We thank the reviewer for the major and minor comment to improve upon the manuscript. We have attached a revised version of the manuscript and supplementary document to address the comments made by the reviewer. The reviewer comments are in black text, the authors' responses are in red text and the revised text is in "double quotation marks" (with Line number(s) in updated manuscript).

Major Comment - Line 177f

"Aerosols in a highly viscous or a semisolid state are assumed to be phase separated in a core-shell morphology as a model simplification (refer to Section 2.3). This simplification is based on recent observations showing higher than anticipated rebound fractions in phase-separated OA that has been attributed to organic aerosol constituents with viscosities > $10^2\ Pa.s$ (Reid et al., 2018)." (Edited now in current updated manuscript text in Section 2.1)

I still do not understand this logic. Just because phase-separated particles have been seen to rebound, does not mean that every particle whose organic phase by itself would be semi-solid, undergoes phase separation.

We completely agree with the reviewer that it is not necessary that rebounding observed off particles with η_{org} > 10^2 Pa.s refer to phase-separated morphology only, in such solid or semi-solid particles. We have edited the text to reflect that in such a case, both phase-separated and homogenous morphologies are possibilities. We now highlight *PhaseSep2* as the centerpiece of our discussion and edited figures, comparing it to *NonPhaseSep* base simulation. While, *PhaseSep* case is changed to a sensitivity case giving an extreme scenario illustrating what would happen if all semi-solid/solid particles were phase-separated. Of course, *PhaseSep* sensitivity would be overstating phase separation frequencies, but serves the purpose of giving a range of uncertainty in phase separation frequency estimates relative to the *PhaseSep2* scenario. The following text was edited as well to address this comment in the updated manuscript (Lines 176-185):

"Aerosols in a highly viscous or a semisolid state can be homogenous or phase-separated in a core-shell morphology, similar to particles with a liquid-like state. For this study we also ran a sensitivity simulation to see the impact if highly viscous particles were phase separated at all times (refer to Section 2.6). The need for this sensitivity is based on recent observations showing higher than anticipated rebound fractions in OA particles with viscosities > $10^2\ Pa.s$ implying a highly viscous particle that can likely exhibit diffusive limitations in reactive uptake (Reid et al., 2018). These viscous aerosols can be assumed to be in an amorphous solid phase, homogenous or phase separated, but unlike liquid particles they can only dissipate energy by rebounding and criteria governing phase separation in them is not well constrained (Bateman et al., 2015a, 2015b, 2017; Reid et al., 2018; Virtanen et al., 2010)."

The authors also state this in their rebuttal:

"Furthermore, there is little information on the criteria necessary for a particle that is specifically in a semi-solid or glassy state to undergo phase separation. (…) The threshold of viscosity > 100 Pa.s, only indicates the transition in phase state of the 'organic coating' from liquid (< 100 Pa.s) to semisolid or solid (> 100 Pa.s) and should not be confused with dictating phase separation itself." (Was in previous author response not in manuscript)

I also cannot find information about the rebound fractions of phase-separated OA in Reid *et al*. (2018).

Yes, we agree that Reid et al. (2018) gives the rebound fractions for OA particles and they can be phase-separated or homogenous and not explicitly a ‘phase-separated’ OA. We misinterpreted that earlier from Reid et al. (2018) and appreciate the reviewer for pointing this out. We remove the erstwhile argument and edited this argument in the updated manuscript text in Section 2.1 (Lines 176-186; see above) to resolve this.

The authors further state:
“Therefore the 100 Pa.s cut off was chosen as an extreme upper bound scenario for particle morphology to better understand the effects of a core-shell morphology on the heterogeneous chemistry of IEPOX.” (Was in previous author response not in manuscript)
Where does the better understanding come from if the authors over-declare particles as phase-separated (68.7 % vs 42.7 % in updated simulation PhaseSep2)? Does this not simply inflate the modelled effect of phase separation?

We reiterate again by agreeing with the reviewer that: Yes, *PhaseSep* case, which takes this ‘simplification’ (100 Pa.s cut-off) into account is now a sensitivity case. *PhaseSep* definitely inflates the modelled effect of phase separation giving an extreme scenario which might be over-declaring particles as phase separated. But it also provides for a range of uncertainty (an upper bound) in phase separation frequency estimates; given there are still lot of unconstrained parameters in models that can change phase state frequencies in future modeling updates.

