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Dear Editor Chuang,

Thanks a lot for volunteering to review our paper. Your invaluable comments are deeply appreciated. The revised paper entitled "Climatic factors contributing to long-term variations of fine dust concentration in the United States" by B. Pu and P. Ginoux are now submitted for consideration for *Atmospheric Chemistry and Physics*. The insightful comments from you and the other reviewer help improved the paper and are sincerely appreciated. Our replies to each reviewer's comments are attached. We also made edits in the manuscript to improve its clarity.

We gratefully appreciate your time and consideration.

Sincerely,

Bing Pu and Paul Ginoux

Interactive comment on “Climatic factors contributing to long-term variations of fine dust concentration in the United States” by Bing Pu and Paul Ginoux

We thank the reviewer for very helpful comments. We reply to your comment (in Italic) below.

This is a well-conceived, well-written, careful, and thorough study that furthers previous work into the investigation of increased dust concentrations in the United States, especially in the Great Plains region. Summertime dust has increased in this region and the authors address the underlying meteorological and large scale climate variability associated with this increase. The work is important for understanding future dust activity in a fairly under-studied region in the U.S. (relative to the Southwest) and provides a needed investigation to address and further our understanding of dust emission and loading. I recommend publication after addressing minor comments below.

Line 101: Technically IMPROVE didn't start sampling every third day until after ~2000 when the network expanded in support of the Regional Haze Rule. Before this it sampled Wed and Sat.

We thank the reviewer for pointing this out. Lines 104-105 are modified to “twice weekly (Wednesday and Saturday; Malm et al. 1994) prior to 2000 and every third day afterwards”.

Line 104: How were data below the minimum detection limit treated?

We did not specifically treat data below the minimum detection limit (MDL). Hand et al. (2017) treated fine iron (Fe) concentration below MDL before 2011 by replacing the original data with 0.5×MDL. Here we used fine soil concentration instead of Fe for analysis. Because fine soil concentration is derived from five different elements (Al, Si, Ca, Fe, and Ti), and MDLs for individual elements are different, it is hard to apply an overall MDL to fine soil. The changes of analytical methods also make MDLs vary with time (Hand et al. 2011).

Line 110: Did the authors apply completeness criteria to compute monthly means?

Monthly data are calculated using all available daily data to increase the coverage of data. We tried the criteria of about 50% completeness (i.e., at least 5 records in each month) for calculating monthly mean, and found about 97% of available data (station×month×year) meet the criteria, and the resulted seasonal mean fine dust data are highly significantly correlated with the unscreened data (Fig. R1). We added lines 117-119 to explain this question.

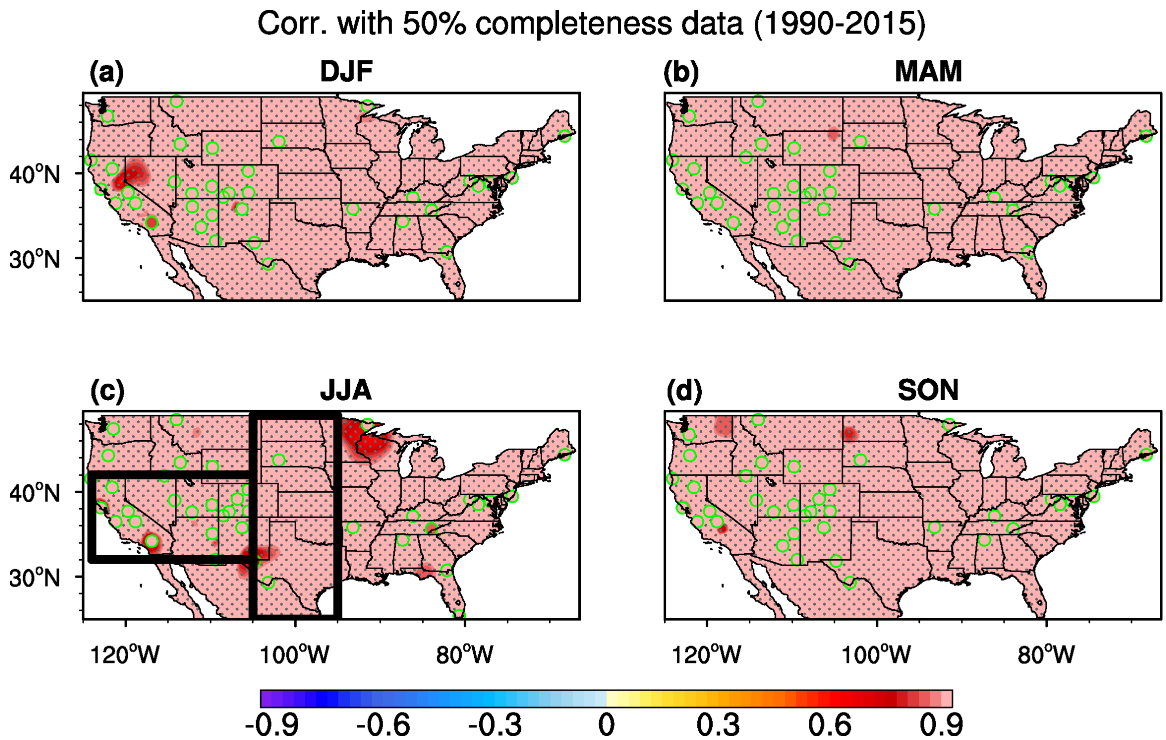

Figure R1. Correlation between seasonal mean screened (with at least 50% completeness) and unscreened fine dust concentrations from 1990 to 2015. Areas significant at the 99% confidence level are dotted in grey. The colored circles show correlations at IMPROVE stations with consecutive records for at least 23 years during 1990-2015. Circles with green outlines denote correlations are significant at the 99% confidence level.

Were there any data requirements for long term site sampling so that the interpolations were not affected by missing sites from year to year? I note on line 209 that 23 consecutive years were required for trend analyses; was this also true for interpolations?

In Fig. 1 we show trends for stations with records for at least 23 years on top of the trend calculated from the interpolated data. When applying the interpolation, all available data are used to increase the coverage of data in each year. And we realize this could introduce uncertainties due to the inconsistent sample sizes from year to year. We added lines 121-124, 227-230 to discuss this issue. Also note, in Fig. 1, most long-term sites show trends similar to those from the interpolated data, with a few exceptions, e.g., over northern Alabama, where interpolated data show positive trends but a long-term site near by shows negative trend. In the analysis for the connection between the Great Plains low-level jet and central Great Plains fine dust (section 3.2) and daily composite analysis (section 3.3) we show analysis for 2002-2015, when the numbers of stations in the region changes little.

Line 209: Add a short description (perhaps earlier- around line 114?) of how trend analyses were performed- OLS? Theil?.

We used least squares linear trend analysis. Lines 120-121 are added to explain the method.

Line 209, Figure 1: Can the authors comment on the interpolated trends over regions with no sites? Were these calculated using sites that were not consecutively sampling? (e.g., over the central US)? Including some sites for only some years could obviously bias the spatial variability in the trends in the interpolated values.

The trends (shading) shown in Fig. 1 are from interpolated data that are not necessarily consecutively sampled. Lines 239-244 are added to clarify this issue: "One thing we want point out here is that most of the stations in the Great Plains have records shorter than 15 years, with only three stations having records for more than 25 years (Fig. S1), therefore the positive trends here are combinations of interpolated information from nearby stations in the early period (before ~2002) and more reliable data largely from local stations in the late period. " We agree cautions are needed to treat results in the early period, that's why we mainly used the data from 2002 to 2015 in the analysis focused on the central Great Plains (sections 3.2 and 3.3). The similar relationships between fine dust in the central Great Plains, CIN, and the low-level jet during 2002-2015 and 1990-2015 to some extent suggest that the time series of fine dust are coherent.

Line 213: How is the "climatological" value computed? (Over which years)

It is averaged between 1990 and 2015 (see figure caption of Fig. S2).

Line 317: Change "Figure 3a show" to "Figure 3a shows"

Done.

Line 449: Change "transports" to "transport"

Done.

Line 464: Figure 7 and 8: In 3 of the cases, but especially 7/2/12, concentrations near Everglades increase also, which might suggest African dust influence especially with the 4km dust level? Did authors investigate the elemental composition on these days to rule out that influence?

The reviewer found the increases of fine dust concentration in the Everglades in Figs. 7-8 may suggest an influence of African dust. Although our analysis in section 3.2 shows that on seasonal scale the influence of African dust is largely over the southeastern U.S. (lines 433-443), it is possible that African dust is transported to higher levels over the central Great Plains in some days. This is confirmed by a quick check on the elemental composition (Figure R2).

Figure R2 shows the anomalies (with reference to 2002-2015 JJA mean) of three elemental compositions, fine aluminum (Al), calcium (Ca), and iron (Fe) for July 2nd, 2011 (top) and July 2nd, 2012 (bottom). We found an anomalous increase of Al and a decrease of Ca in the central Great Plains region. Such a high-Al and low-Ca feature is similar to African dust (e.g., Hand et al. 2017). In both cases, changes of Al and Ca in Virgin Islands National Park (VIIS1, 18.3°N, 64.8°W) are weaker than that over the central Great Plains. This suggests there may be contributions from local or Mexican sources as well.

We added lines 512-515:"The anomalous high fine dust concentration in Everglades National Park (Figs. 7-8) in three of the four cases shown here suggest that there may be a contribution from African dust in these days, but further analysis are needed to clarify the magnitude of its contribution."

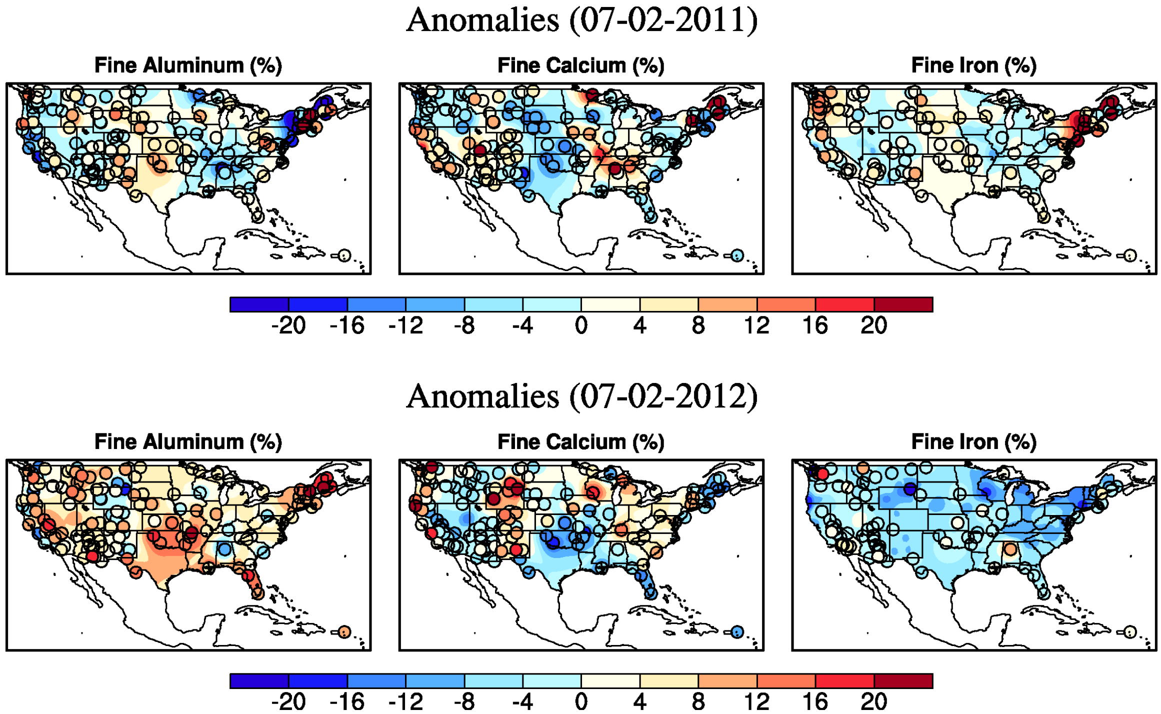

Figure R2. Anomalies (with reference to 2002-2015 JJA mean) of elemental composition (percentage): fine aluminum (Al), fine calcium (Ca), and fine iron (Fe) for July 2nd, 2011 (top) and July 2nd, 2012 (bottom). Shadings are gridded values, while colored dots show values of individual stations.

Line 525: Change "prevent" to "prevents"

Done.

Line 1031: Figure 5 Caption: can the authors add a statement describing the black box in figure 5b?

Done. We added "Black box denotes the CGP region."

Line 1049, Figure 6 Caption: Change "data is" to "data are"

Done.

Line 1082, Figure 7 Caption: can the authors add a statement describing the black box in figure 7?

Done. We added "Black boxes denote the CGP region."

