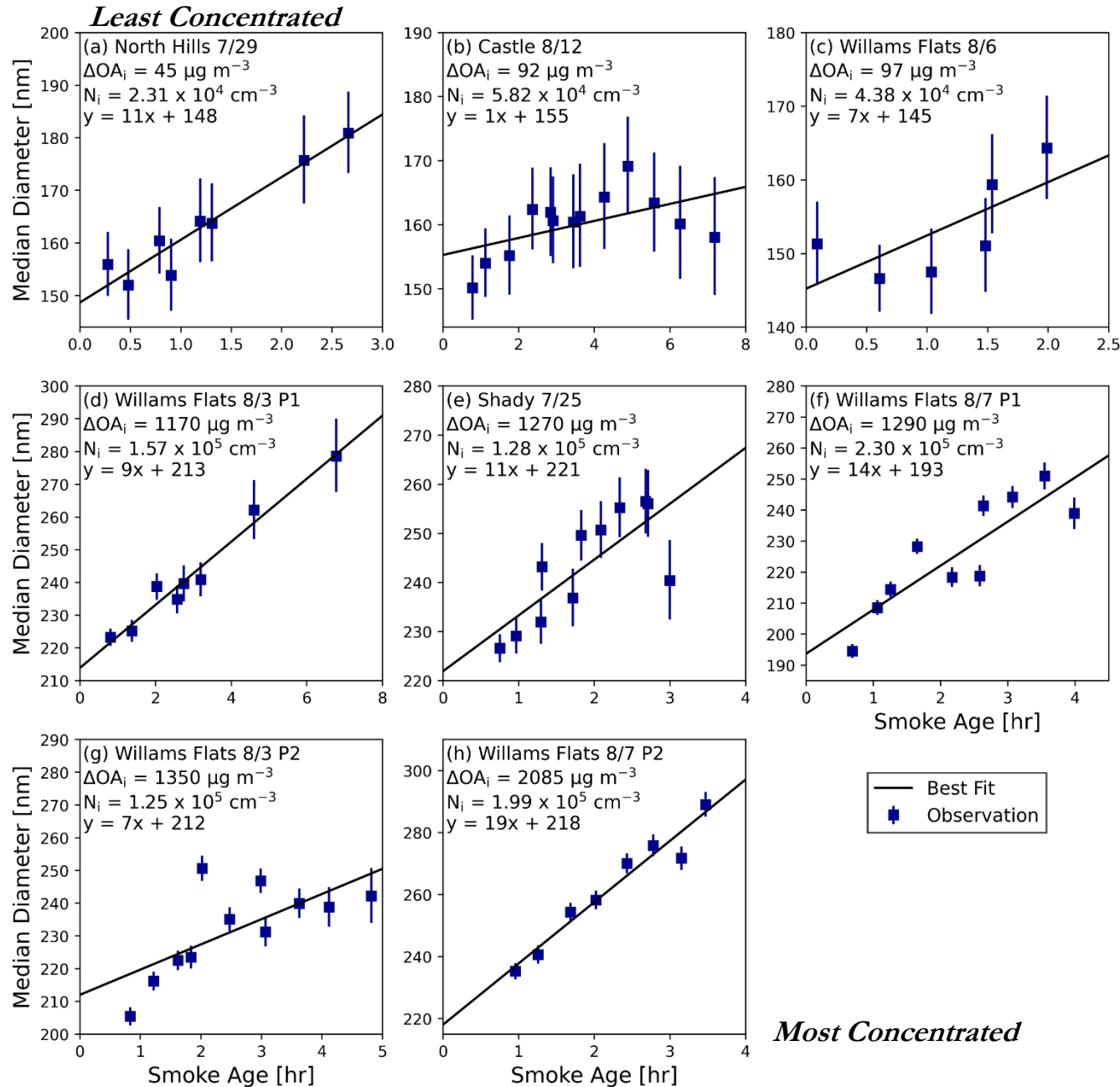


Example plume:
The aerosol diameter increases as smoke ages.



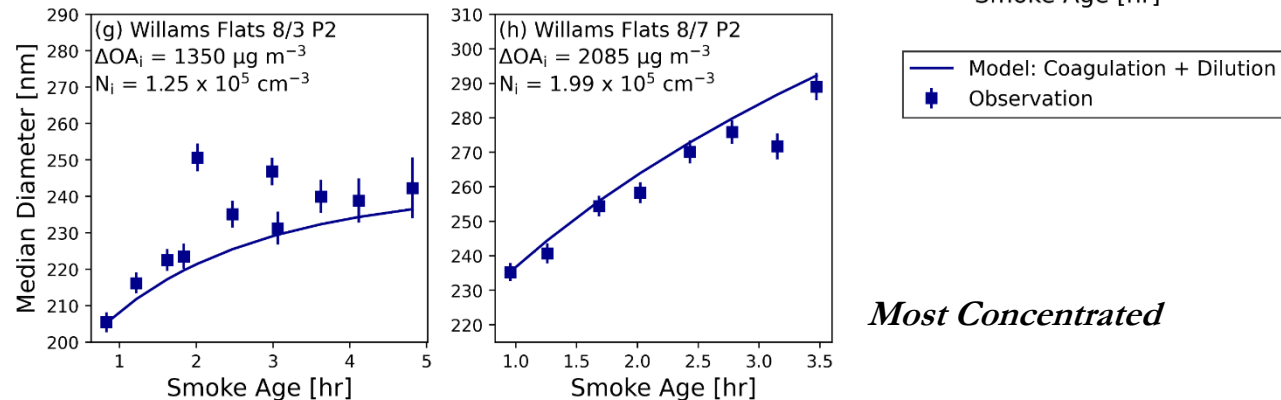
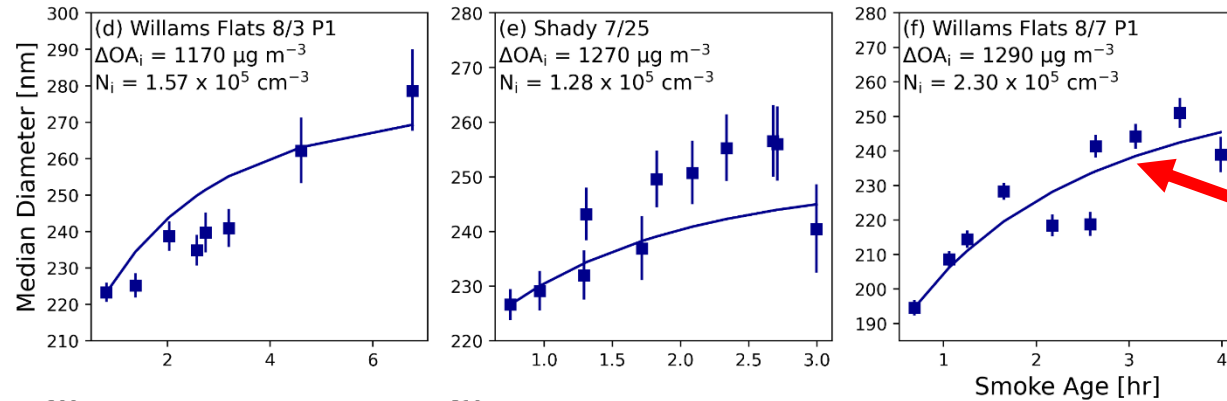
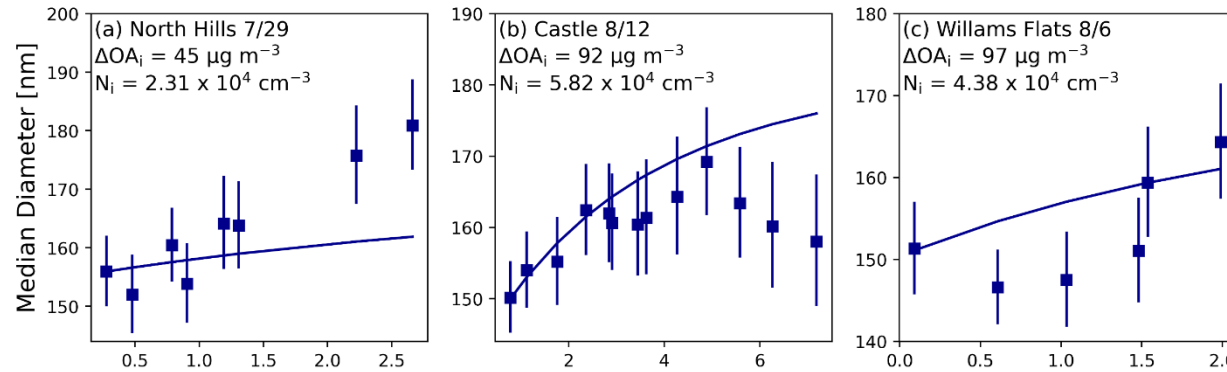
June et al., ACP, 2022

8 observed plumes show particle growth as smoke ages.



Coagulation (and dilution) can explain most of the diameter changes.

Least Concentrated

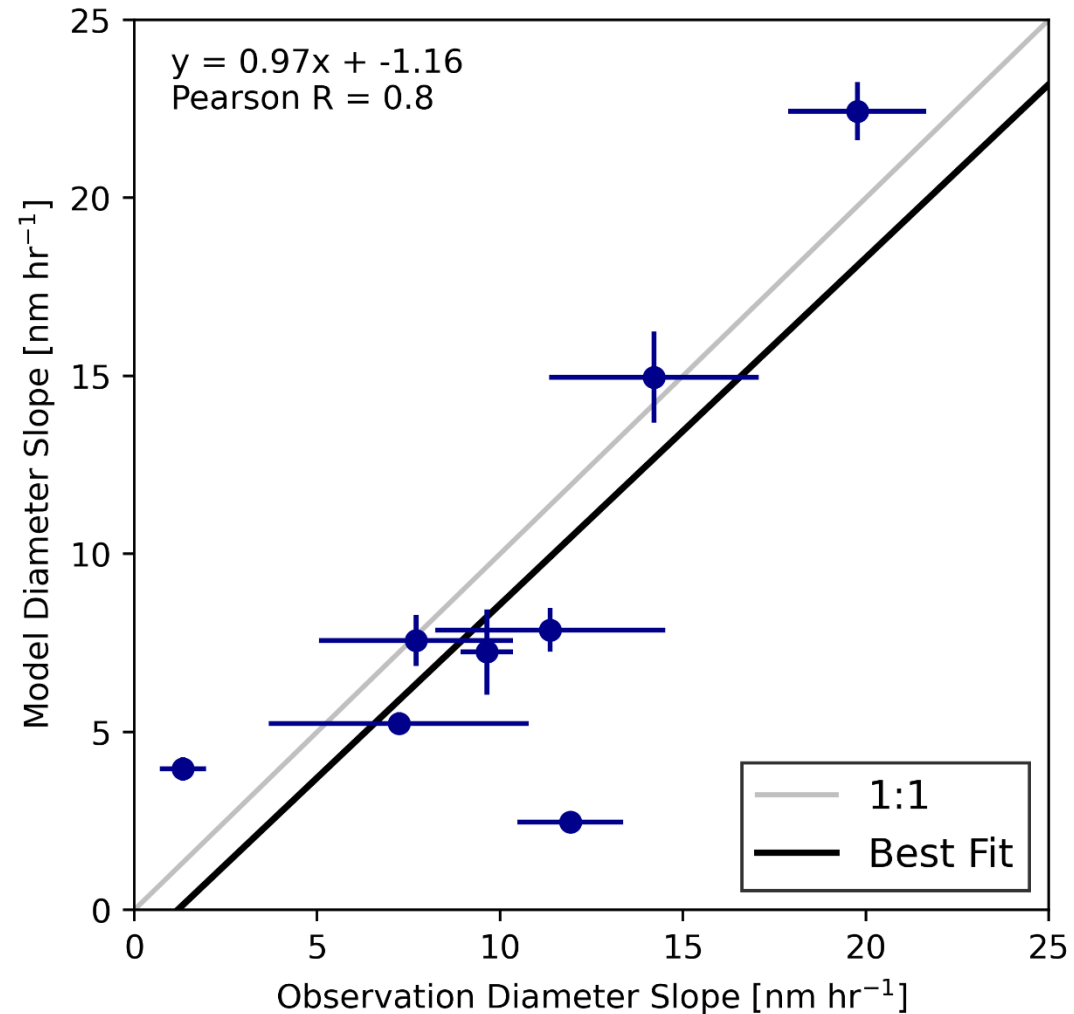


Most Concentrated



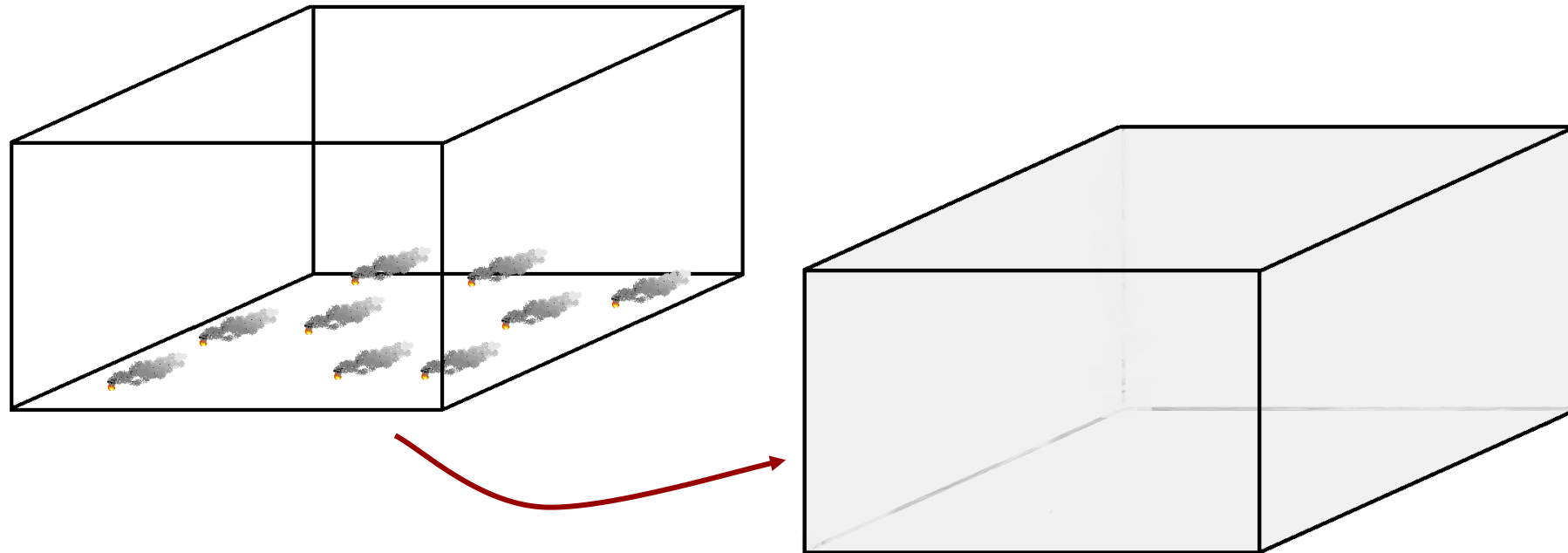
Line is model of coagulation and plume dilution only

We can explain the variability in particle growth by differences in concentration-driven coagulation.