Also refer to updated manuscript text (Lines 186-193), after Lines 176-185:

“The specific conditions under which a particle will form a glassy rather than liquid-like organic shell are unclear, but thought to be driven by the same underlying physical properties that drive viscosity. This led to a sensitivity simulation with the consideration that semi-solid or glassy particles would inherently adopt a core-shell morphology. This sensitivity case can be thought of as an upper bound on the frequency of particles separating into core-shell morphology. For the primary phase separation criteria to be broader, it was not assumed that a semisolid state is always phase-separated, and instead the LLPS criteria were applied for conditions that produce a low aerosol water content (refer to Section 2.3). ”

Also refer to the edited text in Section 2.3 that focuses on *PhaseSep2* now as the main realistic scenario as suggested by the reviewer (Lines 258-266):

“Unlike the case of liquid particles, phase separation frequencies in solid or semi-solid particles are not well understood as stated in Section 2.1. We predict LLPS to occur for aerosols with $\eta_{org} \leq$ 100 Pa·s or $T_{g,org}:T$ < 0.8 (Shiraiwa et al., 2017) and when RH $\leq$ separation relative humidity (SRH_{LLPS}) (Bertram et al., 2011; You et al., 2014). Song et al. (2018) suggests that LLPS always happens when $(O:C)_{avg} \leq 0.56$, which we implemented to predict phase separation. When $(O:C)_{avg} > 0.56$, phase separation (or rather LLPS) is predicted based on the conditions specified in Eqs. (8) and (9). As a model simplification, solid- or semi-solid- phase-separated particles (SSPS) occur following the aforementioned LLPS criteria, but when η_{org} > 100 Pa·s or $T_{g,org}$:$T \geq 0.8$, to create a broader scenario referred as *PhaseSep2* (See Table 3)."

Reference added:

"You, Y., Smith, M. L., Song, M., Martin, S. T., and Bertram, A. K.: Liquid–liquid phase separation in atmospherically relevant particles consisting of organic species and inorganic salts, International Reviews in Physical Chemistry, 33, 43-77, doi:10.1080/0144235X.2014.890786, 2014."

Table 3 contents and Section 2.6 (Lines 346-359) were also edited accordingly.

Fig. S6 paints a much more modest picture of the effects of phase separation compared to Fig. 8. It is difficult to compare these two figures though, since the color axis is very different. In my opinion, this publication should focus on scenario PhaseSep2 as the more realistic scenario. Why is PhaseSep2 not used as default scenario?

We concur with the reviewer and we have now edited the manuscript to focus on *PhaseSep2* in our results and discussions as the default scenario, being the broader scenario, and also predicting phase separation frequencies at isoprene-rich Centerville site more accurately. In that process, we have re-plotted figures in high resolution with proper labels and captions.

We have now updated Figures 7 and 8 combining and compressing the information in older Figures S6; while removing older Figure S6. Similarly, remaining figures have been updated as needed to have better resolution and captioning compared to their earlier versions.

Besides the Figures, Result section has also been updated accordingly to focus on *PhaseSep2* case now as the default, more realistic simulation. Result section has been divided into further sub-sections (See Sections 3.1.1 to 3.1.7) to improve readability.

Minor Comment - Lines 560-565

"Wider ηorg ranges in the higher RH bins can be explained by increased diffusivity with higher aerosol liquid water in SOA causing faster mixing times leading to quick changes in composition. Furthermore, these contain the most atmospherically relevant RH ranges for the model simulation period. Therefore, a wide range of particle compositions is expected which also contributes to the wide range of predicted ηorg values."

This is a poorly crafted argument. (i) Why are "quick changes in composition", affecting ηorg? Why is there a time domain of importance? (ii) a larger sample size should not affect quartile position.

We misinterpreted earlier in our explanation and would like to keep the focus on discernable trends in η_{org} and RH as per Figures 4 and 6. Hence we removed this explanation to avoid further confusion and edited the preceding lines in the Updated manuscript as (Lines 580-582):

“The trends in range of modeled η_{org} are the same as in Fig. 4B, with higher mean and quantiles of η_{org} corresponding to lower RH and vice versa for higher RH.”