Line 1130, Line 1137 Figure 9 and 11 Caption: Same comment as above. Supplemental Information, Figure S5: Change "liner fitting" to "linear fitting"

Done.

Reference:

Hand, Copeland, S. A., Day, D. E., Dillner, A. M., Indresand, H., Malm, W. C., McDade, C. E., Moore, C. T., Pitchford, M. L., Schichtel, B. A., and Watson, J. G. (2011), IMPROVE (Interagency Monitoring of Protected Visual Environments): Spatial and seasonal patterns and temporal variability of haze and its constituents in the United States, Rep. V, Coop. Inst. For Res. In the Atmos., Fort Collins, Colo. [Available at http://vista.cira.colostate.edu/Improve/spatial-and-seasonal-patterns-and-temporal-variability-of-haze-and-its-constituents-in-the-united-states-report-v-june-2011/].

Review of "Climatic factors contributing to long-term variations of fine dust concentration in the United States"
Authors: Bing Pu and Paul Ginoux
Reviewer: Patrick Chuang

We thank Professor Chuang for very helpful comments. We reply to each comment (in Italic) below.

The manuscript examines the meteorological factors responsible for surface fine dust (deduced from IMPROVE network measurements). They build upon earlier work that shows that primary factors affecting dust optical depth include precipitation (which leads to wet scavenging), surface bareness and wind speed (which are associated with dust emissions). This work focuses specifically on PM2.5 dust, and examines the times of year and locations within the US where CIN and CAPE add predictive power to fine dust, and the larger meteorological context that leads to these new factors being important. Overall, the manuscript represents a useful contribution to our knowledge of dust aerosol in the US. Before it is appropriate for publication, however, the figures need to be improved, and some methodological issues clarified and/or fixed.

Main comments:
** The title should read "surface fine dust" since this is the focus of the observations. You should emphasize this once in a while within the text and figures as well.*

We modified the title to "Climatic factors contributing to long-term variations of surface fine dust concentration in the United States". We also emphasize that IMPROVE measures surface fine dust in the text.

** You mention pattern correlation but there is no description of the method. Please add a clear description of what is done.*

Thanks for pointing this out. We add in text (lines 218-219) that we used centered pattern correlation and to save space we provided one reference where the methods to calculate centered pattern correlation is detailed in that paper.

** Why is the reconstructed pattern called "REG"? This is unintuitive.*

We used "Reg" because we reconstruct surface fine dust concentration using a multiple linear regression model.

** Section 3.3.1: your usage of various two-variable regressions seems like a poor choice of methodology. Is there some reason why you don't use multi-variate regression as in the rest of the paper?*

The reviewer found it is odd to use two-variable regression instead of multiple linear regression to understand the connection between CIN and other variables. There may be some misunderstanding. We added lines 454-456 to better clarify our purpose. In this section, we try to understand physically how CIN is connected with surface fine dust concentration, by exploring a variable both connected with CIN and fine dust concentration, i.e.., U^*. Then we further explain how U^* is connected with CIN and found both variables are significantly correlated with surface temperature and T_{700}-T_{dp}. We used two-variable correlation to establish these connections.

Multiple linear regression is used in other sections because we want to identify relative contribution of each controlling factor to fine dust variability. It is true that we can apply multiple regression to examine the relative contribution of individual variables such as surface temperature, friction velocity, surface dryness to the variability of CIN, but that's not the purpose of this analysis.

Also, the lines 450 to 452 basically state that the entire section up to this point is not supported by the NARR reanalysis. If this is so, then is your hypothesis really worth mentioning? To the reader, it feels like I spent a bunch of time working hard to understand a complicated set of figures, tables and text, and in the end, it may be completely spurious which makes me feel like I just wasted my time. Please delete this part if you can't get NARR to tell a consistent story.

We realized that the original text in section 3.3.1 was confusing, so we added lines 454-456 and modified lines 467-515 to increase the clarity of this section. Lines 450-452 in original text only refer to the hypothetical connection between vertical velocity at 850 hPa (w850) and U^* during dusty days (now lines 488-496), not all the analysis before this paragraph. The connection shown in Table 1 and Figure 6 are valid in both ERA-Interim and NARR (now line 486-487).

** I found the examples using CALIOP data (Figs 7 and 8, and text from lines 453 to 467) unconvincing. Unless there was an objective choice for these cases, these are snapshots that could pretty much mean anything. I'm sure one could find two cases that represent the opposite of what you want to show. Please delete. It's a long enough manuscript as it is. I don't think this part adds anything.*

We used CALIOP products for case study and had no intent to establish any statistical conclusion based on case study, as such a negative connection between CIN and fine dust concentration is already shown on the interannual time scale in section 3.1, and will be shown on the daily scale in section 3.3.2. The purpose of Figs. 7-8 is to demonstrate one hypothesis that strong convective inhibition prevents convective mixing between dusty boundary air and clean air above, thus increase surface fine dust concentration. These cases were selected when surface fine dust concentration was greater than one standard deviation.

** Your analysis ignores advection, as you state in your methods. Given the spatial scale of your analysis regions, expected transport patterns and the lifetime of fine dust, can you defend doing so? There are certainly places in the world where there are times of year (say, Korea in the spring) when dust is almost entirely due to advection. I note that surface wind speed, precipitation, CIN and CAPE could all plausibly correlate with advective transport.*

In this study we focus on what local or regional factors may affect surface fine dust concentration. We used surface fine dust concentration from IMPROVE stations, which may contain a portion of transported dust, depending on the location of site. Even though, we can still use regression or correlation to examine how station recorded fine dust concentration (both local and transported) is related to precipitation, wind speed, CIN or CAPE. We mentioned in text (lines 269-271) that in spring Asian dust has been found over the western U.S., while in summer African dust can influence the southeastern U.S. In lines 425-443, we discussed whether African dust may affect surface fine dust concentration over the central Great Plains. To fully understand the relative contribution of transported dust to total fine dust concentration requires a climate model that well captures the dust emission and transport processes or/and additional data (e.g., mineralogy, isotopic compositions), which is beyond the scope of this paper.

The significant correlations between surface fine dust concentration and local controlling factors (e.g., precipitation in the southwestern U.S. in spring and CIN in the central Great Plains in summer) and the trends of these controlling factors (i.e., precipitation and CIN) also indicate that the positive trends of fine dust are largely related to changes of these controlling factors, not caused by the changed fraction of transported dust.

** For all figures where you want the reader to focus on a specific region (Figs 1, 2, 3, 4 and 5), you MUST put the boxes into each panel, not just one of them. Otherwise it is impossible to compare the boxes (I just about went crazy trying to do so). Note that you do have boxes in all panels in in Figs 9 and 11. Also, please consider using different kinds of boxes to outline the Southwest, GP and CGP (say, solid, dashed and dotted or black, blue and green). That way when I see a box in a new figure, the color or line used to draw the box tells me exactly which box it is.*

We've followed the suggestion to add boxes in each plot in Figs. 1-5. We now changed the boundaries of southwestern box to solid black line, the GP box to dashed black line, and CGP box to solid black line. We tried to use a different color for the CGP box, but since the box appears in Figs. 4, 5, 7, 8, 9 and 11, and all these figures used very different colors for shading or contours, it turned out that black is the best choice for the color of the box. We also introduced each of these boxes in corresponding figure captions.

** Overall the text can be a bit tough to follow since your analysis has three regions and four seasons = 12 choices. Exactly which of these 12 you are discussing at any one time keeps changing. I don't have any specific suggestions, but I do recommend that you think about editing the paper to help the reader more easily keep track of exactly what your analysis refers to.*

We modified the whole text to make it easier to follow. In fact, besides Figs. 1-2, we only focused on the fine dust variation in the southwestern U.S. in spring and over the central Great Plains in summer, in other words, two choices. We added in line 352-353 "Specifically, we focus on the positive trends of surface fine dust in the southwestern U.S. in spring and over the CGP in summer (Fig. 1b and c)." to clarify this.

There are minor comments (and some repeats of the above comments) in the attached marked-up PDF file. Please ignore any comments that are redundant with the above.
Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2017-821/acp-2017-821-RC2-supplement.pdf

Thanks for the detailed comments. We rely to each of your comments in the following:

Line 29: "station data from", Must emphasize that this surface fine dust.
We added "surface fine dust" in Line 28.

Line 32-33: "including convective parameters such as convective inhibition (CIN) and convective available potential energy (CAPE)". What is the underlying process? Convection?
We added in lines 33-34: "...that reveal the stability of the atmosphere...".

Line 36, "increasing"
Changed to "increase"

Line 37, "enhancing", "weakening"
Changed to "enhanced" and "weakened"

Line 38, "related to", due to or something stronger.
We changed " related to" to "caused by".

Line 39, "positive trend", positive trend of what property of the jet?
We changed "positive trend" to "strengthening"

Line 41, "its connection", less ambiguous?
We modified lines 42-43 to "via its positive correlation with surface winds and negative correlation with CIN."

Line 51: "and causing two parallel statements are "degrading" and "causing")"
We changed the line to "by degrading visibility and causing traffic accidents..."

Line 52: "diseases" -> disease
Done.

Line 56: "...largely anthropogenic, in associate..." comma splice
We modified the sentence to "Major dust sources in the United States are located over the western and the central U.S. While several deserts are located over the western U.S., e.g., the Mojave, Sonoran, and northern Chihuahuan deserts, over the central U.S. the dust sources are largely anthropogenic, in association with agriculture activities..."

Line 103, "longer than"-> have data extending back more than 10 years.
Done.

Line 109, "(Hand et al., 2011;2012; 2016;2017)." A lot of your multiple references are missing spaces. Please fix all occurrences.
Done.

Line 115, "several..." ->three, since this is the # of boxes you use in this study. Also, how are these regions identified? What happens in areas that are NOT in these identified regions?

We changed "several" to "two". We used same averaging area of the southwestern U.S. and the Great Plains as by Pu and Ginoux (2017), based largely on the geographical locations of dusty regions in the U.S. We added in lines 128-130 "In later analysis, we also focused over the central Great Plains (CGP for short; 32°-40°N, 95°-102°W) in summer to examine the positive trend of fine dust in the region. " In this study we mainly focus on these regions. We displayed the trend of fine dust in the U.S. in Fig. 1, including those areas outside the averaging box.

Line 118: "central Great Plains", CGP for short; ...) Explain why this box was chosen (I assume it's from Fig. 4a).

We added "CGP for short". We chose the region based on Fig. 1c and 4a. We modified in lines 128-130 to clarify this.

Line 128: "backscatter from Level 1 product and depolarization ratio from Level 2 product are used." grammar needs fixing here.

We modified the sentence to "..., both the daily 532 nm total attenuated backscatter from Level 1 product and depolarization ratio from Level 2 product are used."

Line 129: "Depolarization" -> The depolarization ratio

Done.

Line 130, "here"-> here a threshold of 0.2 is used...

Done.

Line 136-137 "and is suitable to study long-term connections between fine dust and precipitation." In what way is it suitable?

We changed lines 147-148 to "Its relative high-resolution and long records are suitable to study long-term connections between fine dust and precipitation".

Line 158, "precipitation", This is a second precip record that is mentioned. Which is actually used, and why?

We added in lines 172-173 "Here daily precipitation is used for daily composite analysis in section 3.3.2." Because PRECL only provides monthly data, for daily composite we used NARR precipitation.

Line 167, "P_{LFC}". Variables must be in italics.

Done.

Line 169, remove "By definition"

Done.

Line 187-188, "surface variable", which ones?

We added in line 202 "(such as surface turbulent stress)"

Lines 189-190:"surface turbulence stress, vertical and horizontal winds, air temperature, and specific humidity..." From what I can tell, only Fig. 11 uses ERA-Interim data. Are all of these used in this figure?

No, not only in Fig. 11. Variables such as vertical velocity, air temperature, specific humidity are used in Fig. 10, as mentioned in figure caption.

Line 197-198: "Here all data are interpolated to a 1° by 1° grid for the regression analysis." Not sure what this means. May need more explanation of the method. Is the regression local? If so, what about advection? Or is it for the entire domain (western or central US)?

We added lines 211-214: "Since multiple controlling factors and gridded surface fine dust have different horizontal resolutions, for the regression analysis we first interpolated all variables to a 1° by 1° grid, then apply the regression at each grid point" We did not separate advection and local fine dust in regression analysis (also see our reply above).

Line 202: "pattern correlation". Can you explain this better? For example, what time scale are the patterns averaged over before running a correlation?

We cited one paper to explain the methods to calculate pattern correlation (line 218-219). Here we compare the patterns of trends (from 1990-2015) calculated from IMPROVE surface fine dust with those calculated from reconstructed surface fine dust.

Line 208: "gridded data (shading)". Why isn't the analysis applied only to the stippled areas where trends are statistically significant?