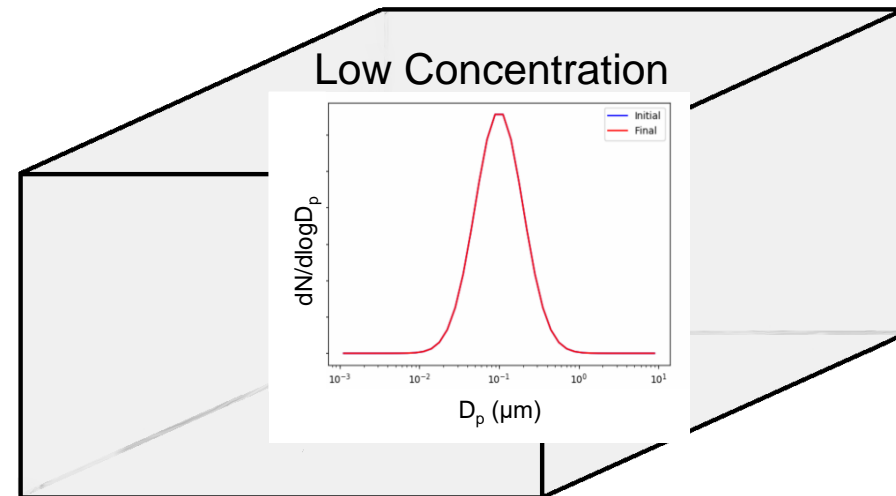
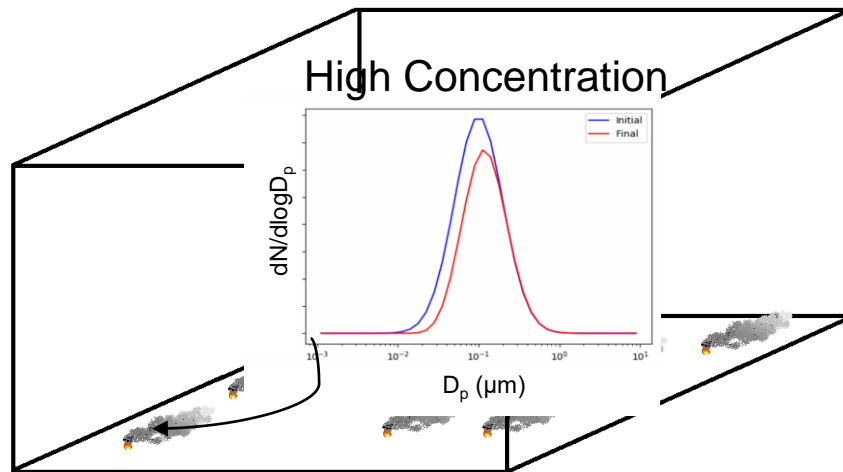


Can we represent this in 3D models?

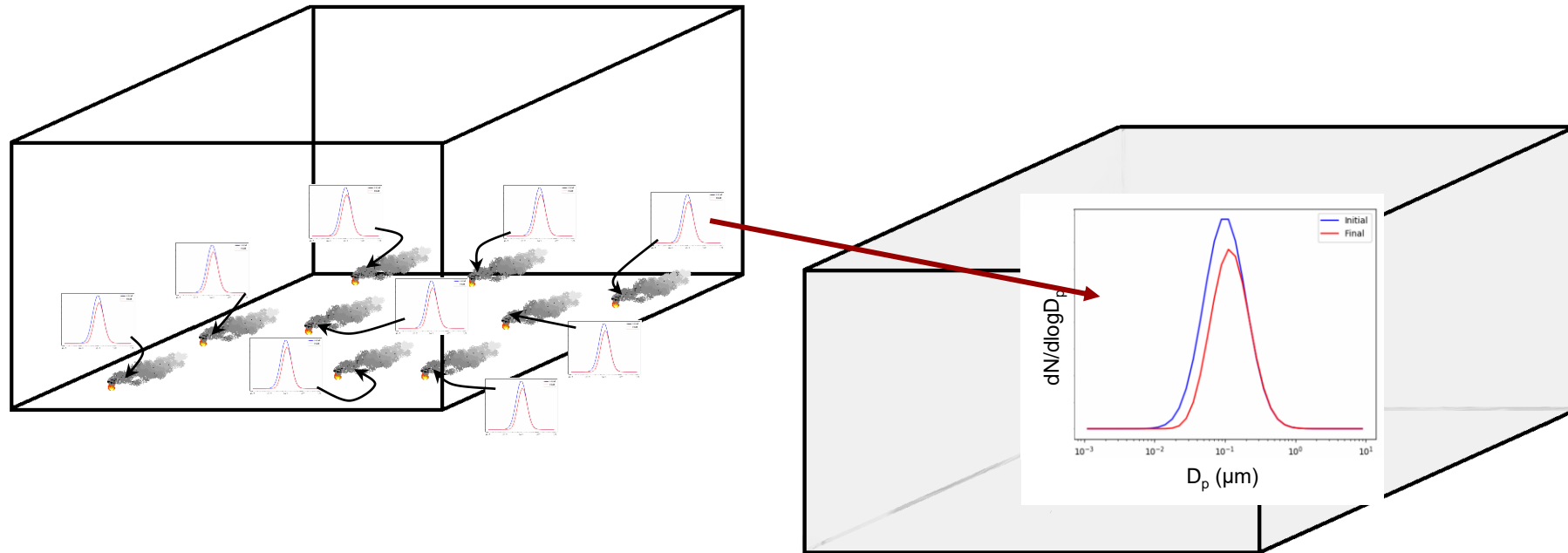
In regional & global models, *instantaneous mixing* through the full gridbox dilutes the aerosol plume.



Inadequate aging due to instant dilution can change the predicted size distribution.



With an in-plume coagulation parameterization, we can include the plume coagulation in the model.



Ran many plume coagulation simulations,
fit results to simple equation for 3D models.

Median diameter after sub-grid coagulation

$$D_{pm} = D_{pm0} + A \left[\frac{\text{Emissions}}{(\text{Wind Speed})(\text{Mixing Depth})} \right]^b (\text{time})^c$$

Emitted median diameter



Sakamoto, K. M., Laing, J. R., Stevens, R. G., Jaffe, D. A., and Pierce, J. R.: The evolution of biomass-burning aerosol size distributions due to coagulation: dependence on fire and meteorological details and parameterization, *Atmos. Chem. Phys.*, 16, 7709-7724, doi:10.5194/acp-16-7709-2016, 2016.

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fit results to simple equation for 3D models.

Median diameter after sub-grid coagulation

$$\downarrow D_{pm} = D_{pm0} + A \left[\frac{\text{Emissions}}{(\text{Wind Speed})(\text{Mixing Depth})} \right]^b (\text{time})^c$$

Emitted median diameter



Sakamoto, K. M., Laing, J. R., Stevens, R. G., Jaffe, D. A., and Pierce, J. R.: The evolution of biomass-burning aerosol size distributions due to coagulation: dependence on fire and meteorological details and parameterization, *Atmos. Chem. Phys.*, 16, 7709-7724, doi:10.5194/acp-16-7709-2016, 2016.

Ran many plume coagulation simulations,
fit results to simple equation for 3D models.

Median diameter after sub-grid coagulation

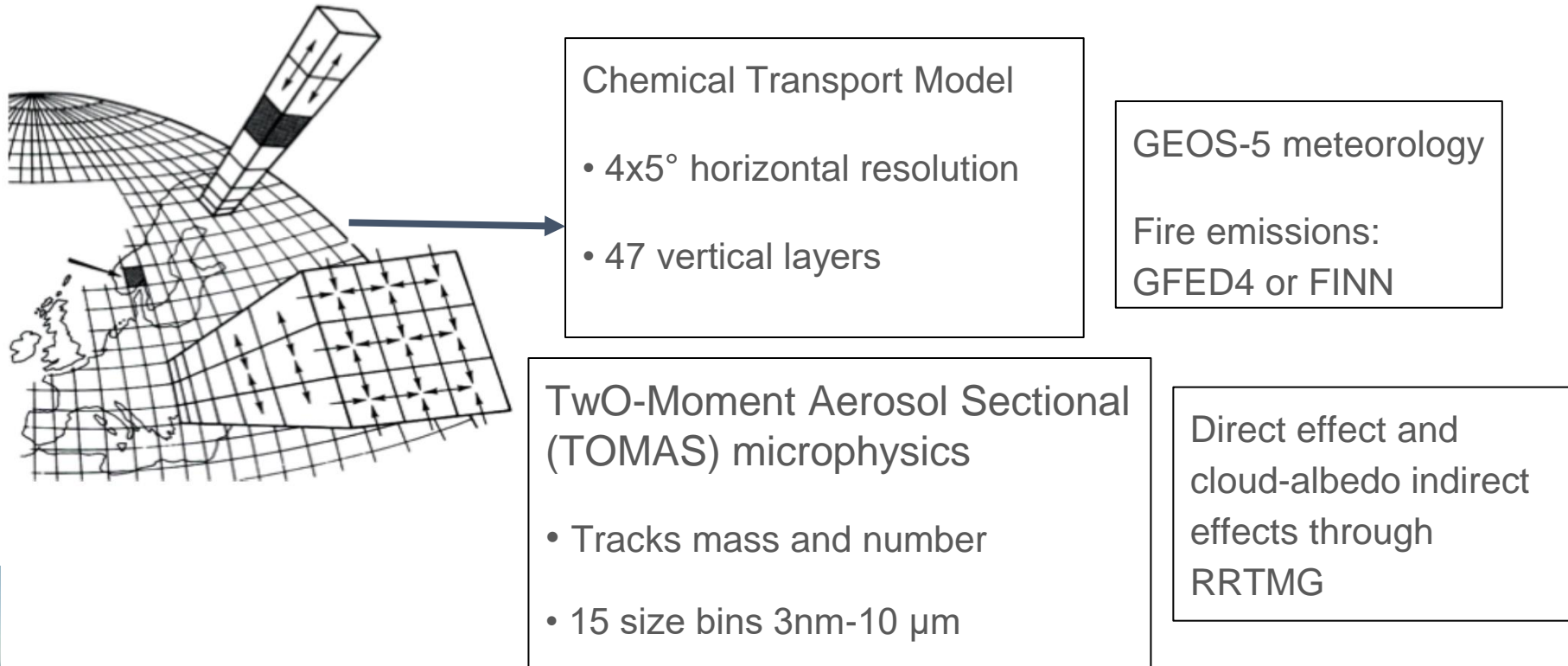
$$\uparrow D_{pm} = D_{pm0} + A \left[\frac{\text{Emissions}}{(\text{Wind Speed})(\text{Mixing Depth})} \right]^b (\text{time}) \uparrow$$

Emitted median diameter



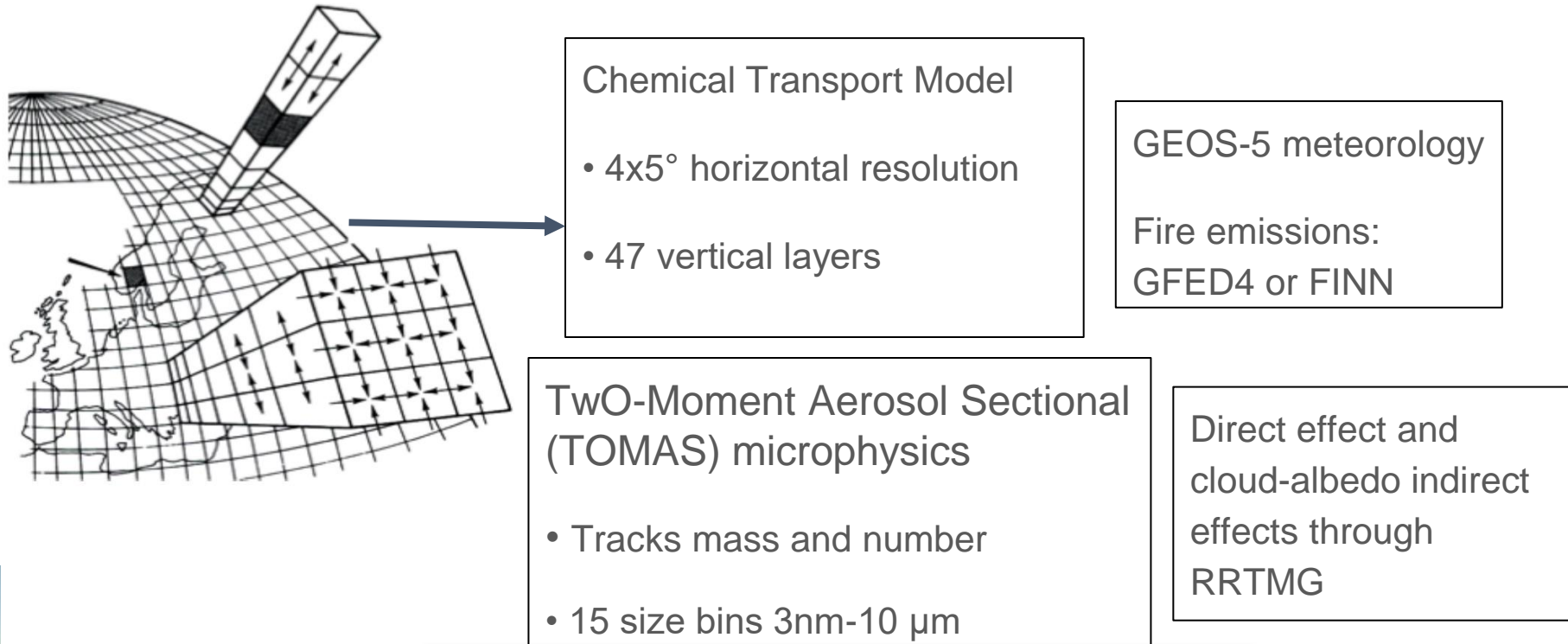
Sakamoto, K. M., Laing, J. R., Stevens, R. G., Jaffe, D. A., and Pierce, J. R.: The evolution of biomass-burning aerosol size distributions due to coagulation: dependence on fire and meteorological details and parameterization, *Atmos. Chem. Phys.*, 16, 7709-7724, doi:10.5194/acp-16-7709-2016, 2016.

Estimate the global effects using GEOS-Chem TOMAS.



Ramnarine, E., Kodros, J. K., Hodshire, A. L., Lonsdale, C. R., Alvarado, M. J., and Pierce, J. R.: Effects of Near-Source Coagulation of Biomass Burning Aerosols on Global Predictions of Aerosol Size Distributions and Implications for Aerosol Radiative Effects, *Atmos. Chem. Phys.*, <https://doi.org/10.5194/acp-2018-1084>, 2019.

Test with and without subgrid coagulation.