Supplementary Material for the ACP manuscript "Predicting Secondary Organic Aerosol Phase State and Viscosity and its Effect on Multiphase Chemistry in a Regional Scale Air Quality Model"

Formatted: Justified

Ryan Schmedding[1,2*], Quazi Z. Rasool[1,3*], Yue Zhang[1,6], Havala O. T. Pye [1,4], Haofei Zhang[5], Yuzhi Chen[1], Jason D. Surratt[1], ~~Ben H. Lee[7], ~~Felipe D. Lopez-Hilfiker[7,10], Joel A. Thornton[7], Allen H. Goldstein[8,9] and William Vizuete[1€]

1. Department of Environmental Science and Engineering, The University of North Carolina at Chapel Hill, Chapel Hill, North Carolina, 27516
2. Present Address: Department of Atmospheric and Oceanic Science, McGill University, Montreal, Canada, H3A 2K6
3. Present Address: Pacific Northwest National Laboratory, Richland, WA, 99354
4. ~~National Exposure Research Laboratory, ~~Office of Research and Development, Environmental Protection Agency, Research Triangle Park, Durham, North Carolina, 27709
5. Department of Chemistry, University of California at Riverside, Riverside, California, 92521
6. Aerodyne Research, Inc., Billerica, Massachusetts, 01821
7. Department of Atmospheric Sciences, University of Washington, Seattle, WA 98195
8. Department of Environmental Science, Policy, and Management, University of California, Berkeley, CA 94720
9. Department of Civil and Environmental Engineering, University of California, Berkeley, CA 94720
10. Present address: Tofwerk AG, CH-3600 Thun, Switzerland.

* Shared lead authorship~~Ryan Schmedding[1,2*], Quazi Z. Rasool[1*], Yue Zhang[1,5], Havala O. T. Pye [1,3], Haofei Zhang[4], Yuzhi Chen[1], Jason D. Surratt[1], Ben H. Lee[6], Felipe D. Lopez-Hilfiker[6,9], Joel A. Thornton[6], Allen H. Goldstein[7,8] and William Vizuete[1€]~~

~~1. Department of Environmental Science and Engineering, The University of North Carolina at Chapel Hill, Chapel Hill, North Carolina, 27516~~

~~2. Present Address: Department of Atmospheric and Oceanic Science, McGill University, Montreal, Canada, H3A 2K6~~

~~3. National Exposure Research Laboratory, Office of Research and Development, Environmental Protection Agency, Research Triangle Park, Durham, North Carolina, 27709~~

~~4. Department of Chemistry, University of California at Riverside, Riverside, California, 92521~~

Formatted: Centered

Formatted: Left, Indent: Left: 0", First line: 0", Line spacing: 1.5 lines

~~*5. Aerodyne Research, Inc., Billerica, Massachusetts, 01821*~~

~~*6. Department of Atmospheric Sciences, University of Washington, Seattle, WA 98195*~~

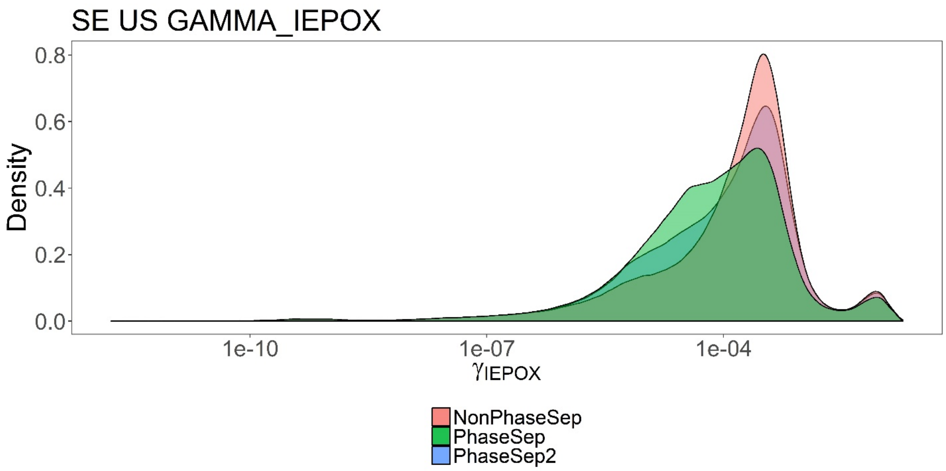
~~*7. Department of Environmental Science, Policy, and Management, University of California, Berkeley, CA 94720*~~

~~*8. Department of Civil and Environmental Engineering, University of California, Berkeley, CA 94720*~~