In Fig. 1 we want to show the trend of fine dust in all seasons. Later, we only focus on two regions with significant positive trend: the southwestern U.S. in spring and the central Great Plains in summer.

Line 243-244: "contribution from Asian dust in spring" This is pretty ambiguous. Is transport a significant contributor?

The relative contribution of Asian dust on DOD or fine dust over the western U.S. is non-conclusive. Hand et al. (2016) also used IMPROVE surface fine dust data and suggested the influence of Asian dust over the northwestern United States is low. To fully address this question needs both modeling study and chemical composition analysis from observations, which is beyond the scope of this study.

Line 248: "variations" On what time scale? This needs to be made clear here and in the figure caption.

We added "on interannual time scale from 1990-2015" in lines 273-274. In Fig. 2 caption, we mentioned that the regression is calculated for 1990-2015.

Line 280: "pattern correlation" I'm assuming these are spatial correlations? This is poorly described. What area is used for the pattern correlation? It seems to be the two boxes from Fig. 1. Do you consider parts of the boxes where the trend is not statistically significant?

Yes, it is spatial correlation for all the area within the boxes. These lines are removed now. We explained pattern correlation in lines 218-219.

Line 340: increasing->increase

Done.

Line 372: "jet index in summer" So does Fig 5a plots summer index vs summer-averaged fine dust or annually averaged fine dust? This is unclear and needs to be clarified in the text and figure caption. Same thing in the maps.

Both fine dust and jet index are calculated in summer as motioned in the caption "in JJA from 1990 to 2015" and in text (line 409) "Figure 5a shows the time series of the jet index in summer". We added "in summer" in line 412 in case the reader is confused. We also plotted the area of jet core (the averaging area for the jet index) in the revised figure (Figs. 5b-c; deep pink boxes).

Line 374: "jet core (25°-35°N, 97°-102°W)". Please mark this core region on the maps in Fig. 5. It's hard to visualize the location of the jet otherwise.

Done.

Line 375 "The jet index is significantly positively correlated with fine dust concentration in the CGP". Why not in the entire jet region? What happens in the part of this core that is outside of the CGP region?

Some part of the jet core is out site of the CGP region as shown in the figure. We chose to use the wind speed at jet core area to represent the strength of the jet, which is widely used in studying the interannual variations of this low-level jet (e.g., Weaver and Nigam 2008). This dose not mean winds in the jet region out side the jet core area are unrelated to the jet core. If we calculate the correlation between the jet index and 900 hPa meridional wind, we would find winds outside the core area in the jet region also significantly positively correlated with wind in the core area.

Line 383: "local temperature" At what altitudes? Warming at surface vs 700 hPa are obviously different in the impact on stability.

The jet core is located below 850 hPa, thus the temperature advection is largely near surface within the boundary layer. But the change of vertical stability may also depend on the surface temperature, so we decided to remove the part "and increasing local temperature via northward warm temperature advection (e.g., Walters and Winkler, 2001; Song et al., 2005; Zhu and Liang, 2013)."

Line 388: "Dust from Africa..." This sentence seems incongruous until you read the next one. I think it should mention the jet explicitly.

Previous studies have not specifically pointed out that the transport of African dust is due to the low-level jet. So we did not mentioned it. We modified lines 427-428 to make the sentence clearer: "Can the intensified jet transport more African dust and thus contribute to the increase of fine dust in the CGP?" .

Line 417: "CIN and friction velocity". Are these calculated from the daily mean *conditions, or from higher frequency values, and then averaged for the day?*

As motioned in line 176, daily CIN from the NARR is used. And in line 203-205 we motioned 3-hourly surface turbulence stress from the ERA-Interim is used to calculate daily friction velocity.

Line 427: "significantly", I think you need to be clear that while the relationship is statistically significant, U only explains something like 15 to 25% of the variance in CIN. So it's far from a majority of the explanation for how CIN changes.*

As shown in Table 1 the correlation between U^* and CIN ranges from -0.44 to -0.54, which indicates that U^* explains 19%-29% variances of CIN, not very high. The point here is not to find which variable dominantly control the variations of CIN, but to understand how CIN is physically related to surface fine dust concentration. It is well known that U^* is related to dust emission. Here we examine the connection between U^* and CIN to explore if the connection between CIN and surface fine dust concentration is due to CIN's connection with U^*.

Line 437-438: "indicating that CIN is connected with U^* via its connection with near surface temperature." I'm not sure you can draw this conclusion. With small fractions of variances explained in these correlations, I don't think this necessarily follows.

The reviewer found that U^* only explains a small portion of the variance of CIN, thus the connection between CIN, U^*, and surface temperature may not be valid. We did not intend to draw any conclusion such as U^* or surface temperature drive the variation of CIN, which requires a higher fraction of variances being explained by U^* or surface temperature. Here based on the correlations shown in Table 1 we simply conclude that both CIN and U^* have significant correlations with surface temperature, thus the significant negative correlation between CIN and U^* may be due to their mutual connection with surface temperature.

Line 440: "Similarly". Is there a reason you don't use a multivariate regression similar to what you've already done in Fig 2? The way this is done isn't particularly convincing, I don't think.

As we explained before, the purpose of this section is not to identify relative contribution of individual factors to the variability of CIN. We tried to explain the physical connection between CIN and surface fine dust connection and found CIN is significant correlated with U^*. Since both U^* and CIN are significantly correlated with surface temperature, we suggest that CIN is connected with U^* via their mutual connections with surface temperature.

Line 451-452: "...suggesting further investigation on this mechanism is needed." If this proposed mechanism isn't borne out in the presumably more refined reanalysis, then why is it even being discussed?

As we replied above, this line only refers to the hypothesis to explain the correlation between U^* and CIN in dusty days (lines 488-496), not the whole analysis in section 3.3.1. As shown in Table 2, the connections among U^*, vertical velocity at 850 hPa (w850), and CIN are valid. These correlations are not valid if using w850 from the NARR, and it's probably because U^* itself is from the ERA-Interim, thus is more consistent with the variations of w850 in the ERA-Interim. This paragraph provides a possible explanation for the significant correlation between CIN and U^* in dusty days, and we prefer to keep it.

Line 453-454: "Despite the connection between CIN and surface variables, the possible mechanism that strong inhibition prevents dilution is also examined." I don't see why these are mutually exclusive ideas. To me they seem perfectly compatible.

We did not mean to suggest that the two mechanisms are against each other. To avoid misunderstanding, we changed the sentence to "In addition to the connection between CIN and surface variables, the possible mechanism that strong inhibition prevents dilution is also examined."

Line 460: "...with CIN anomaly greater than one standard deviation." Again, is this anomaly a daily-averaged value, or specific to nighttime, or to the daytime period prior to the overpass or...?

We added in line 504 and figure caption to clarify that this is daily anomaly.

Line 467: "Nonetheless, more cases are needed to further verify this mechanism." These cases don't add much to the main story of this manuscript. Unless you have an objective way of identifying these days, then this could be interpreted as working backwards from the answer you are looking for. I suggest deleting it.

Thanks for the suggestion. We used case study to provide an explanation on how CIN is related to surface fine dust concentration, not to establish any statistical connection between the two. Based on the analysis in section 3.1, we already found that CIN is significantly negatively correlated fine dust on the interannual time scale. We brought up a hypothesis that "A stable atmosphere prevents deep moist convection, which reduces the chance of scavenging by precipitation, and also likely prevents dilution of fine dust concentration in the boundary layer with the clean air above through convective mixing" at the end of section 3.1. In section 3.3.1 we used case study to demonstrate this mechanism. We selected days when fine dust anomaly is above one standard deviation, while CIN has strongly negative or positive anomalies to illustrate the effect of prohibited dilution when the convection inhibition is high. We do not have enough cases to do statistically test, because CALIOP track dose not always pass the CGP in those dusty days. Later, the daily composite analysis (Fig. 9) showed that in dusty days CIN is anomalously negative (significant at the 95% confidence level).

Climatic factors contributing to long-term variations of surface fine dust concentration in the United States

Bing Pu[1,2] and Paul Ginoux[2]

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Abstract. High concentration of dust particles can cause respiratory problems and increase non-accidental mortality. Studies found fine dust (with aerodynamic diameter less than 2.5 microns) is an important component of the total PM2.5 mass in the western and central U.S. in spring and summer and has positive trends. This work examines climatic factors influencing long-term variations of surface fine dust concentration in the U.S. using station data from the Interagency Monitoring Protected Visual Environments (IMPROVE) network during 1990-2015. The variations of the fine dust concentration can be largely explained by the variations of precipitation, surface bareness, and 10 m wind speed. Moreover, including convective parameters such as convective inhibition (CIN) and convective available potential energy (CAPE) that reveal the stability of the atmosphere better explains the variations and trends over the Great Plains from spring to fall.

While the positive trend of fine dust concentration in the Southwest in spring is associated with precipitation deficit, the increase~~ing~~ of fine dust over the central Great Plains in summer is largely associated with ~~an enhancing~~ enhanced~~of~~ CIN and~~ a weakening~~ weakened ~~of~~ CAPE, which are ~~related to~~caused by increased atmospheric stability due to surface drying and lower troposphere warming. The ~~positive trend~~strengthening of the Great Plains low-level jet also contributes to the ~~increasing~~ increase of fine dust concentration in the central Great Plains in summer via its positive ~~connection~~correlations with surface winds and negative correlation with CIN.

Summer dusty days in the central Great Plains are usually associated with a westward extension of the North Atlantic subtropical high that intensifies the Great Plains

low-level jet and also results in a stable atmosphere with subsidence and reduced precipitation.

1. Introduction

Mineral dust is one of the most abundant atmospheric aerosols by mass. It is lifted to the atmosphere by strong wind from dry and bare surfaces. Severe dust storms have far-reaching socioeconomic impacts, affecting public transportation and health (e.g., Morman and Plumlee, 2013) by degrading visibility~~,~~ and causing traffic accidents, breathing problems, and lung disease~~s~~. Dust storms are found to be associated with increases in non-accidental mortality in the U.S. during 1993-2005 (Crooks et al., 2016).

Major dust sources in the United States are located over the western and the central U.S.~~,~~ While ~~where~~ several deserts are located over the western U.S., e.g., the Mojave, Sonoran, and northern Chihuahuan deserts, ~~and~~ over the central U.S.~~,~~ ~~where~~ the dust sources are largely anthropogenic, in association with agriculture activities (Ginoux et al., 2012). Climate models project a drying trend in the late half of the ~~twenty-first~~21st century over the southwest and central U.S. (e.g., Seager et al., 2007; Cook et al., 2015), regions largely collocated with the major dust sources in the U.S. This raises questions such as how future dust activities will change in the U.S. To project future dust variations, we first need to understand how dust activity varies in the present day. Pu and Ginoux (2017) explored this question using dust optical depth (DOD) derived from MODIS Deep Blue (M-DB2) aerosol products during 2003-2015 and found that variations of dust activity in the U.S. are largely associated with precipitation, near surface wind speed, and surface bareness.

While DOD describes the total optical depth of dust aerosols with different sizes and is widely used to study climate-dust interactions, fine dust with aerodynamic diameter less than 2.5 μm is more frequently used for air quality purposes. ~~The diameter of dust aerosols usually ranges from 0.1 to 50 μm (Duce, 1995), with measured volume median diameters varying from 2.5 to 9 μm (Reid et al., 2003) and clay (diameter < 2 μm) mass fraction representing less than 10% (Kok, 2011). In terms of air quality,~~ F~~f~~ine dust contributes~~ed~~ about 40-50% of total Particulate Matter 2.5 (PM2.5) mass over the southwestern U.S. in spring and about 20-30% over the southwestern to central U.S. in summer (Hand et al., 2017).

Stations in the network of the Interagency Monitoring of Protected Visual Environments (IMPROVE) have collected near surface PM2.5 samples in the U.S. since 1988 (Malm et al., 1994; Hand et al., 2011). Analysis of chemical elements is used to derive surface fine dust concentration. Due to its long temporal coverage, this dataset has been widely used to study long-term variations of surface fine dust in the U.S. Using IMPROVE data, Hand et al. (2016) found an increasing trend of fine dust in spring in the southwestern U.S. during 1995-2014 and related this trend to a negative Pacific decadal oscillation (PDO) from 2007 to 2014. Tong et al. (2017) also found a rapid increase of dust storm activity in the Southwest from 1988 to 2011 and related the trend to sea surface temperature variations in the Pacific. Later, Hand et al. (2017) examined the trends of IMPROVE fine dust concentration in different seasons from 2000 to 2014 and found positive trends over the southwestern U.S. in spring and over the central U.S. in summer and fall. Similarly, Zhang et al. (2017) also found a positive trend of fine dust over the central U.S. from 2005 to 2015 and suggested this trend may contribute to the increase of absorbing aerosol optical depth in the region. Nonetheless, the possible causes of the fine dust trends, especially the increase of fine dust over the central U.S., have not been thoroughly discussed by previous studies. Here, we explore the underlying factors driving the long-term variations of fine dust from 1990 to 2015. We start with local environmental factors and then examine the possible influence of the low-level jet over the Great Plains on fine dust concentration in summer.