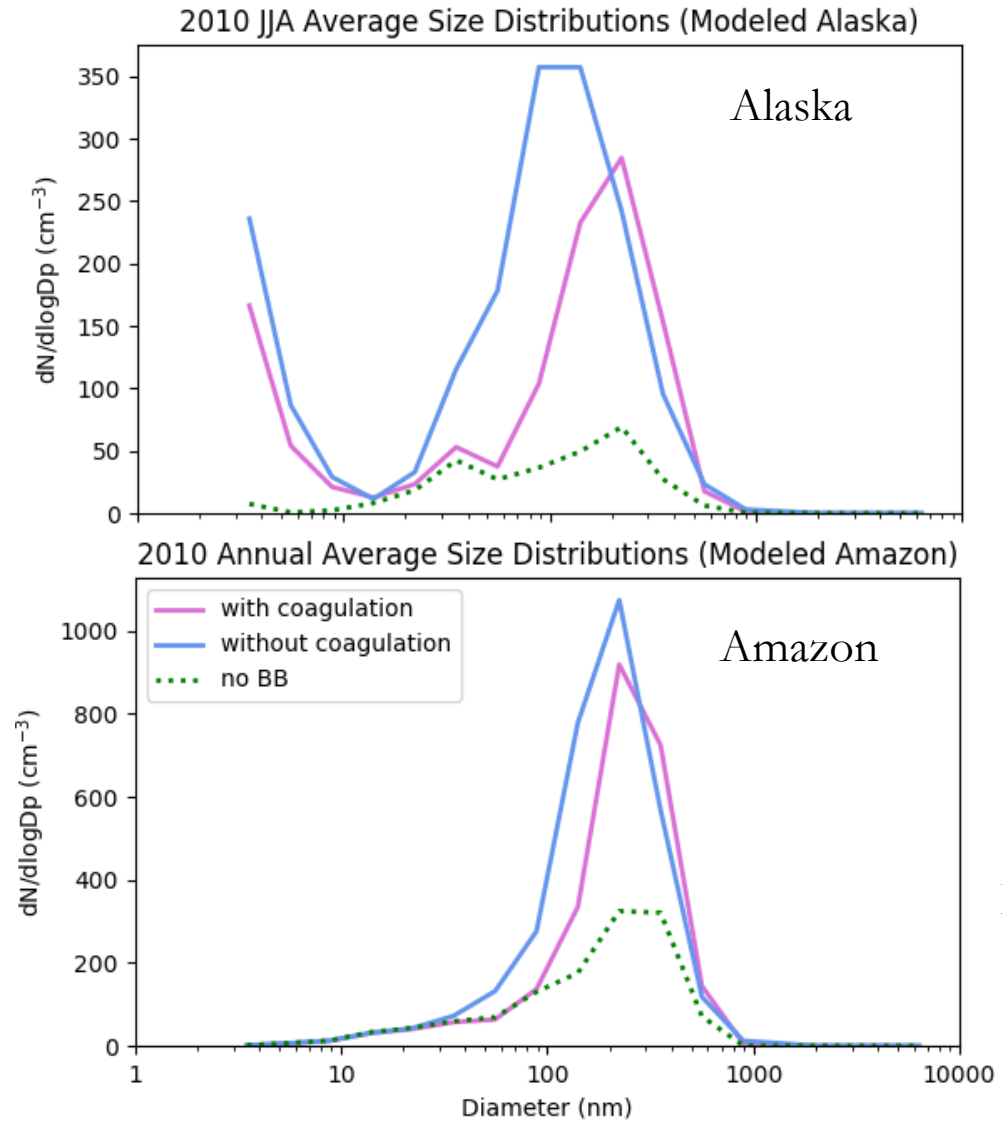


$$D_{pm} = D_{pm0} + A \left[\frac{\text{Emissions}}{(\text{Wind Speed})(\text{Mixing Depth})} \right]^b (\text{time})^c$$



Ramnarine, E., Kodros, J. K., Hodshire, A. L., Lonsdale, C. R., Alvarado, M. J., and Pierce, J. R.: Effects of Near-Source Coagulation of Biomass Burning Aerosols on Global Predictions of Aerosol Size Distributions and Implications for Aerosol Radiative Effects, *Atmos. Chem. Phys.*, <https://doi.org/10.5194/acp-2018-1084>, 2019.

Sub-grid coagulation shifts size distribution in locations dominated by smoke.

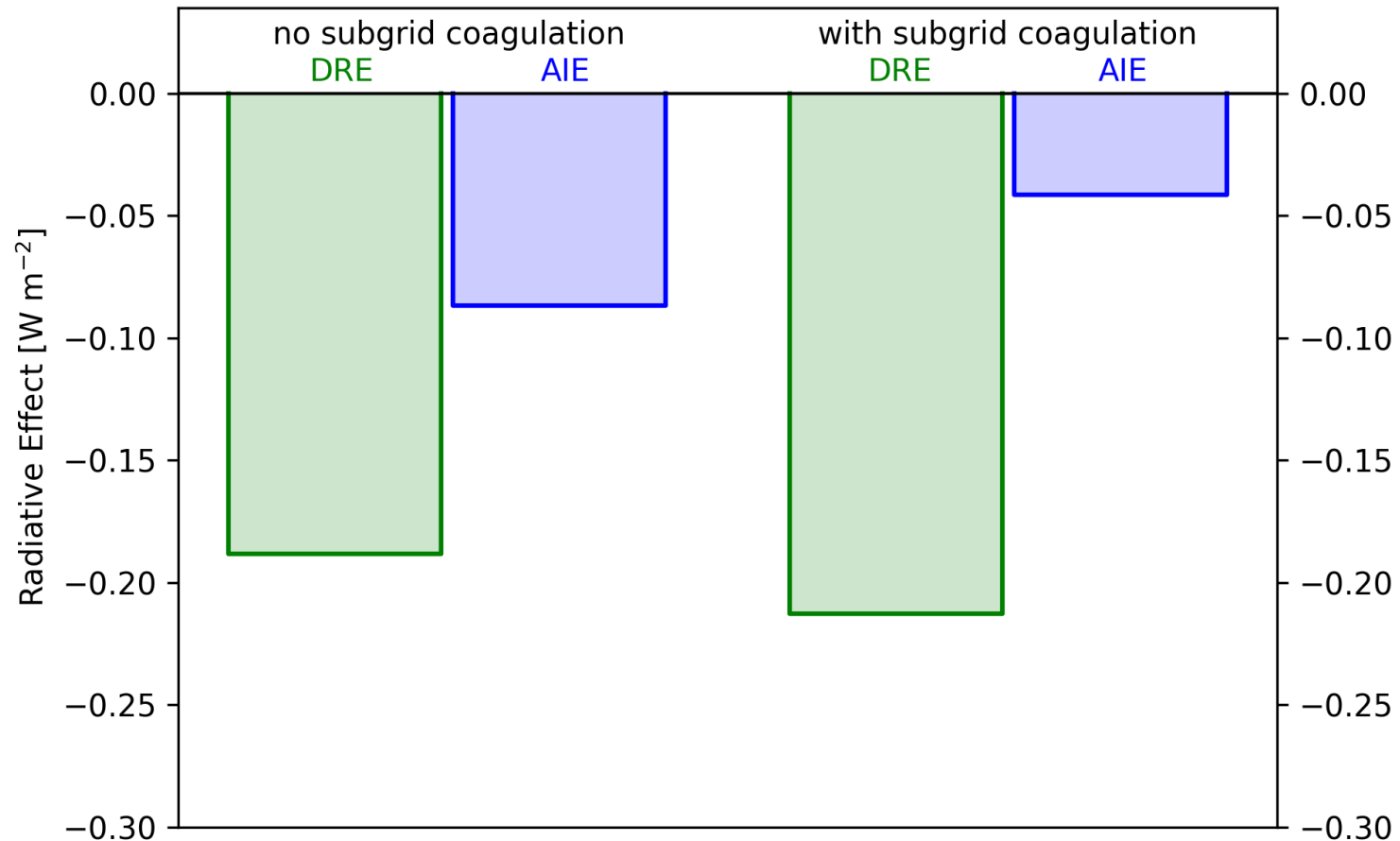


Ramnarine et al., 2019

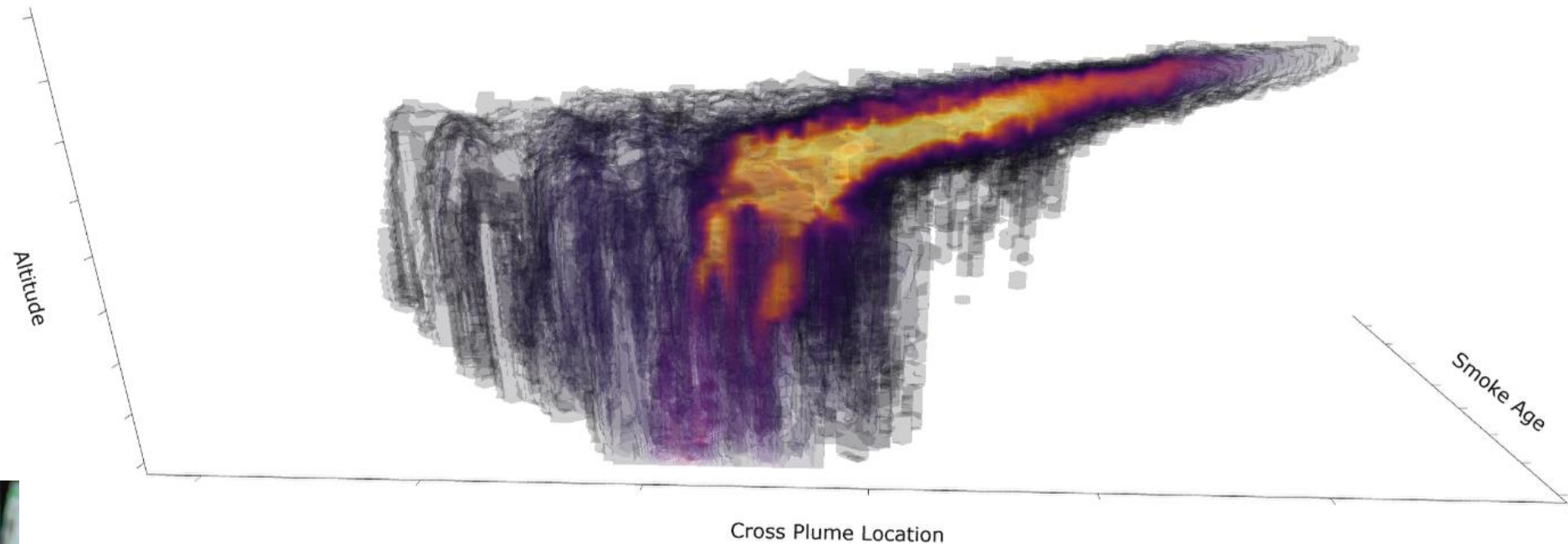
Biomass burning radiative effects:

Subgrid coag decreases AIE

Subgrid coag slightly increases DRE

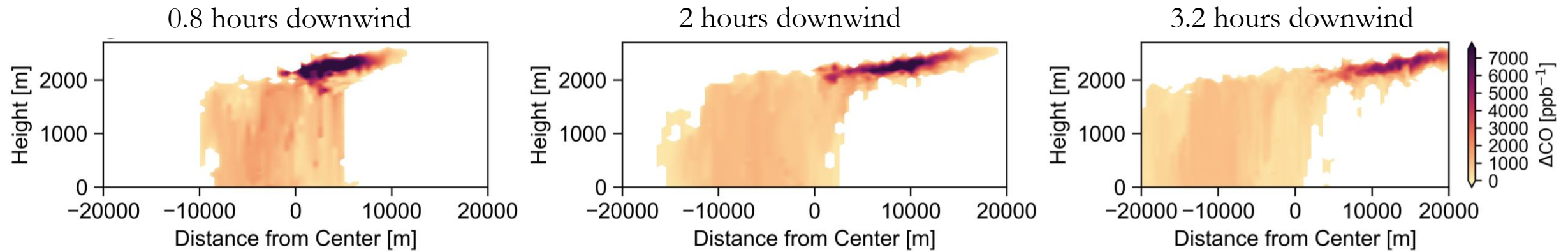


Use Large-Eddy Simulation to simulate plume dispersion and in-plume gradients.