~~*9. Present address: Tofwerk AG, CH-3600 Thun, Switzerland.*~~

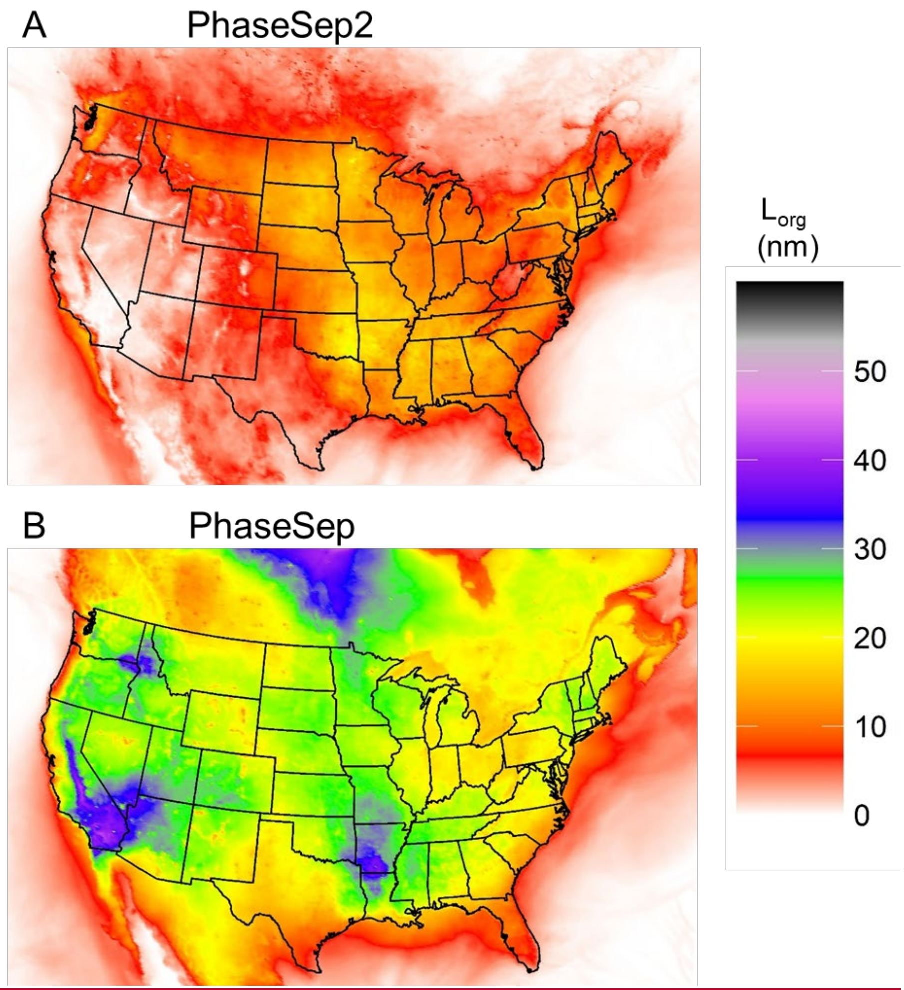
~~** Shared lead authorship*~~

[€] *Corresponding author: e-mail: vizuete@unc.edu; Telephone: +1 919-966-0693; Fax: +1 919-966-7911*



Formatted: Indent: Left: 0"

Figure S1 – For Southeastern United States, Probability distribution of γ_{IEPOX} at the surface level for the *NonPhaseSep* (red), *PhaseSep* (green) and *PhaseSep2* (blue) for SOAS 2013 simulation period.



Formatted: Indent: Left: 0"