The following section describes the data and analysis method used in the paper. Section 3 presents our major results and conclusions are summarized in Section 4.

2. Data and Methodology

2.1 IMPROVE fine dust

IMPROVE stations are located in National Parks and wilderness areas in the United States, with PM2.5 sampling performed twice weekly (Wednesday and Saturday; Malm et al. 1994) prior to 2000 and every third day ~~since March 1988~~afterwards. Records from 204 stations within a domain of 15°-53°N and 60°-127°W are used in this study, and most of the stations have data extending back more than 10 years~~contain data longer than 10 years~~ (Fig. S1 in the Supplement). Elemental concentration is determined from X-ray fluorescence, and fine dust concentration is calculated using the concentrations of aluminum (Al), silicon (Si), calcium (Ca), iron (Fe), and titanium (Ti) by assuming oxide norms associated with predominant soil species (Malm et al., 1994; their Eq. 5). More details regarding IMPROVE stations, sampling, and analysis method can be found in previous studies (Hand et al., 2011; 2012; 2016; 2017).

We averaged daily station data to monthly means and then interpolated them to a 0.5° by 0.5° grid using inverse distance weighted interpolation, i.e., weights depending on the inverse cubic distance between the site location and the interpolated grid point. All daily data are used to calculate monthly mean. We tried the criteria of about 50% completeness (i.e., at least 5 records in each month) for calculating monthly mean, and the results are similar. In daily composite analysis, daily station data are interpolated to a 0.5° by 0.5° grid using the same method. Least squares linear trend analysis is applied to the interpolated data, and student-t test is used for statistical significance test. We realize that the time-varying station numbers could contribute to the uncertainties of our trend analysis; so similar analysis is also applied to station data with long-term records (see Fig. 1 for details).

Following Pu and Ginoux (2017), ~~several~~two ~~–~~ dusty regions are selected for analysis. The southwestern U.S. (WST for short; 32°-42°N, 105°-124°W) and Great Plains (GP for short; 25°-49°N, 95°-105°W) cover the major dust source regions in the U.S. (black boxes in Fig. 1).~~,~~ In later analysis, we also focused over ~~while~~ the central Great Plains (CGP for short; 32°-40°N, 95°-102°W) in summer ~~is chosen~~ to examine the ~~increasing~~ positive trend of fine dust in the region.

2.2 Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) products

CALIOP is the two-wavelength polarization lidar carried by Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) satellite, which was launched in April 2006 (Winker et al., 2004; 2007). CALIOP measures backscattered radiances attenuated by the presence of aerosols and clouds, whose microphysical and optical properties are retrieved. Daily products are available since June 2006. To examine the vertical profile of dust concentration in the U.S., both the daily 532 nm total attenuated backscatter from Level 1 product and the depolarization ratio from Level 2 product are used. The d~~D~~epolarization ratio can be used to separate spherical and non-spherical hydrometeors and aerosols (Sassen, 1991), and here ~~≥~~ a threshold of 0.2 is used to separate non-spherical dust from other aerosols (Li et al., 2010).

2.2 Precipitation

The Precipitation Reconstruction over Land (PRECL; Chen et al., 2002) from the National Oceanic and Atmospheric Administration (NOAA) is a global analysis available monthly from 1948 to present at a 1° by 1° resolution.~~, and~~ Its relative high-resolution and long records are~~is~~ suitable to study long-term connections between fine dust and precipitation. The dataset is derived from gauge observations from the Global Historical Climatology Network (GHCN), version 2, and the Climate Anomaly Monitoring System (CAMS) datasets. Monthly precipitation from 1990 to 2015 is used.

2.3 Leaf area index (LAI)

Monthly LAI derived from the version 4 of Climate Data Record (CDR) of Advanced Very High Resolution Radiometer (AVHRR) surface reflectance (Claverie et al., 2014) and produced by the National Aeronautics and Space Administration (NASA) Goddard Space Flight Center (GSFC) and the University of Maryland is used. The gridded monthly data are on a 0.05° by 0.05° horizontal resolution and available from 1981 to present. ~~This dataset is selected due to its high spatial resolution and long~~

~~temporal coverage. Monthly data from 1990 to 2015 are used.~~ A detailed discussion on the algorithm and evaluation of the dataset can be found by Claverie et al. (2016). This dataset is selected due to its high spatial resolution and long temporal coverage. Monthly data from 1990 to 2015 are used.

Surface bareness is derived from seasonal mean LAI, and is calculated following Pu and Ginoux (2017),

$$Bareness = -exp(LAI) \quad . \qquad (1)$$

2.4 Reanalysis

North American Regional Reanalysis (NARR; Mesinger et al., 2006) provides 3-hourly, daily, and monthly meteorological variables from 1979 to the present at a high spatial resolution (i.e., about 32km horizontally). Precipitation in the NARR is assimilated with observations. Here daily precipitation is used for daily composite analysis in section 3.3.2. The reanalysis reasonably captures the hydroclimatic fields in the continental U.S. on multiple time scales (Ruiz-Barradas and Nigam, 2006; Ruane, 2010a, b), thus is suitable to study the connection between fine dust concentration and local hydroclimatic variables. ~~Here~~ D~~d~~aily and monthly convective variables such as convective inhibition (CIN), and convective available potential energy (CAPE) are used. CIN is defined as the energy that a parcel needs to overcome to rise above the level of free convection (LFC), and is usually written as:

$$CIN = -\int_{P_{sfc}}^{P_{LFC}} R_d\left(T_{vp} - T_{ve}\right) dlnp \quad , \qquad (2)$$

where P_{LFC} is the pressure at LFC, P_{sfc} is the pressure at the surface, R_d is the specific gas constant for dry air, T_{vp} is the virtual temperature of the lifted parcel, and T_{ve} is the virtual temperature of the environment. ~~By definition,~~ CIN is usually a negative variable, with bigger CIN (in absolute value) indicating greater inhibition. On the other hand, CAPE describes the positive buoyancy of an air particle from the LFC to the equilibrium level (neutral buoyancy), and can be written as:

$$CAPE = -\int_{P_{LFC}}^{P_{EL}} R_d\left(T_{vp} - T_{ve}\right) dlnp \quad , \qquad (3)$$

where P_{EL} is the pressure at the equilibrium level. Both CIN and CAPE describe the stability of the atmosphere, and usually convection easily occurs when CAPE is high and CIN is low (in absolute value; e.g., Colby, 1984; Riemann-Campe et al., 2009; Myoung and Nielsen-Gammon, 2010a). Note the two variables can sometimes vary in opposite directions. Indeed, when CAPE is high, strong inhibition may still prohibit the occurrence of deep convection.

In addition, daily and monthly means of horizontal wind speed at 900 hPa, temperature at 700 hPa (T_{700}), 10 m wind speed, dew point temperature (T_{dp}), and 2 m air temperature (T_{2m}), total cloud cover, total and convective precipitation are used.

Another reanalysis used in this work is the ERA-Interim (Dee et al., 2011) from the European Centre for Medium-Range Weather Forecasts (ECMWF). ERA-Interim is a global reanalysis with a horizontal resolution of T255 (about 0.7° or 80 km) and 37 vertical levels, available from 1979 to present. It complements the regional reanalysis by providing a larger domain to analyze circulation variations and also a few surface variables (such as surface turbulent stress) that are not available in the NARR. 6-hourly analysis and 3-hourly forecast variables such as surface turbulent~~ce~~ stress, vertical and horizontal winds, air temperature, and specific humidity from 1000 to 200 hPa, 850 hPa winds and geopotential height are used to calculate daily means of these variables.

2.5 Multiple-linear regression

To understand the connection between the potentially controlling factors and the variation of fine dust concentration, multiple-linear regressions are applied by regressing the observed gridded fine dust concentration onto 3, 4, or 5 standardized controlling factors, a method similar to the one used by Pu and Ginoux (2017). Since multiple controlling factors and gridded surface fine dust have different horizontal resolutions, for the regression analysis we first ~~Here all data are~~ interpolated all variables to a 1° by 1° grid, then apply the regression at each grid point ~~for the regression analysis~~.

The fine dust concentration ~~is then~~can be reconstructed by using the regression coefficients and observed variations of the controlling factors (such as precipitation, surface wind, and bareness). We focus our analysis on two statistical properties: correlations of regional averaged time series and (centered) pattern correlations (e.g., Pu et al., 2016b) for the trends. These two properties are calculated for both observed and regression model estimated (i.e., reconstructed) fine dust concentrations.

3. Results

3.1 Trends of surface fine dust concentration during 1990-2015 and local controlling factors

Figure 1 shows the trend of fine dust concentration from gridded data (shading) and also those from stations with at least 23 years of consecutive records (colored circles) from 1990 to 2015. Most long-term sites show trends similar to those from the interpolated data, with a few exceptions, e.g., over northern Alabama, where interpolated data show positive trends due to the influence of nearby stations with shorter records (Fig. S1 in the Supplement). Significant positive trends are found over the southwestern U.S. in spring (MAM), over the central to southern Great Plains in summer (JJA), and the northern Great Plains in fall (SON). Dust concentration also increases over southwestern Arizona (up to 0.06 μg m^{-3} yr^{-1}), by about 2.5% of its climatological value (Fig. S2 in the Supplement) per year, in all seasons. A similar increasing trend of fine dust in southern Arizona in spring from 1988 to 2009 is also noticed by Sorooshian et al. (2011). A decreasing trend is found over the northeastern U.S. in all seasons as well. The overall pattern is somewhat similar to the trend identified by Hand et al. (2017; their Fig. 9) for 2000-2014, who also found increasing trends of fine dust in the Southwest in spring and over the ~~central Great Plains~~CGP in summer. One thing we want to point out here is that most of the stations in the Great Plains have records shorter than 15 years, with only three stations having records for more than 25 years (Fig. S1 in the Supplement), therefore the positive trends here are combinations of interpolated information from nearby stations in the early period (before ~2002) and more reliable data largely from local stations in the late period.

As suggested by previous studies, the trend of fine dust may be biased due to suspicious trends in some chemical species (Al, Si, and Ti) used to construct fine dust in association with changes of analytical methods (e.g., Hyslop et al., 2015; Hand et al., 2016; Hand et al., 2017). Fe has been suggested as a good proxy of fine dust since it's more stable and is a key component of dust (Hand et al. 2016; 2017). We examined the trend of fine Fe (Fig. S3 in the Supplement), and found the pattern is very similar to the trend of fine dust. In fact, we found the correlations between seasonal mean fine dust and

Fe (both gridded data and long-term stations) are around 0.90 (significant at the 99% confidence level) in most part of the U.S. during 1990-2015 (Fig. S4 in the Supplement). This suggests the trends revealed directly from surface fine dust record are comparably reliable as those calculated from Fe. So we use fine dust concentration for this analysis.

What are the dominant factors influencing the variations of surface fine dust concentration? Hand et al. (2016) found that the PDO played an important role in the variability of fine dust concentration over the Southwest in March by creating a windier, drier, and less vegetated environment. We would like to extend their analysis to other seasons and regions. In addition, we focus on identifying key controlling factors at the local level because remote forcings such as the PDO influence dust variations through their tele-connection with local controlling factors. Pu and Ginoux (2017) found that local precipitation, surface bareness, and surface wind speed could explain 49% to 88% of the variances of dust event frequency (derived from DOD) over the western U.S. and the Great Plains in different seasons from 2003 to 2015. We first examine to what extent these factors can explain the variance of near surface fine dust concentration. Similar to Pu and Ginoux (2017), we do not separate the contribution from local emissions or remote ~~transportation~~ transport to the fine dust concentration, although contributions from Asian dust in spring over the western U.S. (Fischer et al., 2009; Creamean et al., 2014; Yu et al., 2012) and from North African dust in summer over the southeastern U.S. (Perry et al., 1997; Prospero, 1999a) have been observed.

Figures 2a-d show~~s~~ the dominant controlling factor among ~~the three (~~precipitation, surface wind, and bareness~~)~~ for fine dust concentration variations on the interannual time scale from 1990-2015 at each grid point. Precipitation plays an important role in most parts of the southern U.S. in winter. In spring, surface wind starts to dominate the variations of fine dust along the Gulf coast and eastern Great Plains, consistent with the intensification of the Great Plains low-level jet (e.g., Helfand and Schubert, 1995; Weaver and Nigam, 2008; Pu and Dickinson, 2014; Pu et al., 2016a) in April and May, while bareness is important over the western Great Plains and the Midwest. During summer, the influence of surface wind speed gets stronger, especially over western Arizona and the lower Mississippi basin, whereas bareness and precipitation are also important in many parts of the Great Plains and western U.S. Precipitation becomes the dominant factor over most parts of the U.S. again in fall, with surface winds playing a weak role over the southeast and northeast coasts.