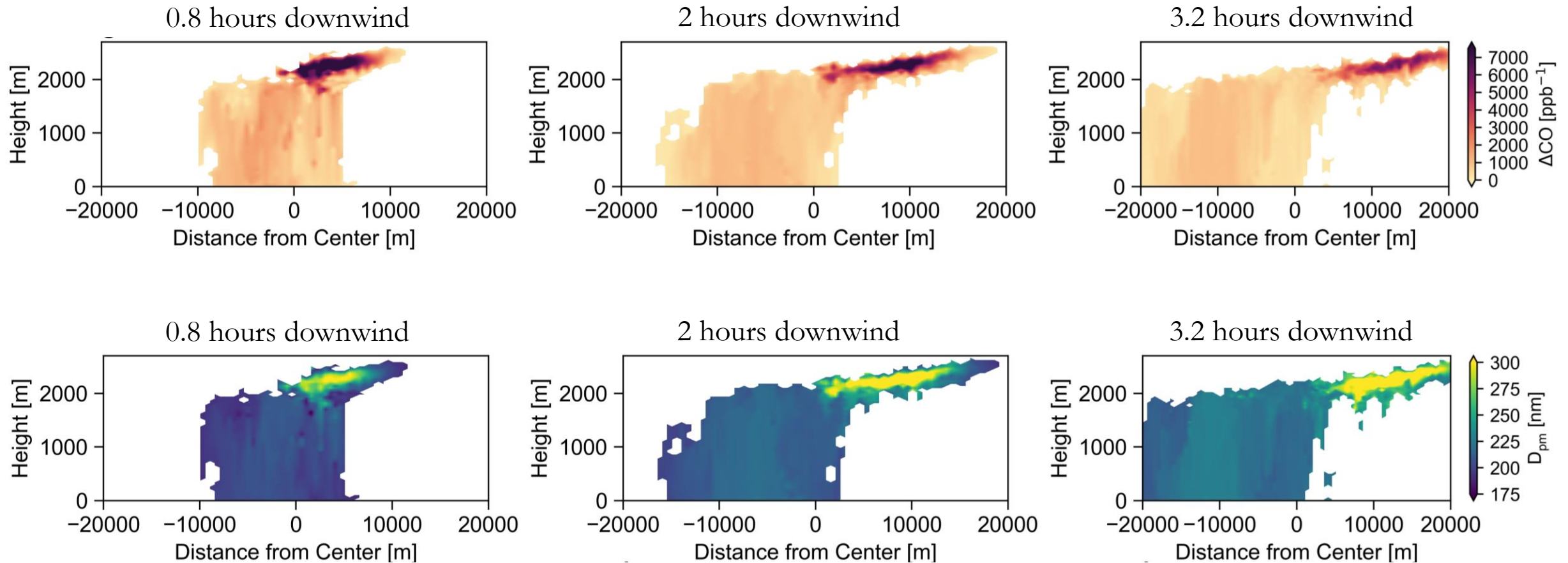


Nicole June

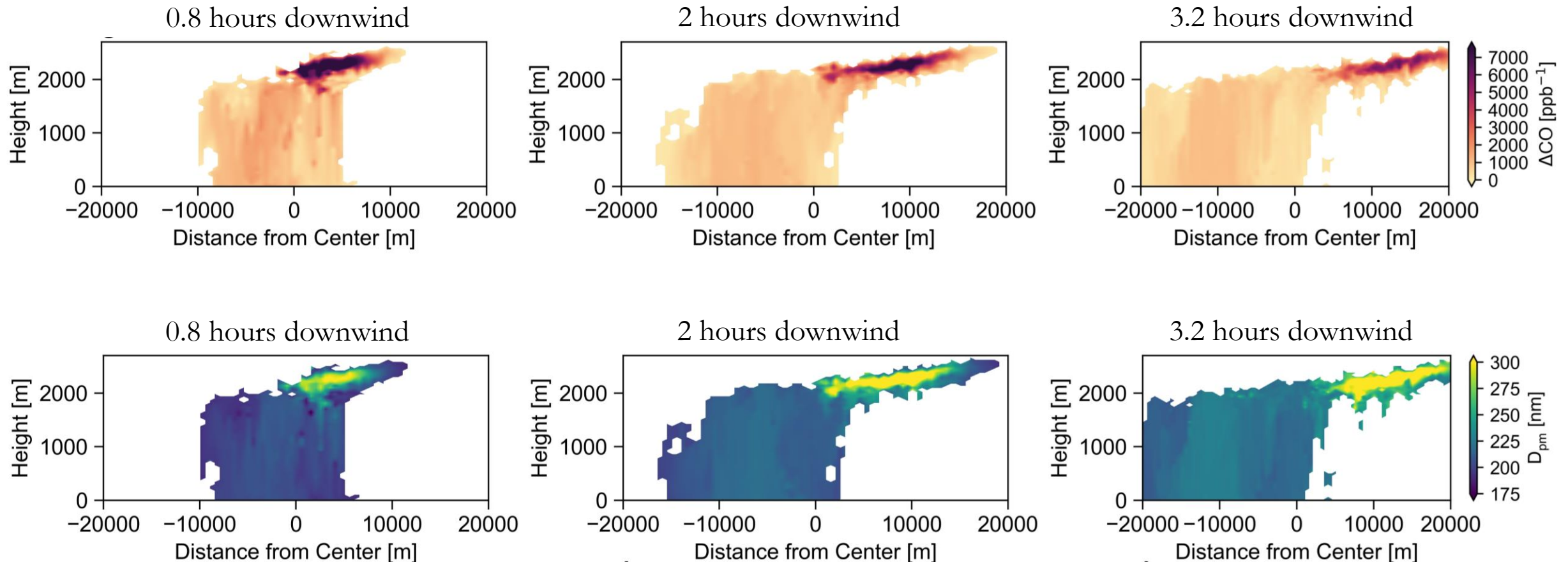
Strong concentration variability in plume “curtains” (downwind cross-sections).



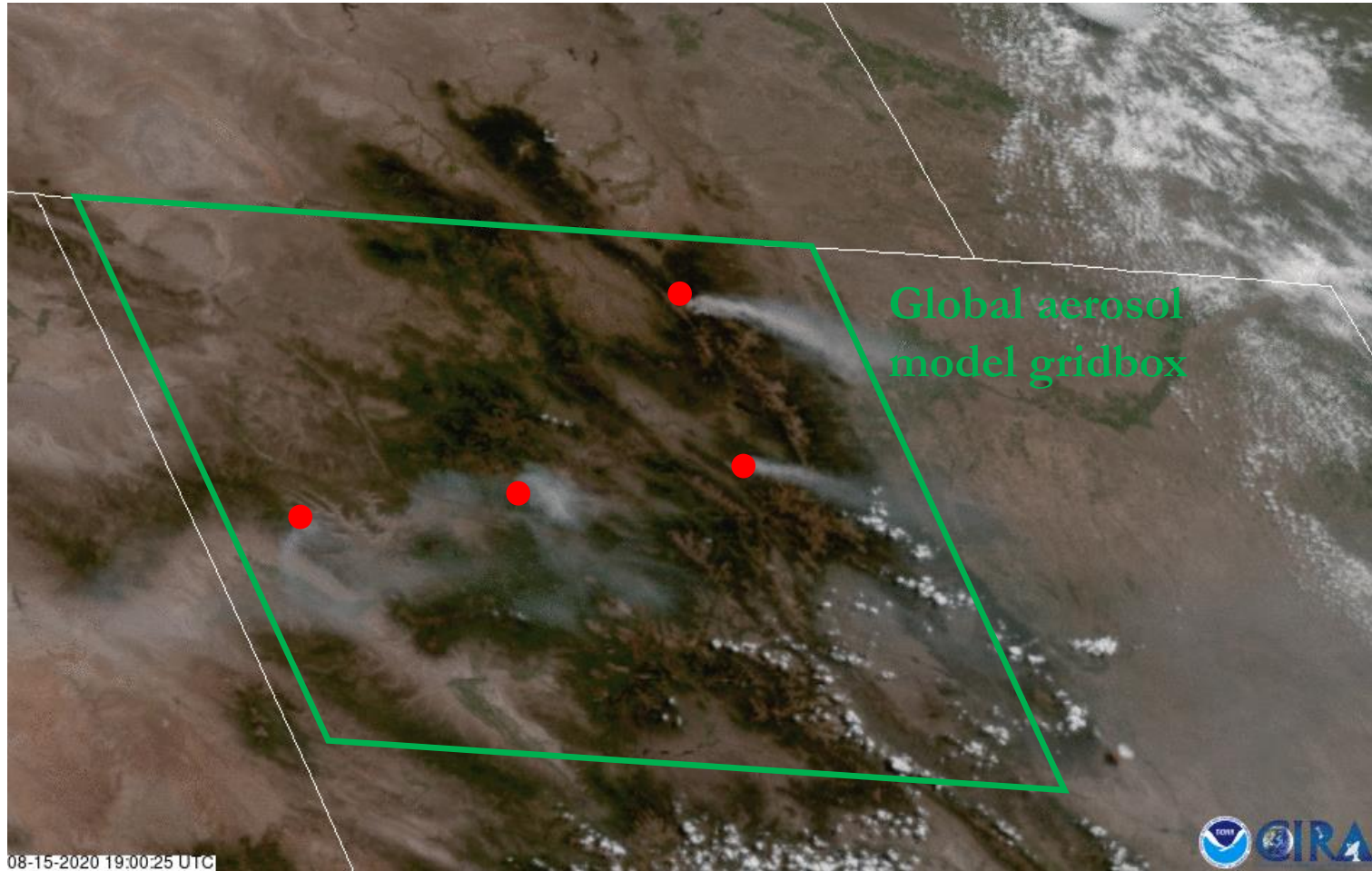
Particles are significantly larger in “core” of plume than edges (because of coagulation).



Particle size can depend on different heights/locations in plumes!
Critical to understand concentration variability to estimate biomass burning aerosol sizes and climate effects.



Plume dilution/concentrations matter for shaping aerosol properties!

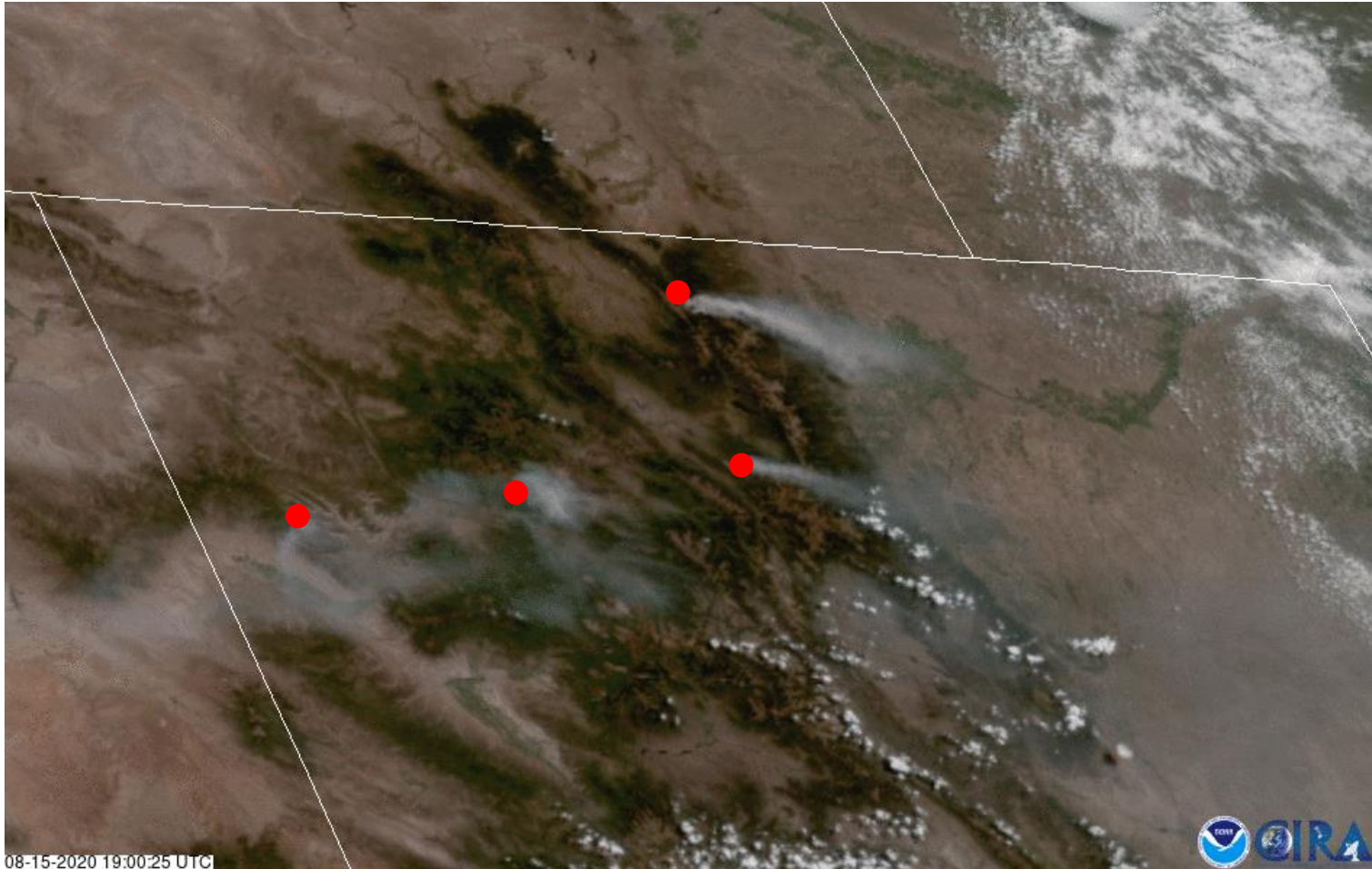


Colorado, Aug 15, 2020.

Two related issues:

1. Smoke mixes instantly into model gridboxes.
2. Real smoke plumes dilute at different rates.

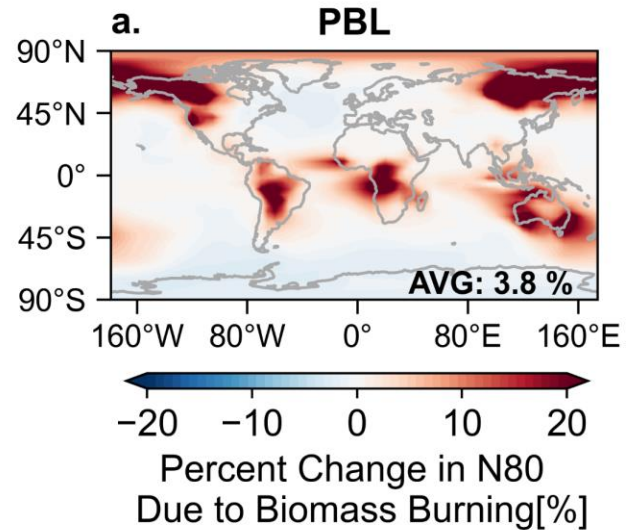
Thank you!



- Improving plume injection height is critical for estimating smoke health and climate effects.
- Plumes are often smaller than model gridboxes.
- Plume concentrations/dilution affect processes (like coagulation) that shape aerosol properties (like size).

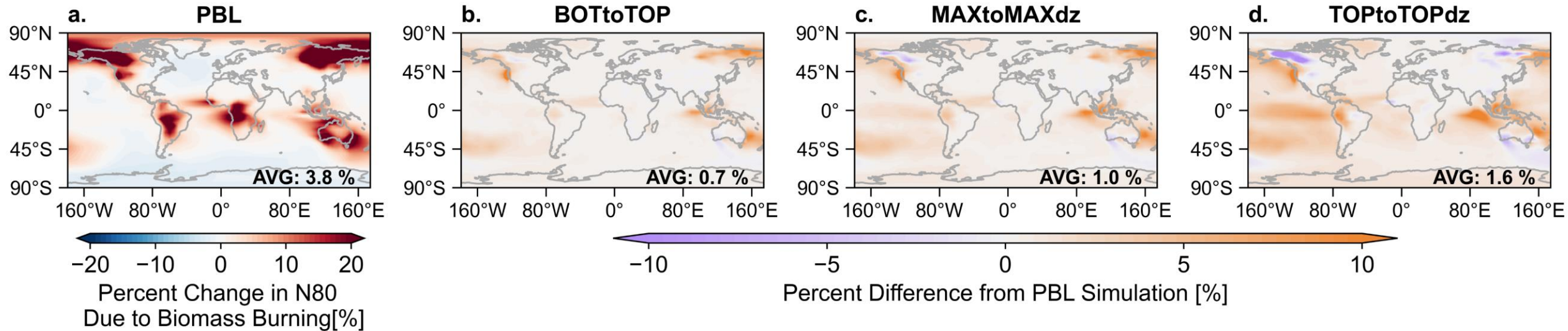
Extras

Biomass burning increases the concentration of CCN-sized particles at low cloud levels ($\eta = 0.9$ to 0.7) in source regions, lowers it in remote regions.