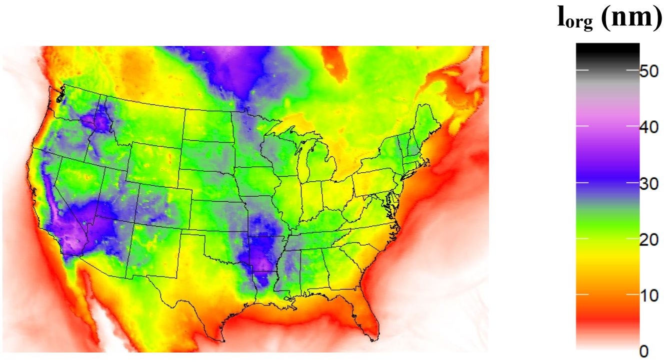
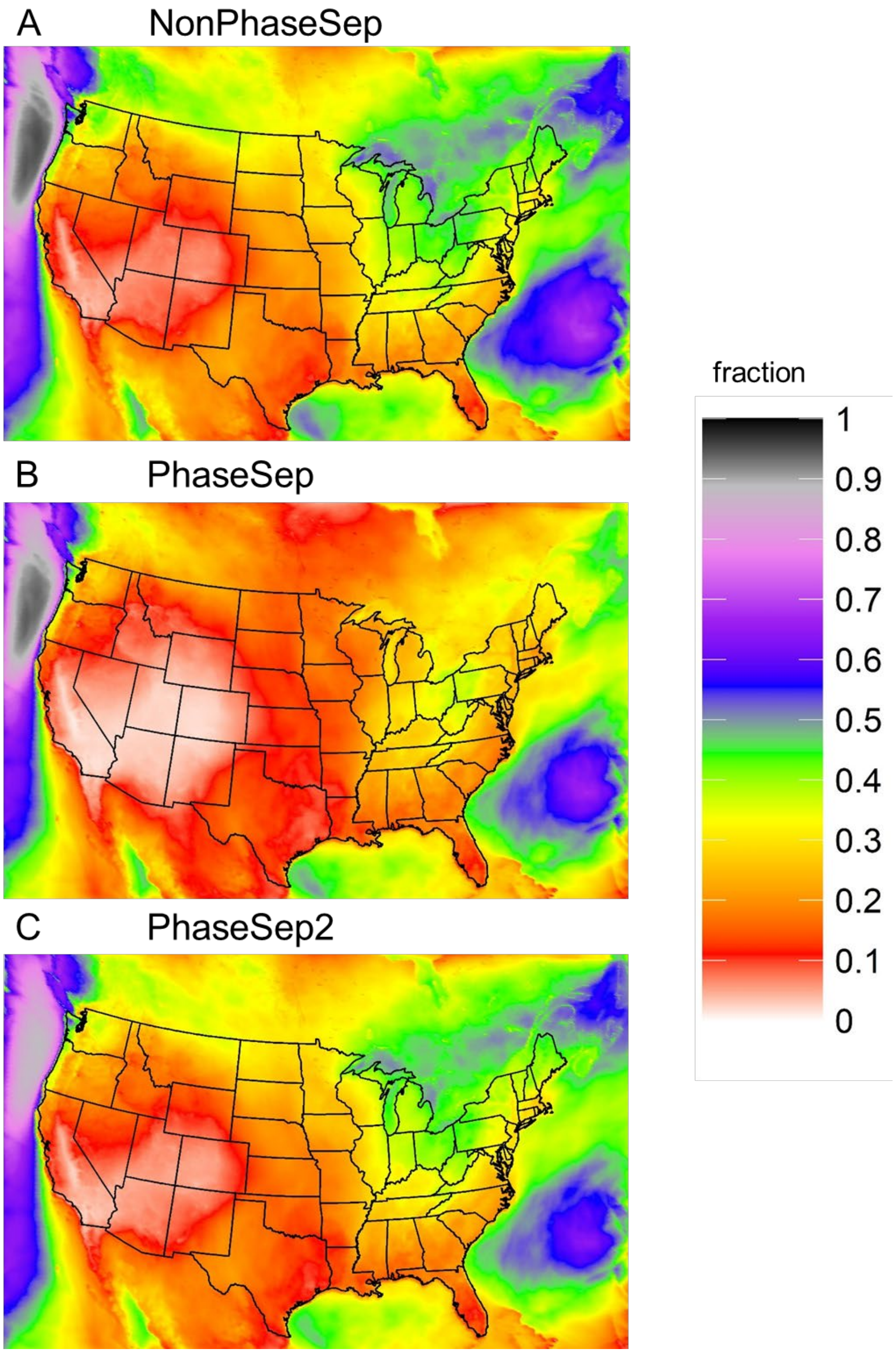


Figure S2 – Average organic coating thickness (l_{org} in nm) at the surface level for **(A)** *PhaseSep2* and **(B)** *PhaseSep* cases for SOAS 2013 simulation period.