The regression coefficients obtained here share some similarity with those shown by Pu and Ginoux, (2017; their Fig 4) using DOD, e.g., the importance of surface bareness in the Great Plains in spring and summer. However, there are also quite large differences, likely due to different periods of regression and the fact that the DOD and surface fine dust concentration are not always linearly related to each other (Fig. S5 in the Supplement). ~~For instance, over the Great Plains and the southwestern U.S., seasonal mean fine dust is linearly related to the DOD in spring but not so in summer. As mentioned earlier, F~~fine dust covers a small fraction of the total mass distribution of dust particles, thus the connections between fine dust concentration and the controlling factors could be different from those with the DOD. For example, the scavenging effect of precipitation is more efficient on small particles (e.g., Zender et al., 2003) and as a result precipitation generally plays an overall more important role on fine dust variations than on the DOD, especially in winter, spring, and fall.

The correlations of regional averaged time series between reconstructed fine dust concentration in the southwestern U.S. (using regression coefficients and observed variations of precipitation, surface wind, and bareness) and that from the IMPROVE range from 0.69 in fall to 0.82 in winter, indicating that the above three factors explain about 48% to 67% variances of fine dust in the Southwest from 1990 to 2015. Over the Great Plains, ~~correlations between the reconstructed and observed fine dust concentration ranges from 0.57 in summer to 0.69 in winter~~these factors only ~~,~~ explain~~ing~~ 32% to 48% variances statistically, much lower than over the Southwest. Also note the low confidence level of the regression coefficients over the CGP in summer (Fig. 2c), which indicates that the above three factors are not sufficient to well explain the variations of fine dust in the Great Plains.

~~The pattern correlations between the observed trend and the trend from reconstructed fine dust are all above 0.80 in the Southwest except in the summer, whereas in the Great Plains region, the pattern correlations are much lower, from 0.06 in fall to 0.48 in winter. In fact, the reconstructed trend missed the observed positive trend of fine dust over the central Great Plains in summer (not shown). This is consistent with the low confidence level of the regression coefficients over the central Great Plains in summer (Fig. 2c) and indicates that the above three factors are not sufficient to well explain the variations of fine dust in the central Great Plains.~~

The development of dust storms has long been related to convection and atmospheric stability (e.g., Marsham et al., 2008; Cuesta et al., 2009). Here we examine whether the variances of fine dust concentration and trend can be better represented by adding CIN (i.e., four-factor) and both CIN and CAPE (i.e., five-factor) in addition to the three factors (i.e., three-factor) discussed above.

Figure 2e shows correlations (blue bars) between the observed and the reconstructed regional mean fine dust concentration ~~from~~ using three-, four-, and five-factor regressions, and corresponding pattern correlations (pink dots) between trends from the observed and reconstructed fine dust for the Great Plains and the southwestern U.S. Over the Great Plains, pattern correlations are largely improved when including CIN and CAPE, especially in spring (from 0.30 to 0.89) and summer (from 0.34 to 0.93), although slightly decrease in winter. The correlations of regional mean time series between the reconstructed and observed fine dust are also slightly improved from three-factor regression to five-factor regression. Over the Southwest, the improvement of pattern correlation is smaller, and the correlations of time series change little when including CIN and CAPE. ~~In both regions, correlations of interannual variation between the reconstructed and observed fine dust are slightly improved from three-factor regression to five-factor regression. Pattern correlations are largely improved over the Great Plains when including CIN and CAPE, especially in spring (from 0.30 to 0.89) and summer (from 0.34 to 0.93), although slightly decreased in winter, whereas the improvement of pattern correlations in the Southwest is much weaker.~~

The collinearity among the factors used in the multiple linear regression can be examined by the variance inflation factor (VIF; O'Brien, 2007; Abudu et al., 2011), and usually values between 5 and 10 are considered high collinearity and the results of regression are less reliable. Increasing the number of predictors in multiple linear regression generally increase VIFs. The VIFs for three-factor regression are around 1 and

2 in most areas, with a few spots around 3 (not shown), while the VIFs for five-factor regression are slightly higher, especially for CIN and CAPE over the Southwest (Figs. S6 and 7 in the Supplement). The increase of VIF and relatively weak improvement in the correlations in the Southwest when adding the convective factors suggest that three factors (precipitation, surface wind, and bareness) are sufficient to capture the variations and trend of surface fine dust in the region. Over the Great Plains, adding CIN and CAPE can better explain the variations.

We now examine key factors driving the observed positive trends of fine dust concentration in spring and summer, the dustiest seasons (Fig. S2 in the Supplement), based on the above analysis. Specifically, we focus on the positive trends of surface fine dust over the southwestern U.S. in spring and over the CGP in summer (Fig. 1b and c). Figure 3a shows the trend of observed and reconstructed fine dust concentrations in spring along with three components contributed to the reconstructed trend (i.e., from precipitation, bareness, and surface wind). The reconstructed trend (Reg (all)) largely captures the positive trend in the Southwest shown in the observation (Obs). Among the three factors, precipitation plays the most important role in contributing to the positive trend over the Southwest, consistent with its dominant role in explaining observed interannual variability (Fig. 2b). The increase of fine dust is mainly associated with a decreasing trend of precipitation in the Southwest (Fig. 3b). Such a drying trend has been related to an increase of anticyclonic conditions in the North East Pacific (Prein et al., 2016) and an intensification of Pacific trades during 2002-2012 (Delworth et al., 2015).

The reconstructed summer trend using coefficients from five-factor regression is very similar to the observation, with a pattern correlation of 0.95 in the domain (Fig. 4a).

The positive trend over the ~~central Great Plains~~CGP is largely contributed by CIN, with a positive center at northern Texas, western Kansas, and Oklahoma. Parts of the positive trend over Oklahoma and western Kansas are contributed by CAPE. In fact, both CIN and CAPE have significant negative trends over the ~~central Great Plains~~CGP, although the trend of CAPE is slightly weaker than that of CIN (Fig. 4b). A decrease of CIN (i.e., an increase in its absolute value) denotes an increasing inhibition of convection, while ~~a weakening of~~weakened CAPE denotes a decreasing instability associated with moist convection. Note that CIN is also significantly negatively correlated with fine dust concentration on interannual time scale (r= -0.39, p= 0.05). This again indicates that CIN plays a more important role than CAPE in the recent positive trend of fine dust.

Both the trends of the CIN and CAPE denote an ~~increasing~~ increase of atmospheric stability. Changes of CIN and CAPE have been related to boundary layer or near-surface temperature and moisture (e.g., Ye et al., 1998; Gettelman et al., 2002; Alappattu and Kunhikrishnan, 2009). Myoung and Nielsen-Gammon (2010b) found that the variations of CIN over Texas in the warm season can be well represented by the differences of temperature at 700 hPa (T_{700}) and surface dew point temperature (T_{dp}), i.e., T_{700}-T_{dp}. While T_{700} is a good proxy for temperature at the free-troposphere below the LFC, T_{dp} denotes the dryness at the surface. Thus, T_{700}-T_{dp} represents a joint effect of surface drying and warming at 700 hPa, a positive anomaly of which indicates increased atmospheric stability~~generally stable atmosphere~~. Here we find both CIN and CAPE have significant negative correlations with T_{700}-T_{dp} over the ~~central Great Plains~~CGP (Fig. 4c). A significant positive trend of T_{700}-T_{dp} is also found, supporting the assumption that the atmospheric stability is enhanced during the period. Such a changes of stability is largely due to the increase of T_{700}, although surface drying also contributes.

CIN is also found to be significantly correlated with rain days (daily precipitation $\geq$ 1 mm day^{-1}) in summer in Texas (Myoung and Nielsen-Gammon, 2010b). Here a similar positive correlation between CIN and rain days in the ~~central Great Plains~~CGP is~~n~~ also found from 1990 to 2015 (r=0.79, p<0.001), suggesting that CIN could influence fine dust concentration via its connection with rain days. A stable atmosphere prevents deep moist convection, which reduces the chance of scavenging by precipitation, and also likely prevents dilution of fine dust concentration in the boundary layer with the clean air above through convective mixing. The connection underlying CIN and fine dust concentration is further discussed in section 3.3 using daily data.

3.2 The connection between the Great Plains low-level jet and summertime fine dust variations in the ~~central Great Plains (~~CGP~~)~~

An important feature related to the moisture and heat transport and precipitation in the Great Plains from late spring to summer is the Great Plains low-level jet~~ (LLJ)~~, which develops in April and reaches its maximum wind speed in June and July at around 900 hPa (e.g., Weaver and Nigam, 2008; Pu et al., 2016a). The southerly jet covers most of the southern to ~~central Great Plains~~CGP, and turns into a westerly around 40° N passing through the Midwest. How this jet may influence the dust concentration in the C~~P~~GP in summer is examined here.

Figure 5a shows the time series of the jet index in summer following the definition of Weaver and Nigam (2008) by averaging 900 hPa meridional wind speed at the jet core (25°-35°N, 97°-102°W) from 1990 to 2015. The jet index is significantly positively correlated with fine dust concentration in the CGP in summer (r= 0.56, p<0.01) and also has a significant positive trend~~ in summer~~, suggesting that the jet also contributes to the increasing of fine dust in the CGP. Such a positive connection between the jet and fine dust concentration can be explained by jet's negative correlation with CIN and positive correlation with the near surface wind speed in the CGP (Figs. 5b). An intensified jet increases the near surface wind speed and meanwhile increases the stability of atmosphere over the CGP by advecting moisture away to the Midwest. ~~and increasing local temperature via northward warm temperature advection (e.g., Walters and Winkler, 2001; Song et al., 2005; Zhu and Liang, 2013).~~

Because most of the IMPROVE sites (4 out of 6) in the ~~central Great Plains~~CGP only have records since 2002, correlations between the jet index and fine dust concentration, CIN, and surface wind for 2002-2015 are also calculated (Fig. 5c). The patterns are similar to those during 1990-2015.

Dust from Africa can be transported to the southeastern U.S. and even Texas in summer (e.g., Perry et al., 1997; Prospero, 1999b, a; 2010; 2014; Bozlaker et al., 2013). Can the intensified jet transport more African dust and thus contribute to the increase of fine dust in the CGP? ~~Does African dust also contribute to the positive trend of fine dust in the CGP via the jet?~~ Fully addressing this question will require a dust model that can well reproduce the emission and transport processes of African dust, which is beyond the scope of this paper. Here we discuss this question based on observational analysis. The regression and trend analysis above suggests that local atmospheric stability largely contributes to the positive trend. Since African dust is transported to the continental U.S.

passing through the Caribbean Sea and the Gulf of Mexico, we assume that the variations of fine dust in stations nearby would reveal the influence of African dust. Two of such stations, VIIS1 (18.3°N, 64.8°W) in the Virgin Islands National Park and EVER1 (25.4°N, 80.7°W) in the Everglades National Park, are used. It is found that the records from these stations have significantly positive correlations with fine dust concentration over the southeastern U.S. in JJA, but not over the CGP (Fig. S8 in the Supplement). This suggests that the influence of African dust is largely over the Southeast on seasonal mean, consistent with the results of Hand et al. (2017), who found the influence of North African dust are mainly over the Southeast, Appalachia, and Virgin islands regions in summer as indicated by a shift of elemental composition in IMPROVE sites.

3.3 Factors contributing to high dust concentration over the CGP in summer

While the negative correlation between fine dust concentration and precipitation in the Southwest is straightforward, the correlation between fine dust and CIN in the CGP is less obvious. Here we further examine the connection between fine dust and CIN and other factors associated with high dust concentration in the area using daily events. As mentioned earlier, since most stations in the CGP have records since 2002, the following analysis focuses on summer during 2002-2015.

3.3.1 Connection between surface fine dust concentration and CIN

What's the physical connection between CIN and surface fine dust concentration? Here we first explore the connection between CIN and a variable that is closely related to dust emission. Figures 6a-c show the scatter plot of standardized (means are removed and then divided by one standard deviation) CIN and friction velocity (U^*) anomalies, for all the days in summer from 2002 to 2015, days when IMPROVE records are available (431 days), and dusty days, defined as days when daily anomaly of IMPROVE observation is greater than one standard deviation (52 days), respectively. U^* is defined as the following,

$$U^* = ([\tau/\rho])^{1/2} = [(\overline{u'w'})^2 + (\overline{v'w'})^2]^{1/4} \ , \qquad (4)$$

where τ is the Reynolds stress and ρ is air density, and $\overline{u'w'}$ and $\overline{v'w'}$ are vertical flux of horizontal momentum. We calculated U^* using ~~the~~ components of surface turbulent~~ce~~ stress~~es~~ ($-\rho\overline{u'w'}$, $-\rho\overline{v'w'}$) from the ERA-Interim. U^* has long been related to dust emission (e.g., Gillette and Passi, 1988; Marticorena and Bergametti, 1995; Zender et al., 2003). As shown in Figs. 6a-c, CIN is significantly negatively related to U^* in all summer days and dusty days~~the friction velocity, which is associated with turbulent fluctuations in the boundary layer~~. This indicates a large negative CIN, or great inhibition for convection, is related to stronger near surface turbulent~~ce~~ fluxes and U^*. ~~Such a negative connection is robust both in days with fine dust records and in dusty days. CIN represents the integrated inhibition from the surface to LFC (Eq. 2), then~~ How does CIN ~~relate to~~influence ~~surface turbulence fluxes and~~ U^*?