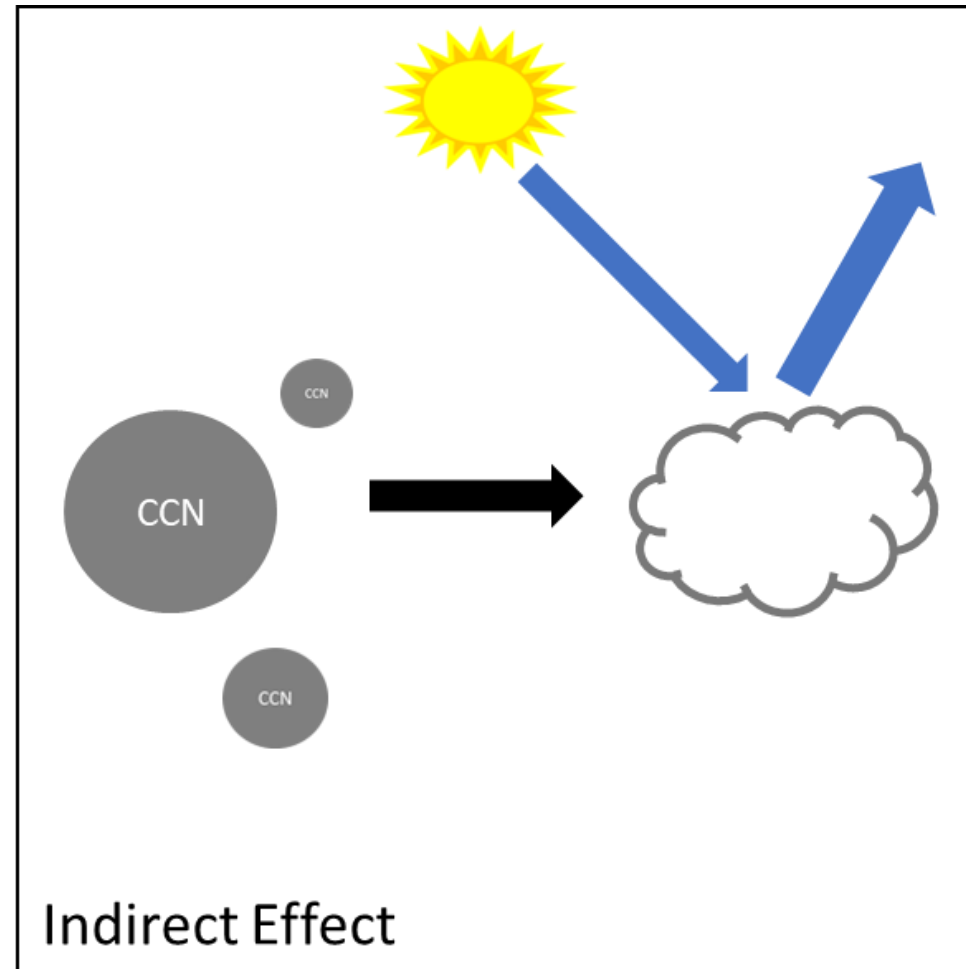
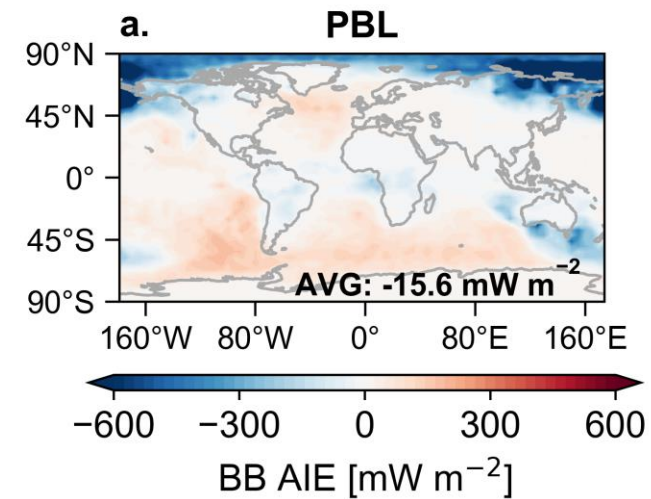


CCN sized particles at low cloud levels ($\eta = 0.9$ to 0.7) increase with increasing PIH.

Increasing Plume Injection Height \rightarrow

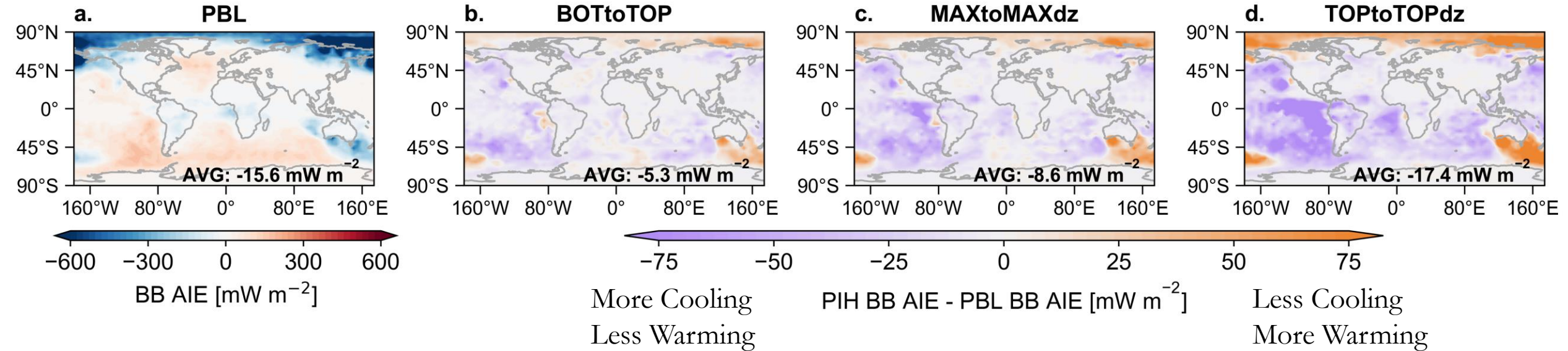


Biomass burning aerosol indirect effect mirrors CCN changes.

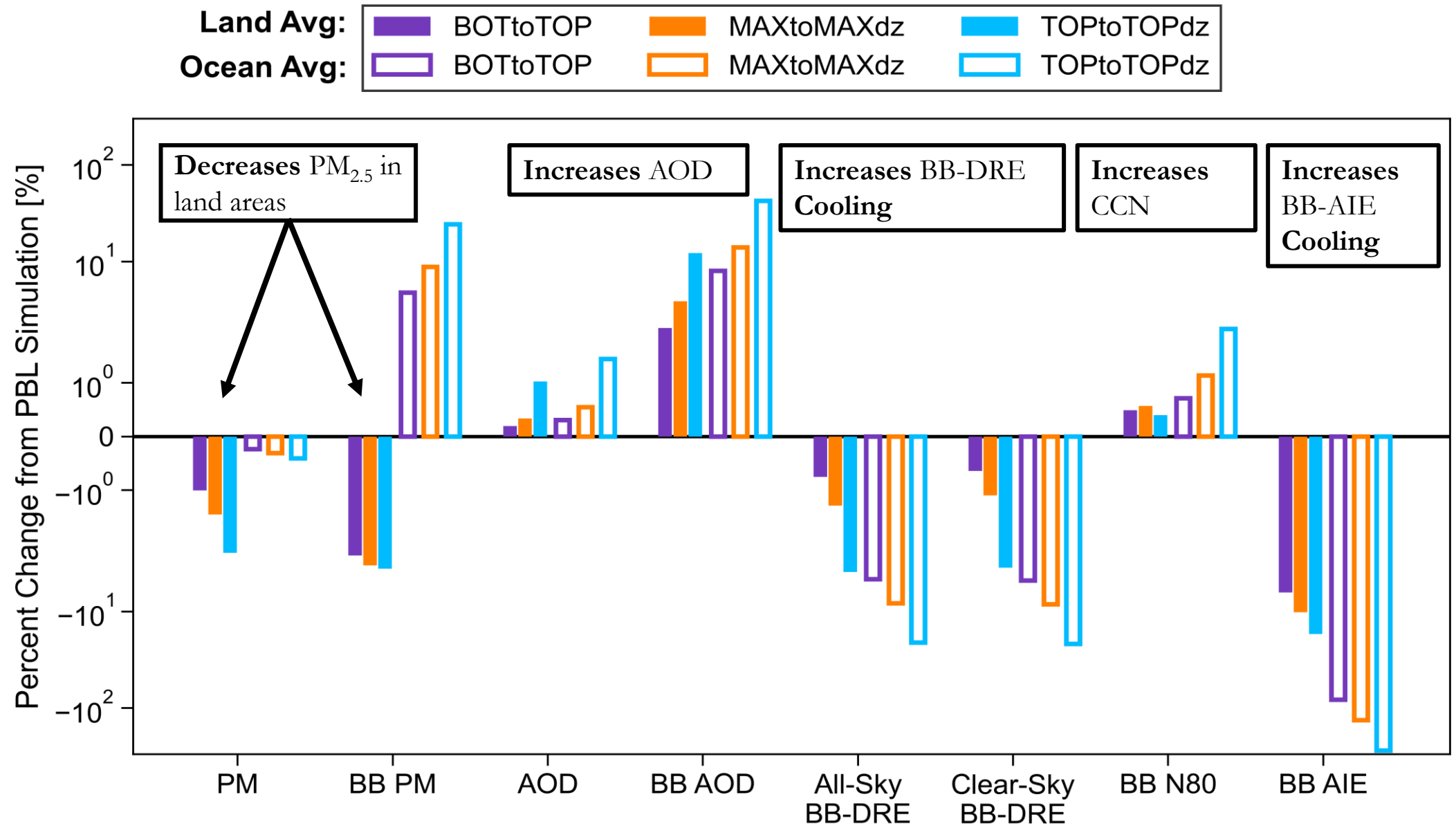


With increasing plume injection height, the cooling aerosol indirect effect of BB increases.

Increasing Plume Injection Height



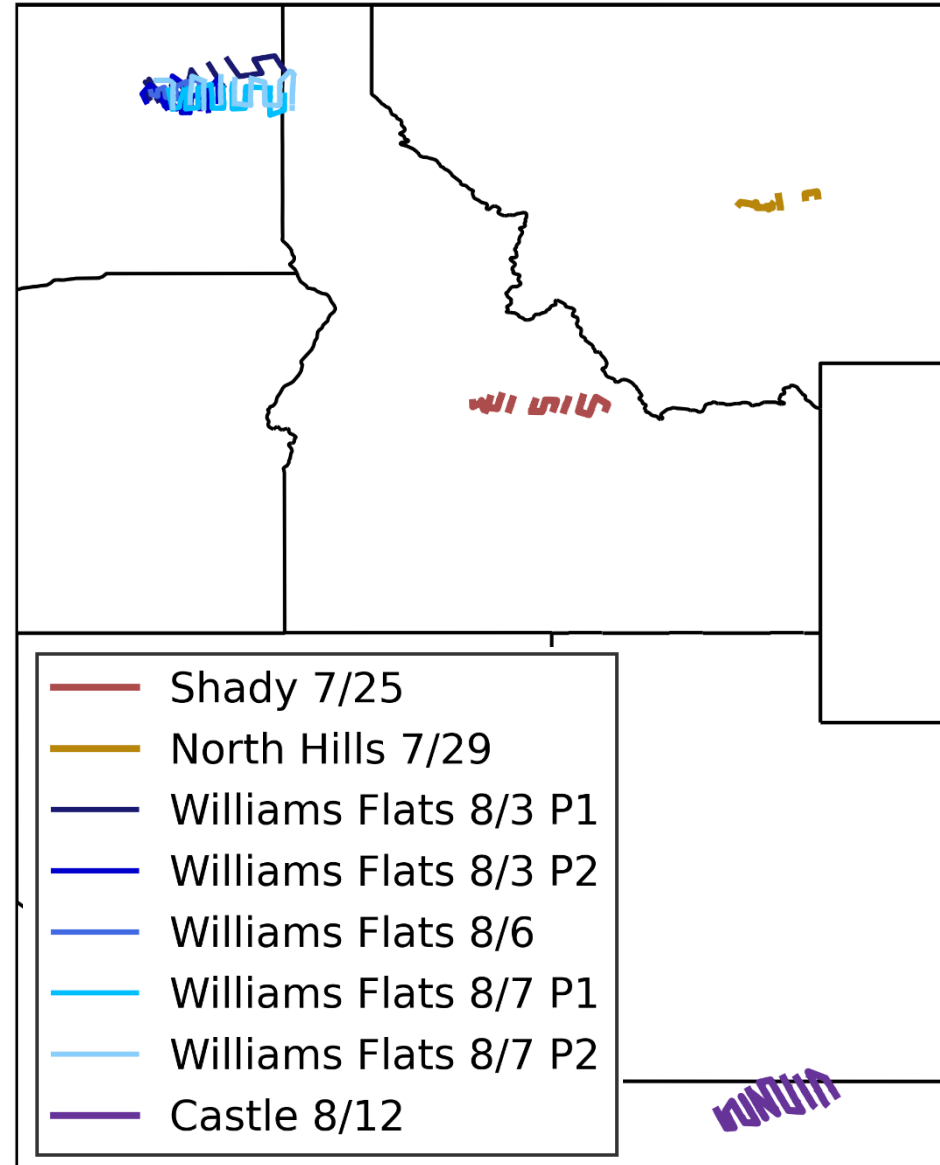
Raising the biomass burning plume injection height:



FIREX-AQ (western US portion)

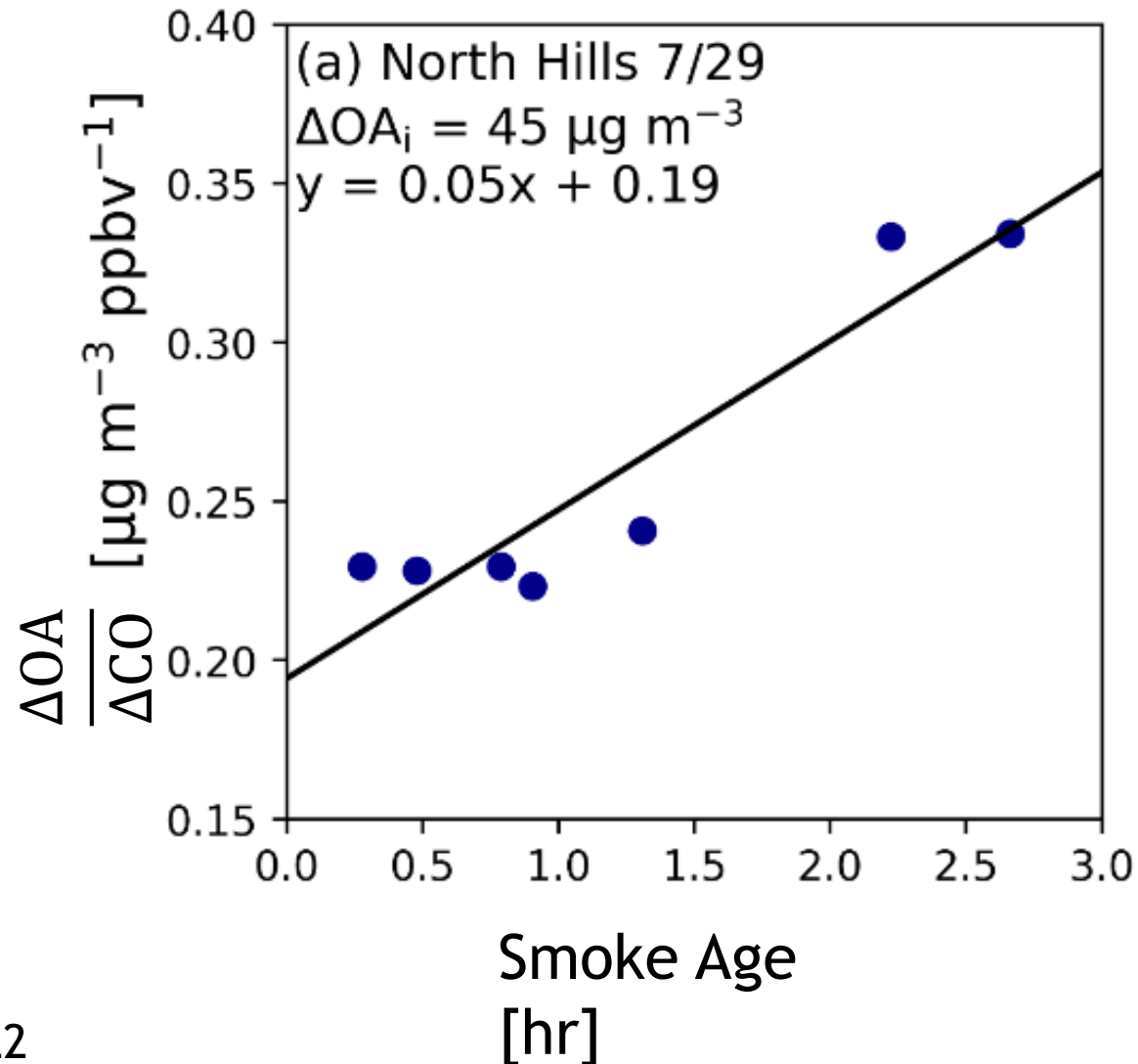


- July-August 2019
- 8 sets of pseudo-Lagrangian transects
- Measurements:
 - Aerosol size
 - OA mass
 - CO