Formatted: Font: Bold

Formatted: Indent: Left: 0"


A
NonPhaseSep
B
PhaseSep
C
PhaseSep2
fraction
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0

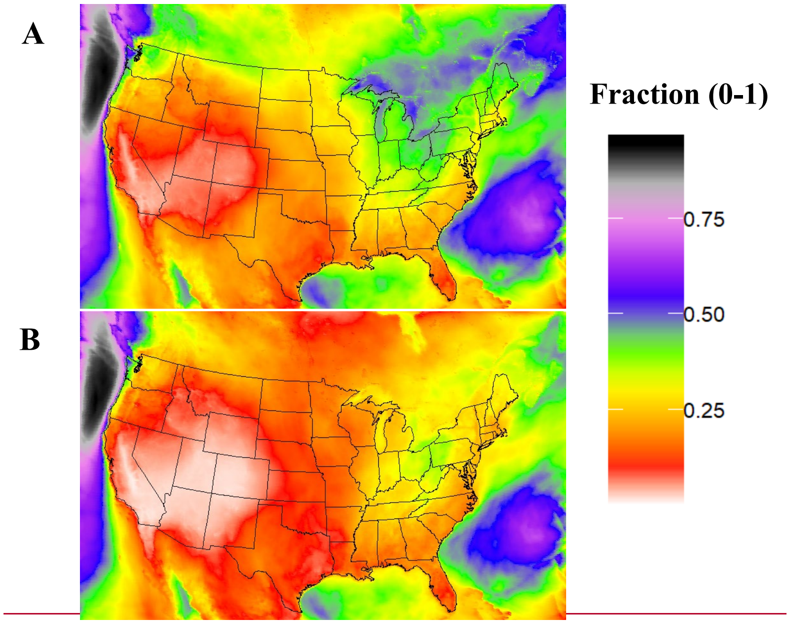


Figure S3 – Average fraction of IEPOX-derived SOA in biogenic SOA mass at the surface level for: **(A)** *NonPhaseSep*, **(B)** *PhaseSep* and (~~B~~C) *PhaseSep2* case for SOAS 2013 simulation period.

Formatted: Font: Not Italic

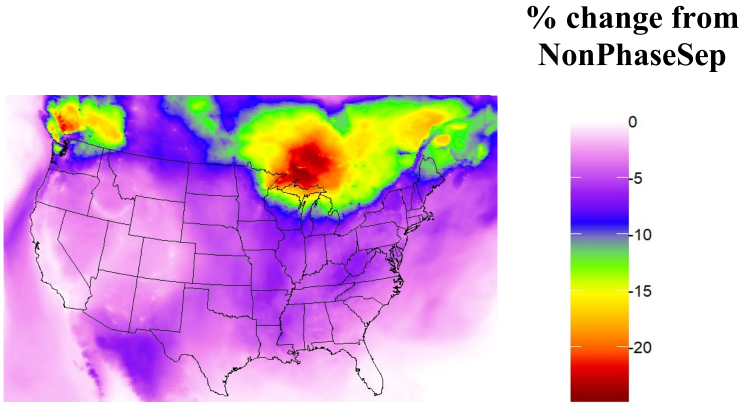


Figure S4 – Spatial map of the mean percent relative change of surface $PM_{2.5}$ organic carbon (OC) mass in *PhaseSep* case relative to the *NonPhaseSep* Simulation.