In the CGP, both CIN and U^* are significantly correlated with near surface temperature, T_{2m}, in JJA and for days when fine dust records are available (Table 1), ~~CIN is significantly negatively correlated with near surface temperature, T_{2m}, i.e., a strong inhibition is associated with higher T_{2m}, for all days in JJA and for days when fine dust records are available (Table 1). This is consistent with previous study over Texas (Myoung and Nielsen-Gammon, 2010b). Meanwhile, U^* is significantly positively~~

~~correlated with T_{2m} (Table 1),~~ indicating that CIN is connected with U^* via their mutual ~~its~~ connection with near surface temperature. ~~Also, N~~Note such a connection seems not valid during dusty days (correlation between T_{2m} and U^* is not significant). Similarly, we found ~~a close connection among~~significant correlations between CIN and ~~,~~ T_{700}-T_{dp}, and between T_{700}-T_{dp} and U^* (Table 1). This again, suggests that CIN ~~can influence~~is connected with U^* via its connection with surface variables such as temperature and dryness. Variables in Table 1 are all from the ERA-Interim (except CIN) to be consistent with U^*, results are similar if using NARR variables.

One hypothesis for the connection between CIN and U^* in ~~for~~ dusty days is shown in Table 2. A significant positive correlation between CIN and vertical wind at 850 hPa (w850) is found, indicating that when the inhibition is strong, it favors subsidence. This is consistent with the finding by Riemann-Campe et al. (2009) who found in climatology high CIN value is located over subtropical regions with strong subsidence. The subsidence may transport~~s~~ momentum downward and promote~~s~~ U^*. This is consistent with the negative correlation between U^* and w850 (Table 2). However, we also notice that the above connections in dusty days are not valid if using w850 from~~in~~ the NARR, suggesting further investigation on this mechanism is needed.

~~Despite~~ In addition to the connection between CIN and surface variables, the possible mechanism that strong inhibition prevents dilution is also examined. We found four examples in CALIOP snapshots over the CGP when the daily anomaly of near surface fine dust concentration from the IMPROVE network is greater than one standard deviation. Figure 7 shows nighttime 532 nm total attenuated backscatter (shading) on August 10th, 2007 (top) and on June 21st, 2013 (bottom). Black contours show area with depolarization ratio ≥ 0.2, denoting dust aerosols. In both cases, the inhibition is quite strong, with daily CIN anomaly greater than one standard deviation. The difference between the two cases is that on June 21st, 2013, CAPE is higher, which leads to some convection as denoted by the clouds above. However, in both cases, with strong inhibition, dust particles are largely located in a layer between the surface and 2 km. Figure 8 shows a different situation when CIN has positive anomaly (i.e., weak inhibition). In these cases, dust particle extends up to 4 km, and surface fine dust concentrations in the CGP (with anomalies of 2.3 and 2.1 μg m^{-3}) are also lower than those in Fig. 7 (with anomalies of 4.0 and 7.1 μg m^{-3}). Nonetheless, more cases are needed to further verify this mechanism. The anomalous high fine dust concentration in Everglades National Park (Figs. 7-8) in three of the four cases shown here suggest that there may be a contribution from African dust in these days, but further analysis are needed to clarify the magnitude of its contribution.

3.3.2 Large-scale circulation pattern in dusty days

Figure 9 shows the daily composites of related meteorological ~~metrological~~ variables in dusty days, i.e., when daily anomaly of CGP fine dust concentration is greater than one standard deviation. Anomalous high fine dust concentration is associated with a reduced CIN (Fig. 9b) in the CGP, but not so much with CAPE (Fig. 9c). CAPE is anomalously enhanced over the northern Plains and the Midwest. Both the ~~LLJ~~Great Plains low-level jet, near surface wind, and friction velocity are enhanced (Figs. 9d-f). Precipitation (mostly convective precipitation) in the CGP also decreases with reduced cloud cover, but increases in the north (Figs. 9g-i), consistent with enhanced CAPE there.

These features are quite consistent with our analysis above on the favorable condition of enhanced fine dust in the CGP.

Figure 10 shows the composites of vertical velocity (shading), vertical and meridional wind vectors, specific humidity (purple contours), and potential temperature (grey contours) zonally averaged over the ~~central Great Plains~~CGP (95° -102° W), along with fine dust concentration (orange line). Anomalous dry subsidence is centered at 30°-36°N, with anomalous southerly winds at low-level associated with an intensified jet, while a rising motion of moist air is located around 38-42°N with a maximum at 700-400 hPa. The dipole pattern of anomalous vertical velocity is consistent with the precipitation anomaly in the area (Figs. 9g-h). The anomalous potential temperature contour is quite uniform near the surface at 30°-36°N with an inversion around 700 hPa, indicating a well-mixed boundary layer in the region with increased fine dust.

What causes the changes of atmospheric stability, precipitation, and winds? Figure 11 shows the composites of T_{2m} and geopotential height and winds at 850 hPa during dusty days. Following Li et al. (2012a), 1560 gpm contour is used here to denote the western edge of the North Atlantic subtropical high in the 2002-2015 climatology (blue) and in dusty days (red). A westward extension of the subtropical high during dust days is quite evident, with enhanced geopotential height over the southeastern U.S. and the Gulf of Mexico (Fig. 11b). Such a westward extension of the subtropical high intensifies the ~~LLJ~~ low-level jet by increasing the zonal pressure gradient, and also contributes to the anomalous precipitation and vertical velocity patterns, as similar patterns are found in previous studies associated with a westward extension of the subtropical high (e.g., Li et al., 2012a; their Figs. 3a and 4a). The formation of the North

Atlantic subtropical high has been related to the land-sea heating contrast (Wu and Liu, 2003; Liu et al., 2004; Miyasaka and Nakamura, 2005; Li et al., 2012a; Li et al., 2012b). One possible reason of the westward extension of the subtropical high is the anomalous surface warming over large part of the central and eastern U.S. (Fig. 11a) in dusty days that enhances the land-sea temperature gradient.

4. Conclusions

Fine dust is an important component in the total PM 2.5 mass in the western to central U.S. in spring and summer (Hand et al. 2017). Previous studies found positive trends of fine dust concentration in the southwestern U.S. in spring and the central U.S. in summer in the past 20 years (Hand et al., 2016; 2017; Zhang et al., 2017), but the underlying causes are not clear, especially for the positive trend over the central U.S. This study examined local controlling factors associated with variations of near surface fine dust concentration from Interagency Monitoring of Protected Visual Environments (IMPROVE) stations for 1990-2015~~ in each season~~. While precipitation, surface bareness, and surface wind speed largely control the variation of fine dust concentration in the southwestern U.S., including two convective parameters that reveal the stability of the atmosphere, convective inhibition (CIN) and convective available potential energy (CAPE), better explains the variations over the Great Plains from spring to fall.

In particular, we found that the increasing trend of fine dust concentration over the Southwest in spring is associated with a significantly decreasing trend of precipitation, while the positive trend of fine dust over the central Great Plains (CGP) is largely due to enhanced atmospheric stability revealed by ~~an enhancing of~~enhanced CIN (greater inhibition) and ~~a decreasing~~decreased ~~of~~ CAPE. Such a stability change is associated with surface drying and warming in the lower troposphere around 700 hPa, i.e., a positive trend of T_{700}-T_{dp}. A stable atmosphere prevents moist convection that can remove fine dust by in-cloud or precipitation scavenging and also likely prevents the dilution of fine dust concentration by prohibiting convective mixing between the dusty boundary layer air and the clean air above.

The variations of the fine dust concentration in the CGP are also significantly correlated to the Great Plains low-level jet, with a stronger jet corresponding to higher fine dust concentration. Such a connection is largely due to jet's positive correlation with surface wind speed and negative correlation with CIN.

The influence of CIN on dust emission in the CGP is examined using daily data in summer. It is found that CIN is significantly negatively related to surface friction velocity (U^*), i.e., with greater inhibition in association with stronger U^*. Such a connection is largely due to CIN's connection with surface variables such as 2m temperature and dew point temperature. During dusty days, another possible connection is that the anomalous subsidence associated with strong inhibition may transport momentum downward and increase surface U^*.

Dusty days in the CGP in summer are associated with a westward extension of the North Atlantic subtropical high that intensifies the Great Plains low-level jet and surface wind speed, increases atmospheric stability, and also creates anomalous subsidence over the southern to central Great Plains and reduces precipitation~~and the Southeast and rising motion over the Midwest, and correspondingly a south-north dipole pattern of~~

~~precipitation anomaly~~. The westward extension of the subtropical high is likely associated with the anomalous surface warming over the central to eastern U.S.

Our findings have important implications for future projections of fine dust variation in the U.S. Climate models have projected drying trends over the southwestern and the central U.S. (e.g., Seager et al., 2007; Cook et al., 2015) as well as an intensification of the North Atlantic subtropical high (Li et al., 2012b) in the late 21st century, all favorable to an increase of fine dust in the Southwest and ~~central Great Plains~~CGP. Whether current increasing trends of fine dust will persist into the future requires further investigations that include factors not discussed here such as changes of anthropogenic land use, local synoptic-scale systems (e.g., cyclones and fronts), and remote forcings.

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Table 1 Correlations between friction velocity (U^*) and CIN, CIN and 2 meter temperature (T_{2m}), T_{2m} and U^*, T_{700}-T_{dp} (the differences between air temperature at 700 hPa and 2m dew point temperature) and CIN, T_{700}-T_{dp} and U^* for all days in JJA from 2002 to 2015 (1288 days), days when fine dust concentration is available (431 days), and dusty days (52 days). All values are significant at the 95% confidence level (t-test) except those listed in italic.

Table 2 Correlations between U^* and CIN, CIN and vertical wind speed at 850 hPa (w850), w850 and U^* during dusty days in JJA from 2002 to 2015. All values are significant at the 95% confidence level except the value significant at the 90% confidence level is labeled with a "+" (t-test).

Figure 1. Trend (shading) of fine dust concentration (μg m^{-3}) from 1990 to 2015 in (a) DJF, (b) MAM, (c) JJA, and (d) SON from IMPROVE gridded data. Dotted areas are significant at the 95% confidence level. The colored circles show the trend at IMPROVE stations with consecutive records for at least 23 years during 1990-2015. Circles with green outlines denote that the trend is significant at the 90% confidence level. Black boxes denote the averaging areas of the southwestern U.S. (~~left~~solid) and the Great Plains (dashed~~right~~).

Figure 2. (a)-(d) Multiple linear regression coefficients calculated by regressing fine dust concentration from 1990-2015 onto standardized precipitation (purple), bareness (orange), and surface wind (green). Color denotes the most influential factor at each grid (i.e., the largest regression coefficient in absolute value among the three), while saturation of the color shows the magnitude of the coefficient (0 to 0.3). Areas significant at the 95% confidence levels are dotted. (e) Bar-plot showing the correlations between observed regional mean fine dust concentration and the reconstructed concentration using 3, 4, and 5 controlling factors (light, median, and deep blue), and pattern correlation between trends from the observation and from reconstructed fine dust using 3, 4, and 5 factors (light, medium, and deep pink) in the Great Plains (GP) and the southwestern U.S. (WST, black boxes in a-d~~Fig. 1~~). “3-factor” denotes precipitation, bareness, and surface wind, “4-factor” denotes precipitation, bareness, surface wind, and CIN, “5-factor” denotes precipitation, bareness, surface wind, CIN, and CAPE. Black boxes denote the averaging areas of the WST (solid) and GP (dashed).

Figure 3. (a) Observed (Obs) and reconstructed (Reg) trends of fine dust concentration (μg m^{-3}) using three factors in spring from 1990 to 2015. The contributions from each factor (precipitation, bareness, and surface wind) to the overall reconstructed trend are also shown (second row). Dotted areas are significant at the 90% confidence level. Pattern correlation between reconstructed dust concentration trends and observed trends in the domain (25°-49.5°N, 66.5°-125°W) are shown at the top right corner of each plot. Black box denotes the southwestern U.S. (WST). (b) Time series of fine dust concentration (cyan) and precipitation (purple) averaged over the WST and their linear trends (dashed lines; values are listed at bottom left) in spring from 1990 to 2015. Gray shading denotes ±one standard error of the observations. The correlation between fine dust and precipitation is also listed at the bottom in purple.