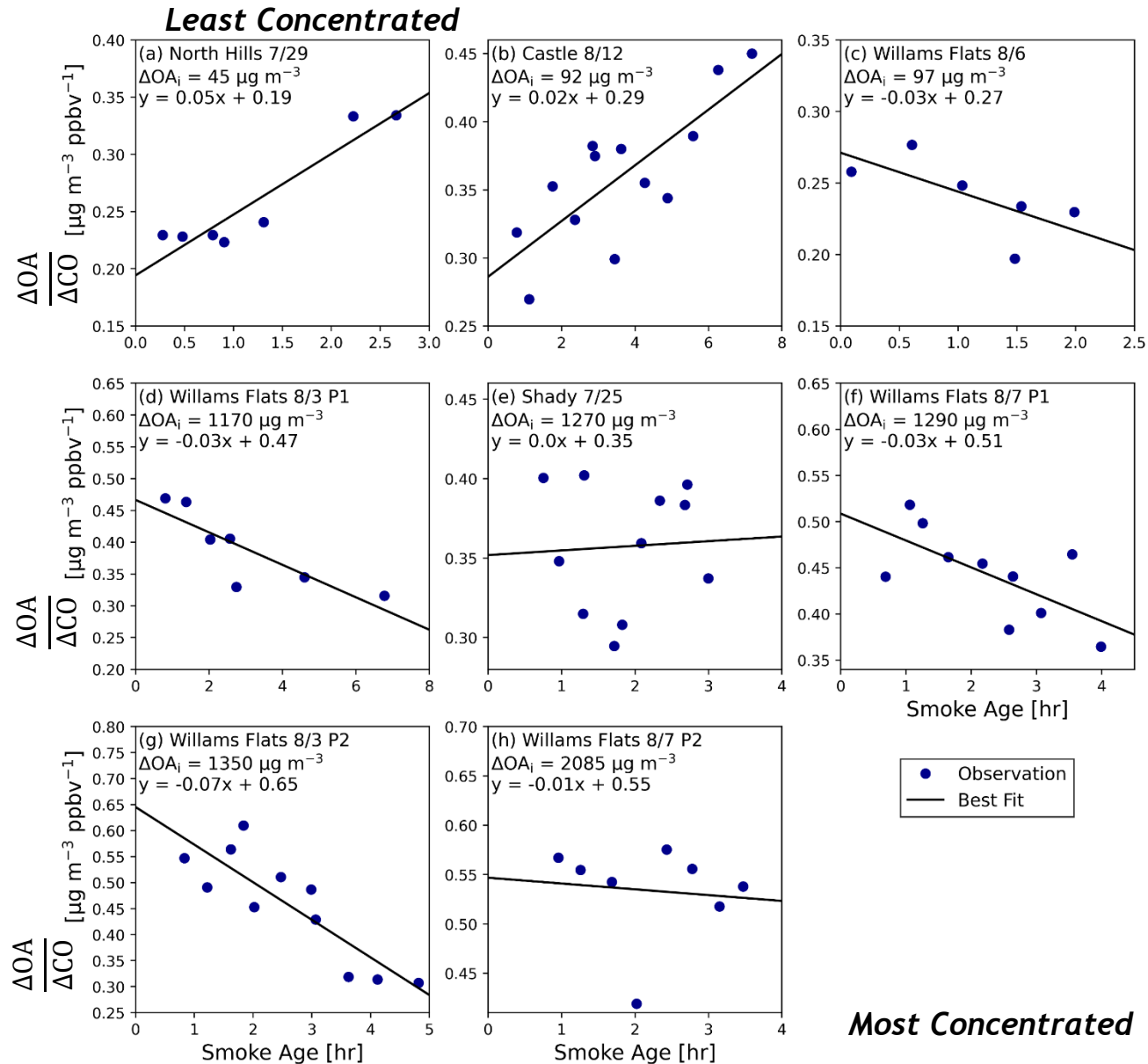


Example plume:

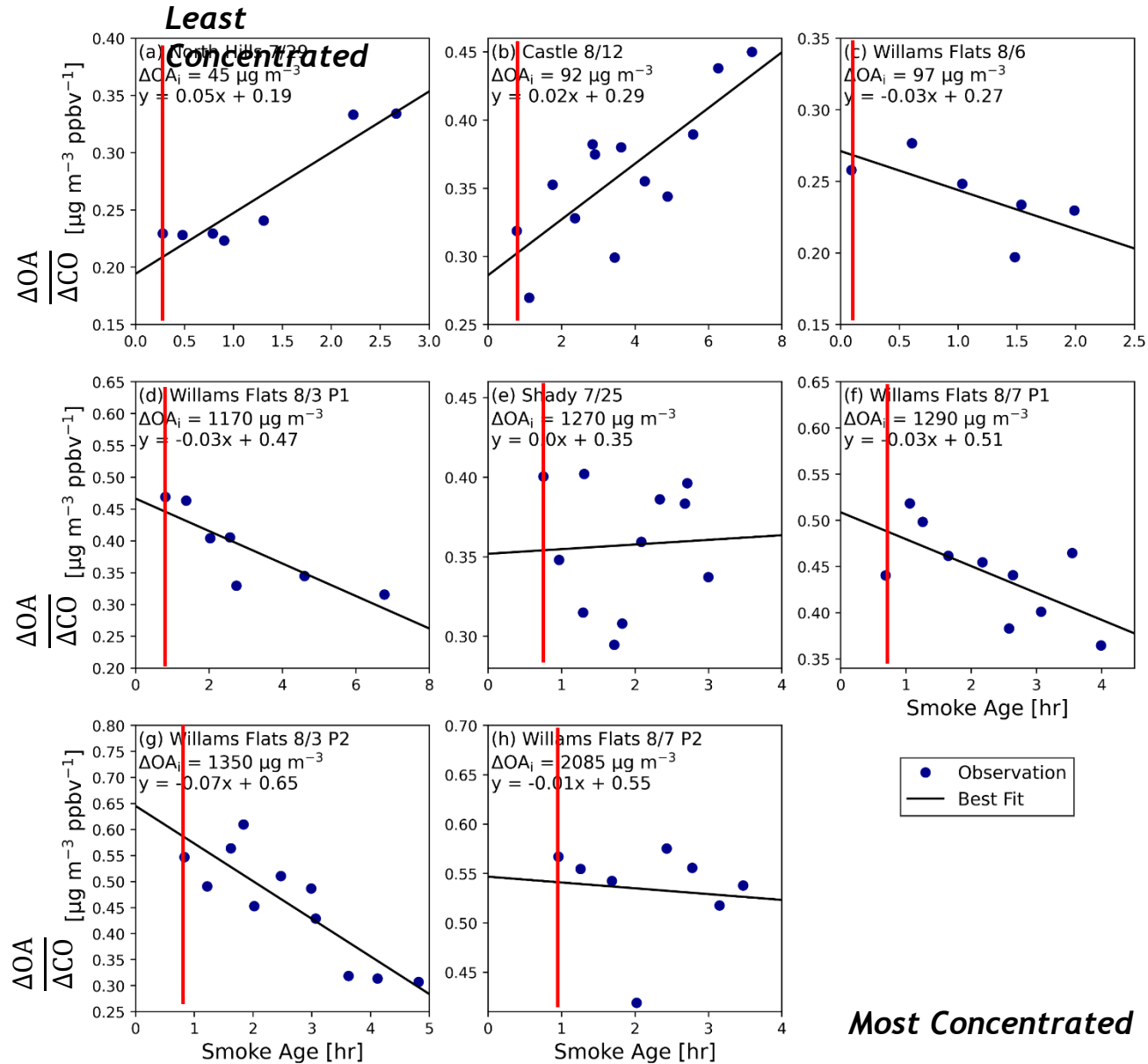
The OA enhancement ratio ($\Delta\text{OA}:\Delta\text{CO}$) changes over time.



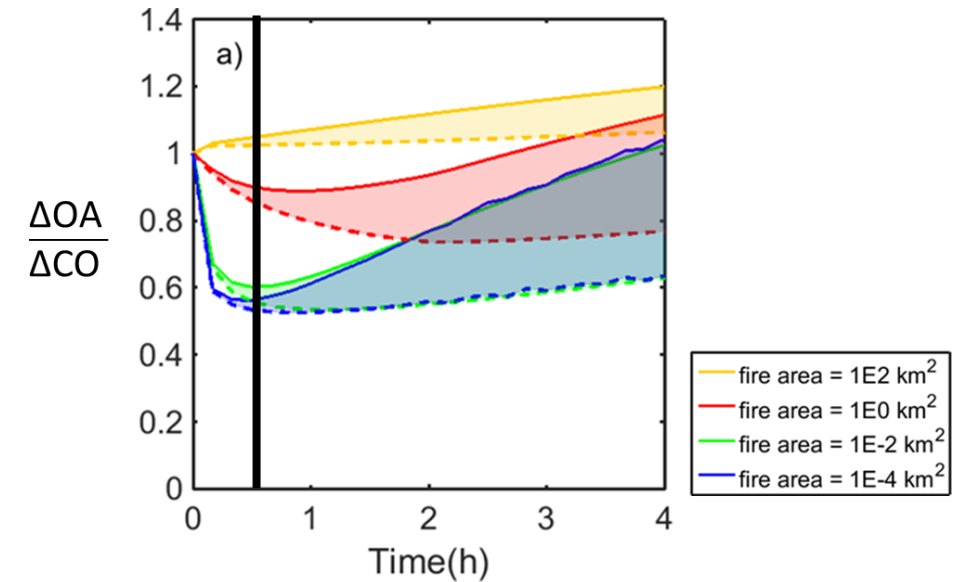
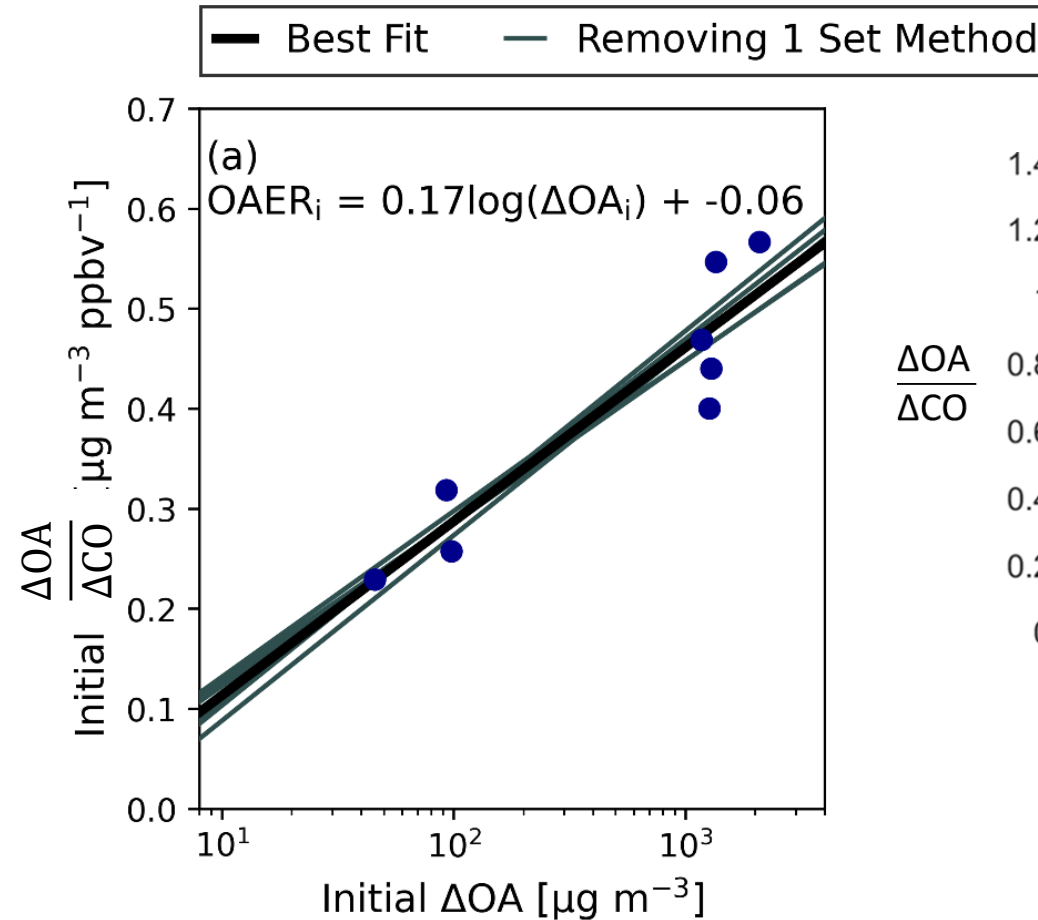
All 8 plumes: OA enhancement ratio ($\Delta\text{OA}:\Delta\text{CO}$) increases, decreases, or remains constant as smoke ages.



Are there systematic differences in $\Delta\text{OA}/\Delta\text{CO}$ at the initial transect with plume concentration?