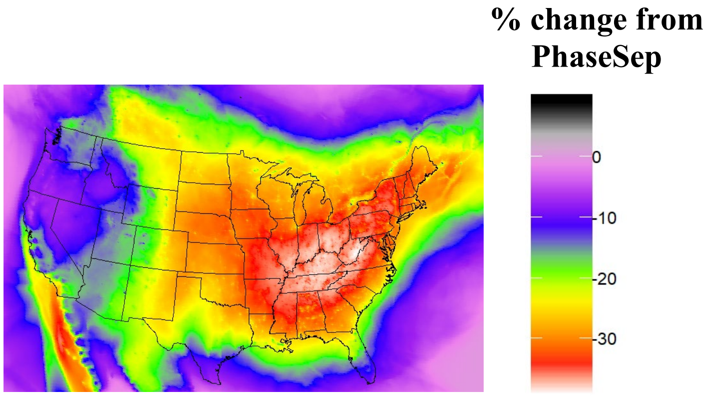
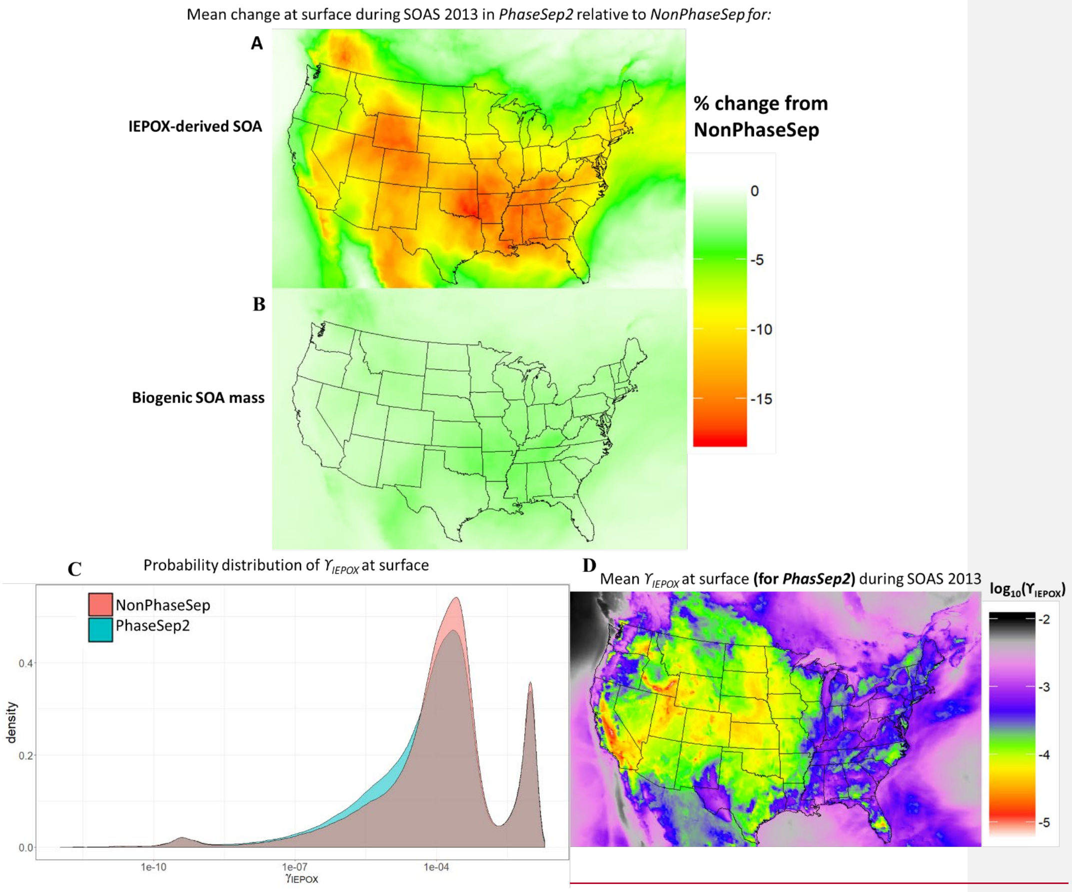


Figure S5 – Spatial map of the mean percent relative change of $PM_{2.5}$ sulfate mass at surface in *Emission Reduction* sensitivity case relative to the *PhaseSep* Simulation.



~~**Figure S6** – Spatial map of the mean percent relative change of **(A)** IEPOX SOA and **(B)** biogenic SOA mass in *PhaseSep2* sensitivity case relative to the *NonPhaseSep* Simulation. **(C)** For Continental US, probability distribution of γ_{IEPOX} at the surface level for the *NonPhaseSep* (red), *PhaseSep2* (green) for SOAS 2013 simulation period. **(D)** Mean γ_{IEPOX} at the surface level for the *PhaseSep2* case for SOAS 2013.~~

Table S1 – Comparison of different simulations on phase separation frequencies (for Continental US) and model performance in isoprene-abundant southeastern United States (rural Centreville, AL and urban Atlanta sites).

Parameter (units)		*PhaseSep*	*PhaseSep2*	*Emissions Reductions*	*HighHorg*
For Continental US~~**LLPS Frequency (%)**~~	**LLPS Frequency (%)**~~**SSPS Frequency (%)**~~	13.7	13.7	13.5	12.5
	SSPS Frequency (%)	54.8	29	57	55.8
NMB (%, compared to SEARCH PM$_{2.5}$ OC)	**Rural Centreville, AL forest site**	-36	-33	-44	-32
	Urban Jefferson Street, Atlanta site	-21	-18	-29	-18

Formatted Table

Formatted: Subscript