Figure 4. (a) Observed (Obs) and reconstructed (Reg) tends of fine dust concentration (μg m^{-3}) using five factors in summer from 1990-2015. The contributions from each factor (precipitation, bareness, surface wind, CAPE, and CIN) are also shown (second and third rows). Dotted areas are significant at the 90% confidence level. Pattern correlation between reconstructed dust concentration trends and the observed trends in the domain (25°-49.5°N, 66.5°-125°W) are shown at the right corner of each plot. Black box denotes the central Great Plains (CGP). (b) Time series of fine dust concentration (cyan), CIN (orange), and CAPE (deep blue) averaged over the CGP and their linear trends (dashed lines) in summer from 1990-2015. Gray shading denotes ±one standard error of the observations. (c) Time series of T_{700}-T_{dp} (black), T_{700} (green) and T_{dp} (light blue) and their linear trends (dashed lines) in summer from 1990 to 2015.

Figure 5. (a) Time series of fine dust concentration (μg m^{-3}) averaged in the CGP (cyan) and the index of the Great Plains low-level jet (magenta) and their trends (dashed line) in JJA from 1990 to 2015. Gray shading denotes ±one standard error of the observations. Correlations between the jet index and fine dust concentration, CIN, and near surface wind speed for (b) 1990-2015 and (c) 2002-2015. Colored circles denotes correlations at IMPROVE stations, with green outlines denotes the correlation is significant at the 90% confidence level. Areas significant at the 95% confidence level are dotted in (b) and significant at the 90% confidence level are dotted in (c). Black box in (b)-(c) denotes the CGP region, and deep pink box denotes the averaging area for the jet index.

Figure 6. Scatter plot of standardized friction velocity (U^*) and CIN anomalies for (a) all days in JJA from 2002-2015, (b) days when fine dust data are~~is~~ available, and (c) dusty days (when daily fine dust concentration anomaly is greater than one standard deviation).

Figure 7. Nighttime 532 nm total attenuated backscatter (shading) and depolarization ratio (black contours, values ≥ 0.2 are shown) from CALIOP on August 10th, 2007 (top left) and on June 21st, 2013 (bottom left), along with daily anomalies of fine dust concentration ~~anomaly~~ (μg m^{-3}; shading, right column) and CIN ~~anomaly~~ (blue contour, only negative values from -60 to -120 J kg^{-1} are shown). CALIOP orbit tracks are shown in grey lines (right column) with cyan part and sampling points (A-F) denote the cross-section shown on the left column. Black boxes denote the CGP region.

Figure 8. Same as Fig. 7~~10~~ but for July 2nd, 2011 (top) and July 2nd, 2012 (bottom). Only positive CIN anomalies from 25 to 50 J kg^{-1} are shown (light purple contour).

Figure 9. Daily composites of the anomalies of (a) fine dust concentration (μg m^{-3}), (b) CIN (J kg^{-1}), (c) CAPE (J kg^{-1}), (d) 900 hPa wind speed (m s^{-1}), (e) 10 m wind speed (m s^{-1}), (f) U^* (m s^{-1}), (g) total precipitation (mm day^{-1}), (h) convective precipitation (mm day^{-1}), and (i) total cloud cover (%) during dusty days in JJA from 2002 to 2015. Dotted areas are significant at the 95% confidence level. 900 hPa and 10 m wind anomalies (green vectors) significant at the 95% confidence level are shown in (d) and (e), respectively. Black boxes denote the CGP region.

Figure 10. Daily composite of the anomalies of vertical velocity (shading; 10^{-2} m s^{-1}), potential temperature (grey contours; K), and specific humidity (purple contours; g kg^{-1}) from the ERA-Interim, and fine dust concentration anomalies (bottom; orange line) averaged between 95° and 102° W for dusty days in JJA from 2002 to 2015. Dotted area denotes vertical velocity significant at the 90% confidence level. Topography is masked out in grey. Cyan lines denote the domain of the CGP.

Figure 11. Daily composites of the anomalies of (a) T_{2m} (K) and (b) 850 hPa geopotential height (gpm) and horizontal wind vectors (m s^{-1}; grey) from the ERA-Interim averaged over dusty days in JJA from 2002-2015. Blue and red contours in (b) denote 1560 ~~geopotential height~~gpm in the climatology (2002-2015) and during dusty days, respectively. Areas significant at the 95% confidence level are dotted. Wind vectors

significant at the 95% confidence level are plotted in green. Black boxes denote the CGP region.

Table 1 Correlations between friction velocity (U^*) and CIN, CIN and 2 meter temperature (T_{2m}), T_{2m} and U^*, T_{700}-T_{dp} (the differences between air temperature at 700 hPa and 2m dew point temperature) and CIN, T_{700}-T_{dp} and U^* for all days in JJA from 2002 to 2015 (1288 days), days when fine dust concentration is available (431 days), and dusty days (52 days). All values are significant at the 95% confidence level (t-test) except those listed in italic.

Variables	**All days in JJA**	**Available days**	**Dusty days**
U^*, CIN	-0.54	-0.54	-0.44
CIN, T_{2m}	-0.59	-0.59	-0.39
T_{2m}, U^*	0.39	0.37	*0.19*
CIN, T_{700}-T_{dp}	-0.59	-0.62	-0.59
T_{700}-T_{dp}, U^*	0.37	0.38	*0.14*

Table 2 Correlations between U^* and CIN, CIN and vertical wind speed at 850 hPa (w850), w850 and U^* during dusty days in JJA from 2002 to 2015. All values are significant at the 95% confidence level except the value significant at the 90% confidence level is labeled with a "+" (t-test).

Variables	**Dusty days**
U^*, CIN	-0.44
CIN, w850	0.28^+
w850, U^*	-0.32

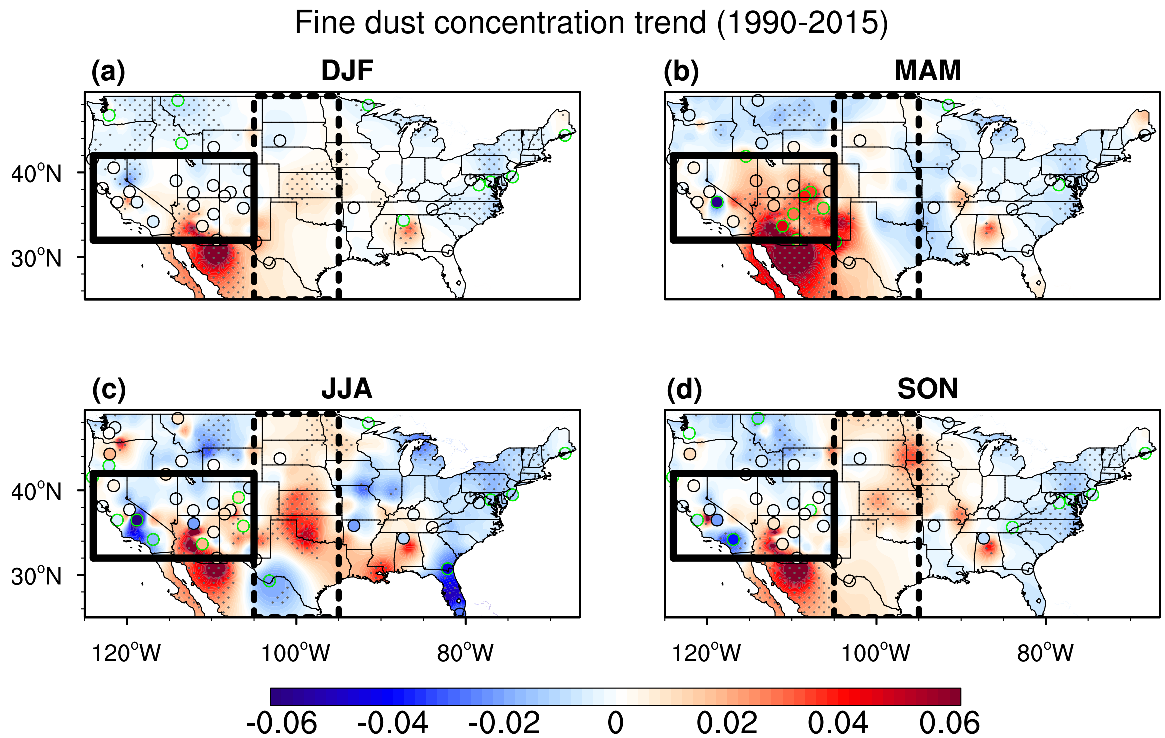


Figure 1. Trend (shading) of fine dust concentration (μg m^{-3}) from 1990 to 2015 in (a) DJF, (b) MAM, (c) JJA, and (d) SON from IMPROVE gridded data. Dotted areas are significant at the 95% confidence level. The colored circles show the trend at IMPROVE stations with consecutive records for at least 23 years during 1990-2015. Circles with green outlines denote that the trend is significant at the 90% confidence level. Black boxes denote the averaging areas of the southwestern U.S. (solid~~left~~) and the Great Plains (dashed~~right~~).

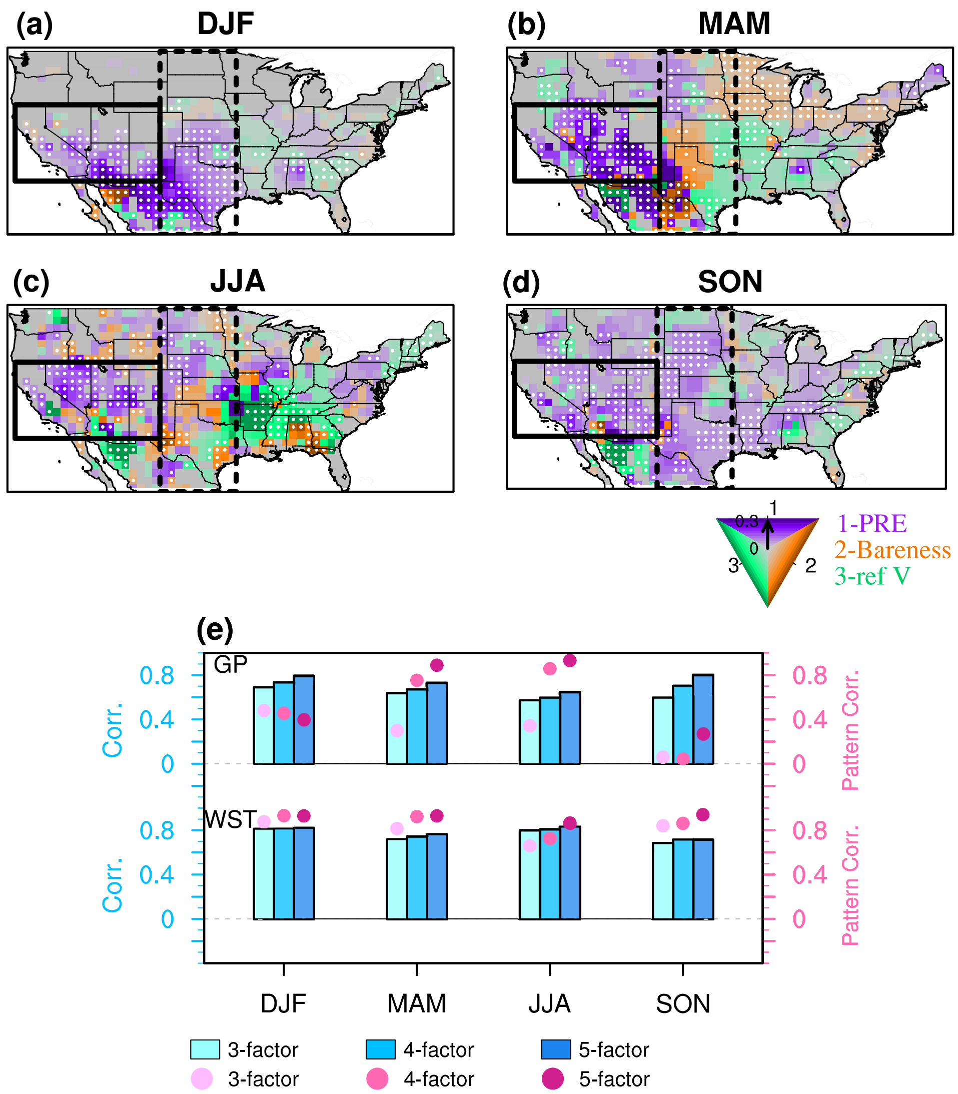
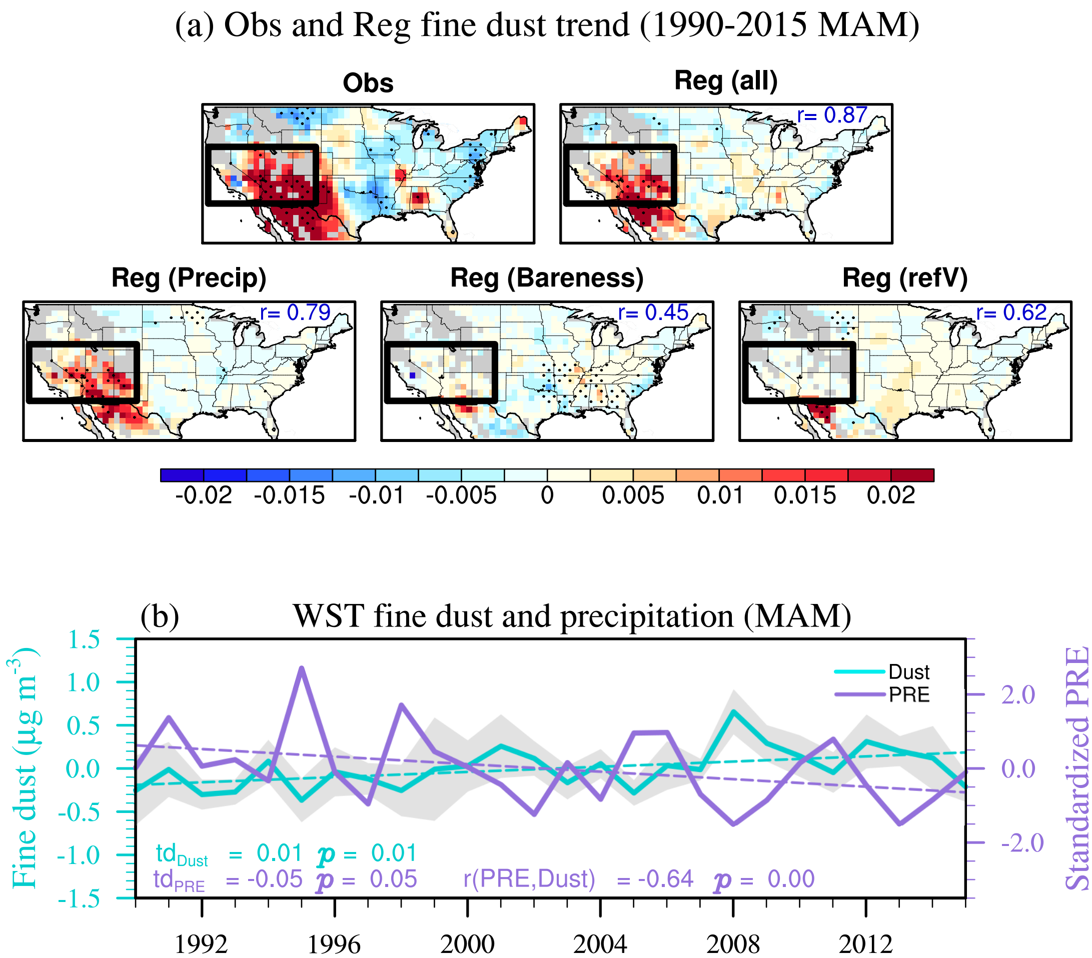