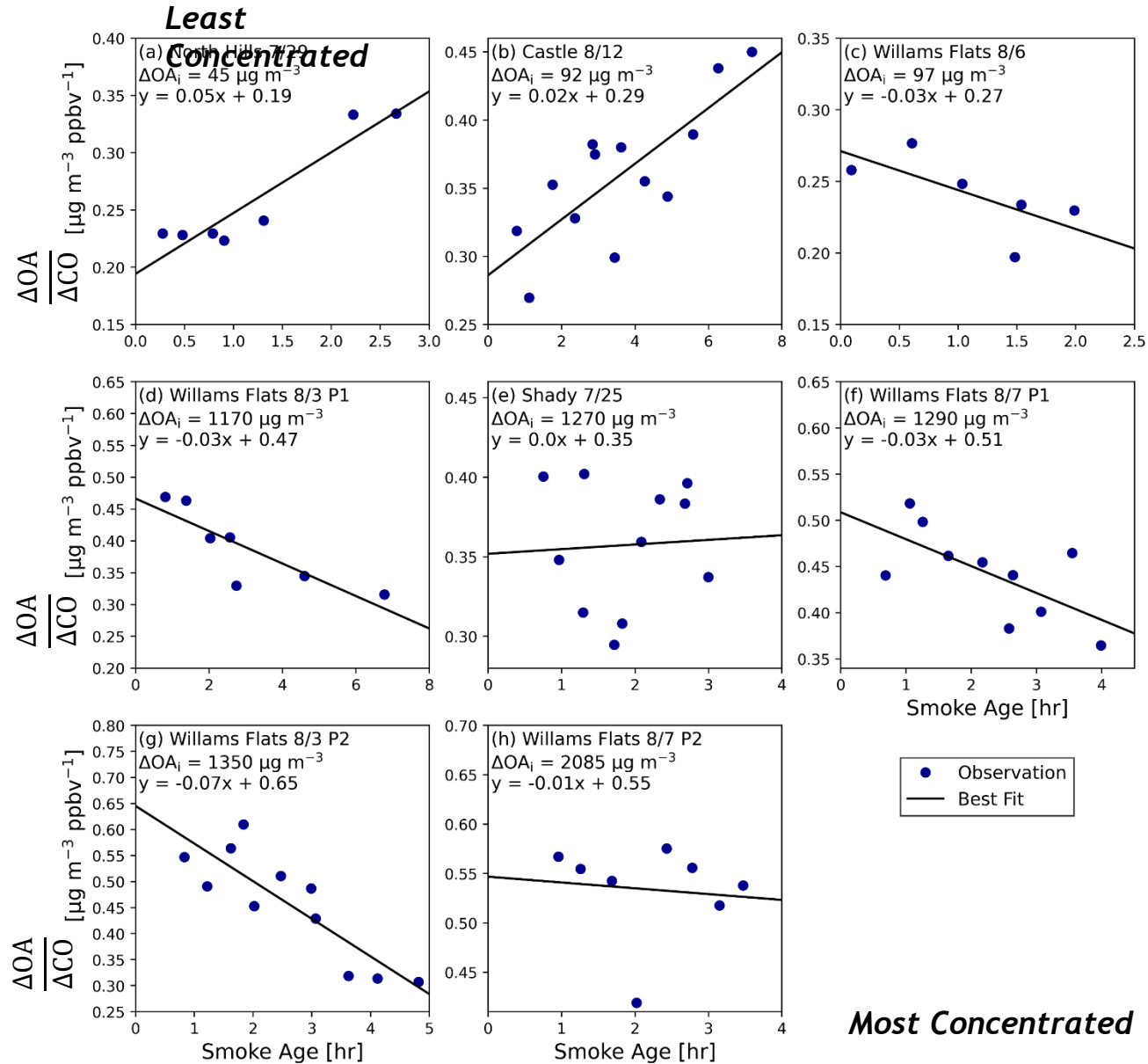


Initial $\Delta\text{OA}:\Delta\text{CO}$ increases as plume concentration increases.

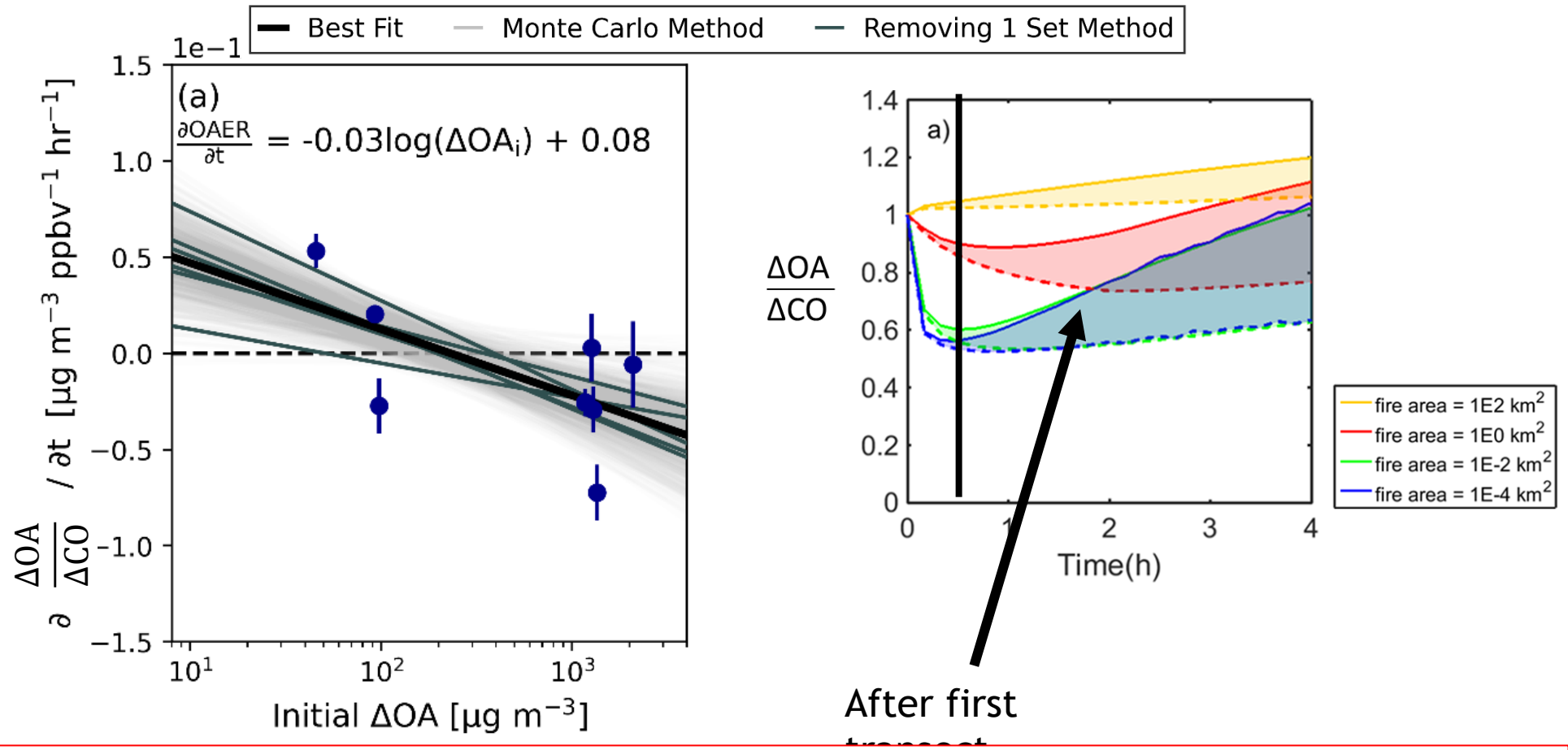


Strong evidence of evaporation before the first transect

Are there systematic differences in the slope (aging) of $\Delta\text{OA}/\Delta\text{CO}$ with plume concentration?



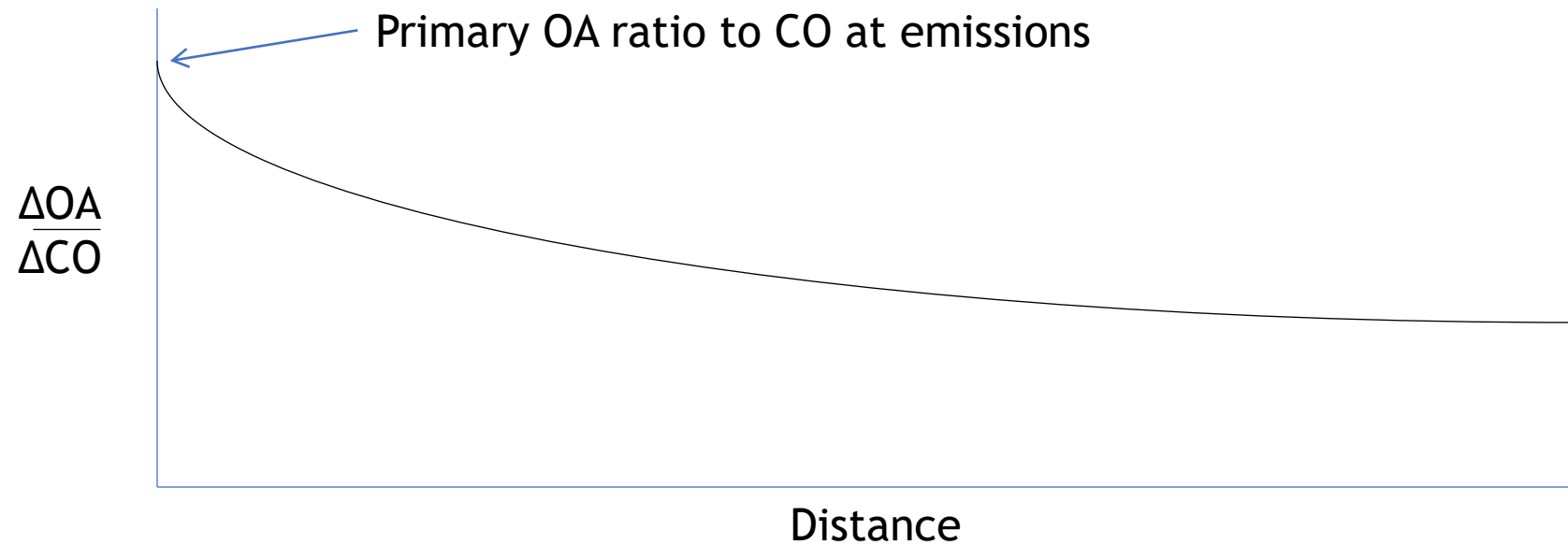
Between ~ 1 -5 hours, OA evaporation is seen more often at higher concentrations in these plumes.



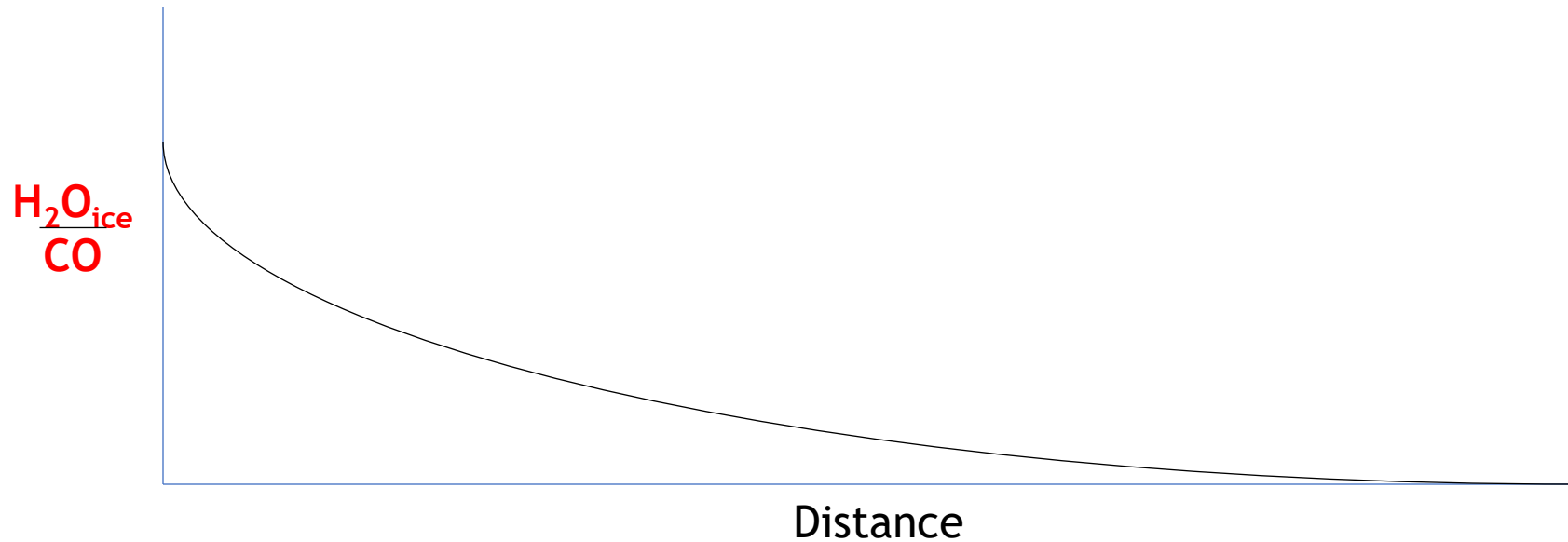
Smaller, more-dilute plumes evaporate prior to the first transect but then gain mass.

Larger, less-dilute plumes evaporate slowly but consistently across several

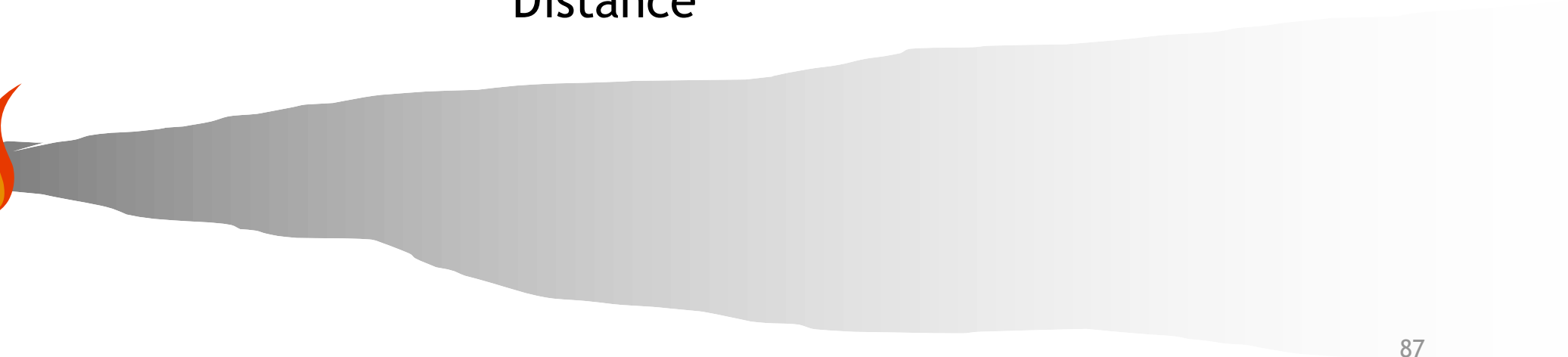
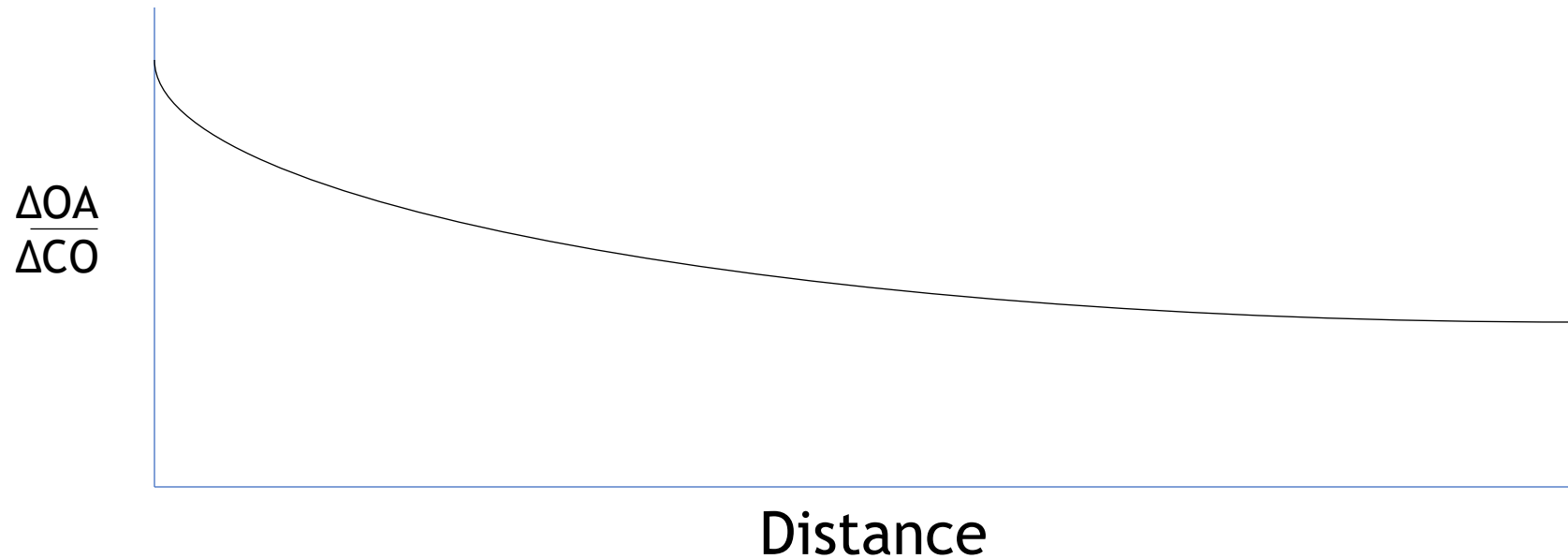
Organic aerosol (OA) is semivolatile: Evaporates as smoke dilutes



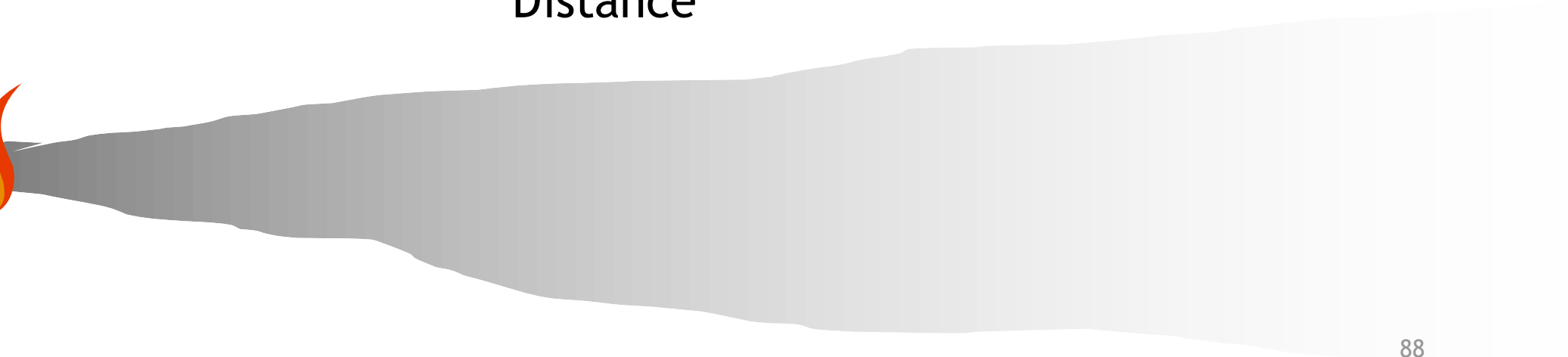
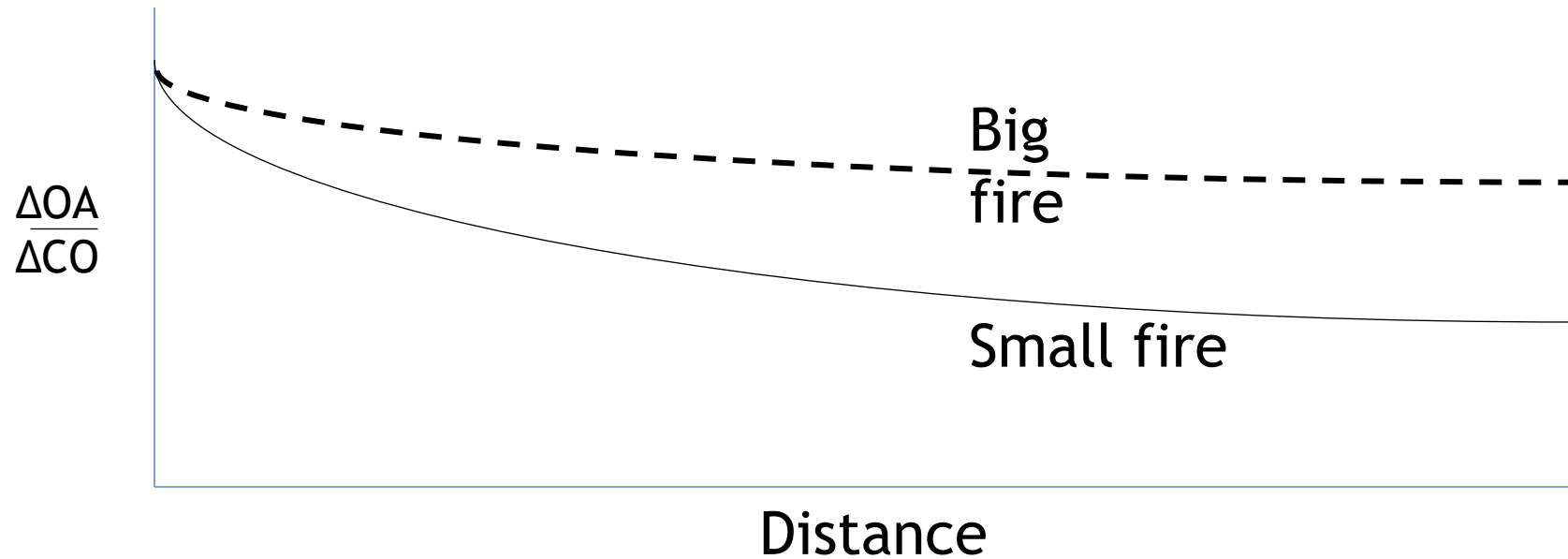
OA evaporates in diluting plumes:
Same phenomena as evaporating contrails



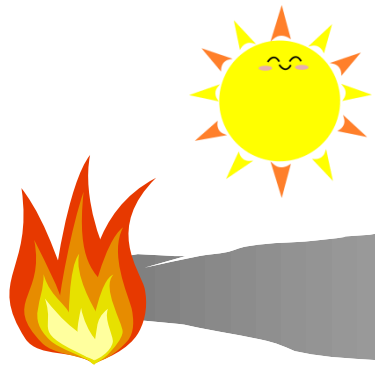
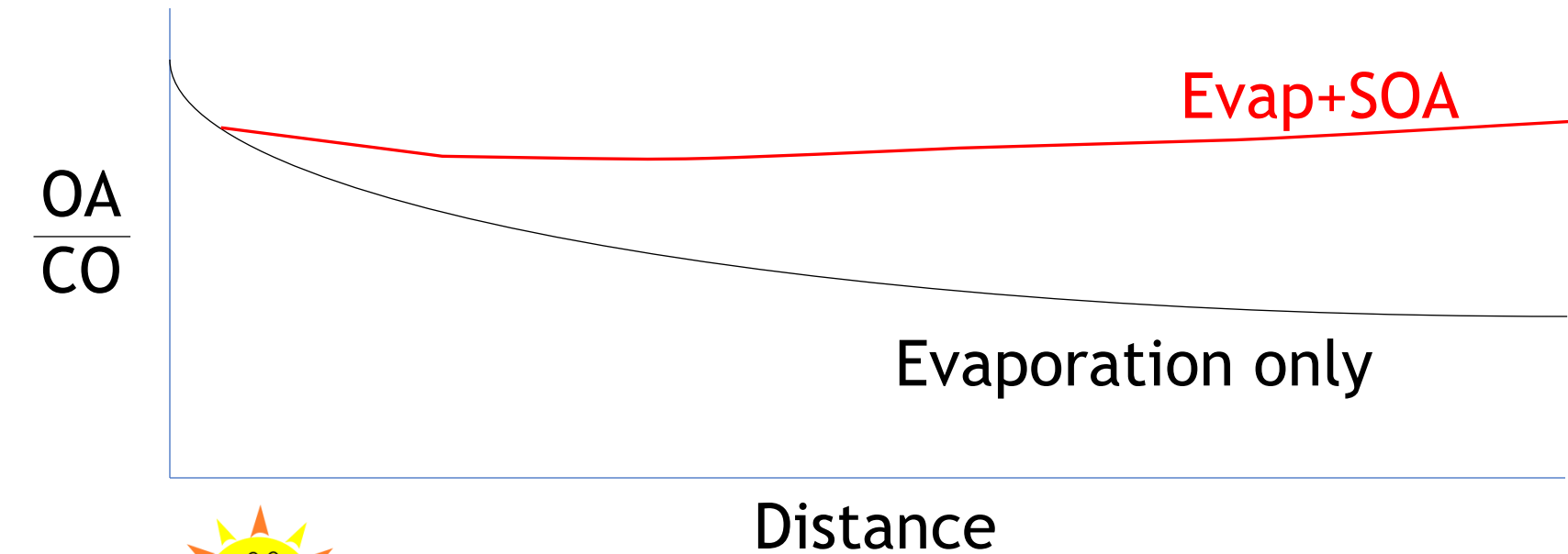
BB aerosol has organic molecules with a range of volatilities:
Not everything evaporates



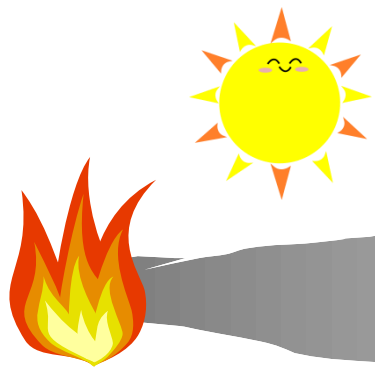
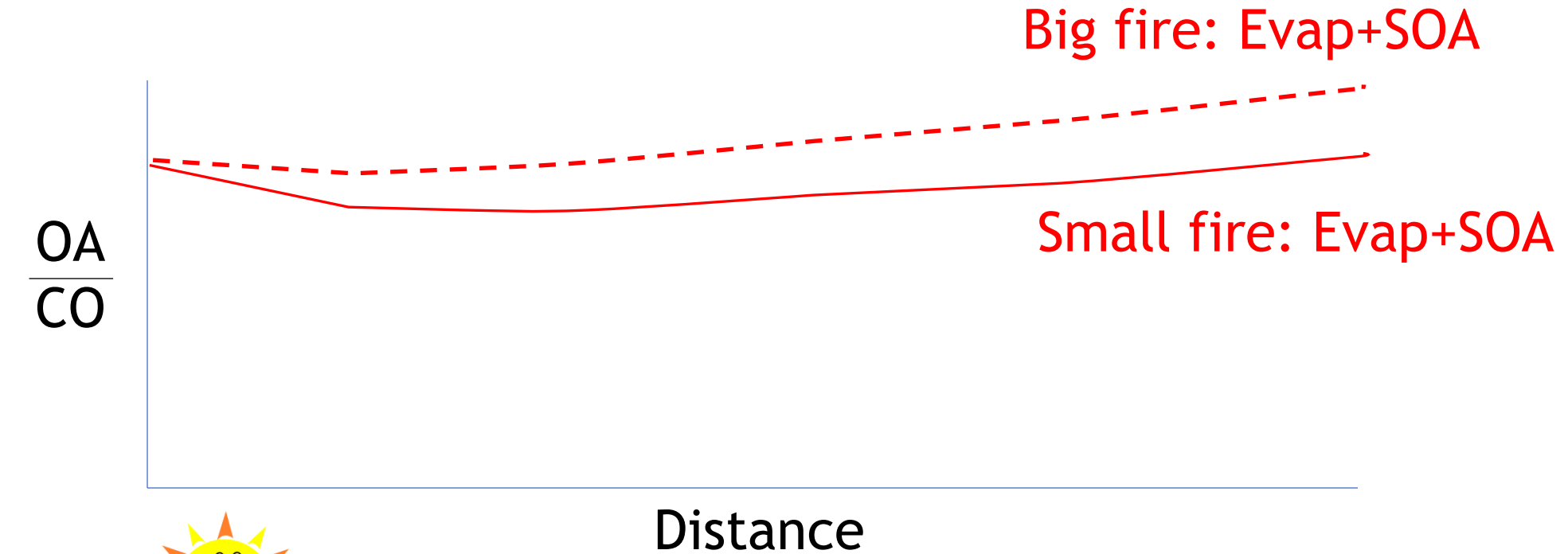
Big fires dilute more slowly than smaller fires
and may evaporate less



Chemistry: Secondary Organic Aerosol (SOA) formation may increase mass

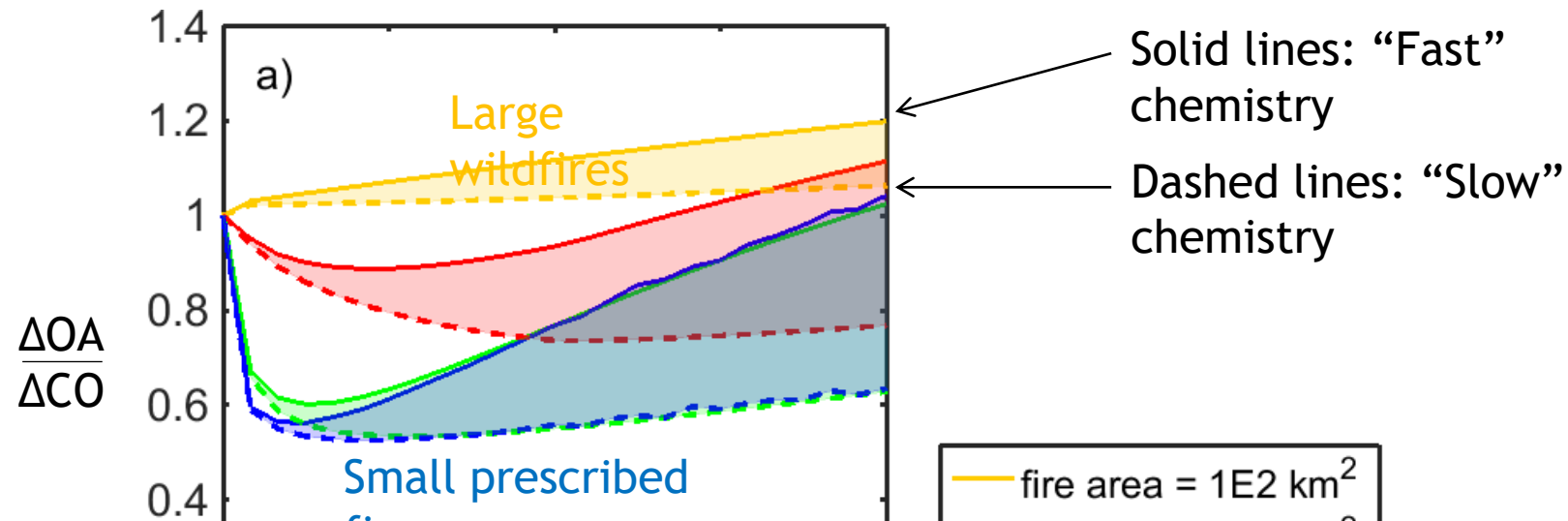


Evaporation vs. SOA formation may differ between large and small fires



Plume simulations with OA evaporation and SOA formation also suggests this result

Little OA change overall



Through investigating several field campaigns:
POA evaporation \approx SOA formation after several hours.

Changes in OA mass within the uncertainties in emissions, so should we neglect OA changes in models (for now)?