Figure 2. (a)-(d) Multiple linear regression coefficients calculated by regressing fine dust concentration from 1990-2015 onto standardized precipitation (purple), bareness (orange), and surface wind (green). Color denotes the most influential factor at each grid (i.e., the largest regression coefficient in absolute value among the three), while saturation of the color shows the magnitude of the coefficient (0 to 0.3). Areas significant at the 95% confidence levels are dotted. (e) Bar-plot showing the correlations between observed regional mean fine dust concentration and the reconstructed concentration using 3, 4, and 5 controlling factors (light, median, and deep blue), and pattern correlation between trends from the observation and from reconstructed fine dust using 3, 4, and 5 factors (light, medium, and deep pink) in the Great Plains (GP) and the southwestern U.S. (WST, black boxes in a-d~~Fig. 1~~). "3-factor" denotes precipitation, bareness, and surface wind, "4-factor" denotes precipitation, bareness, surface wind, and CIN, "5-factor" denotes precipitation, bareness, surface wind, CIN, and CAPE. Black boxes denote the averaging areas of the WST (solid) and GP (dashed).



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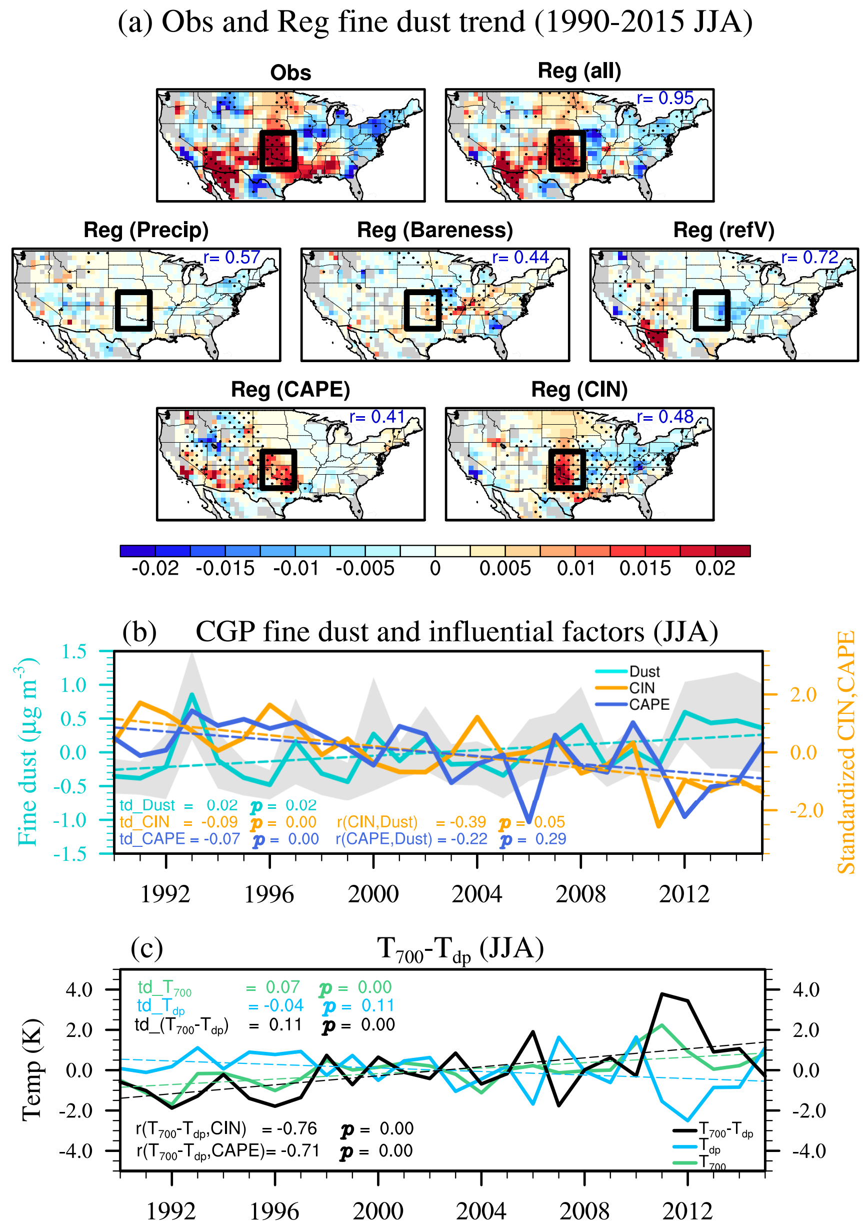

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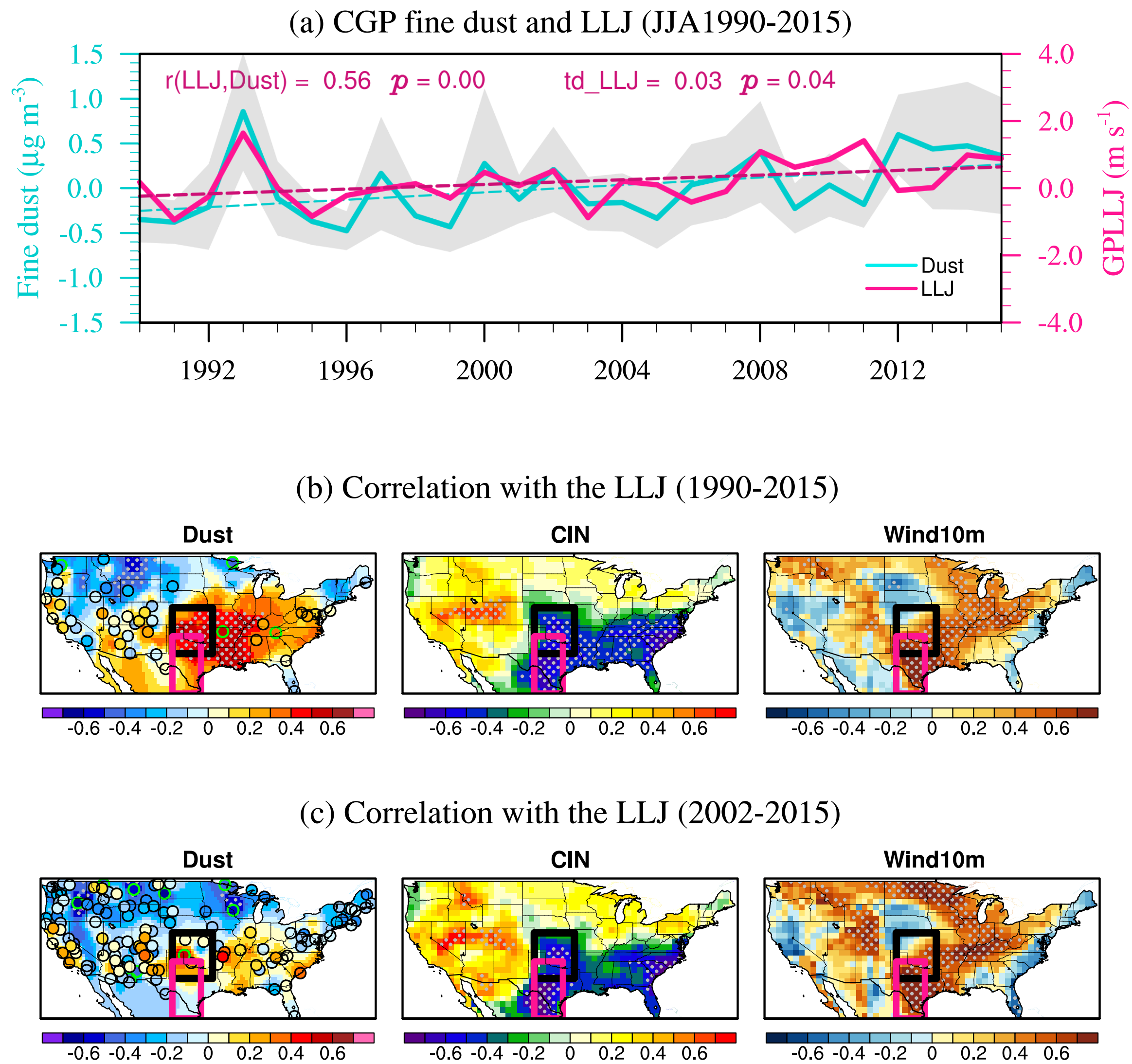

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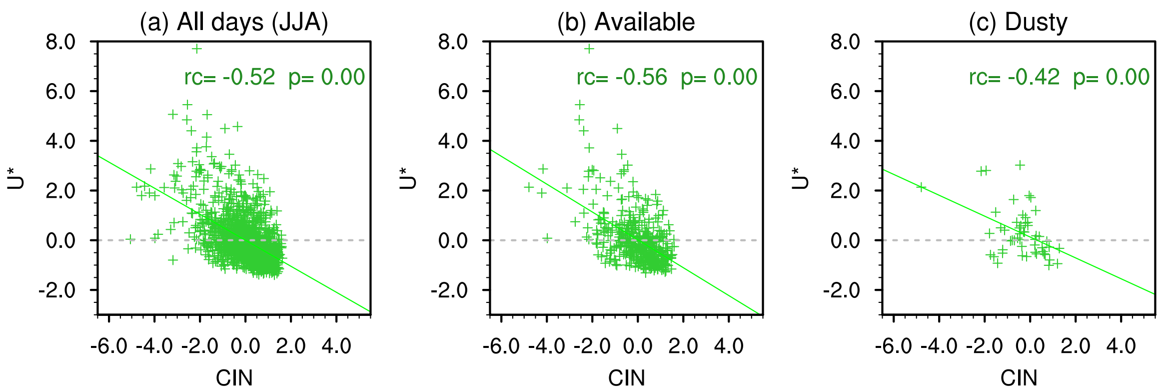

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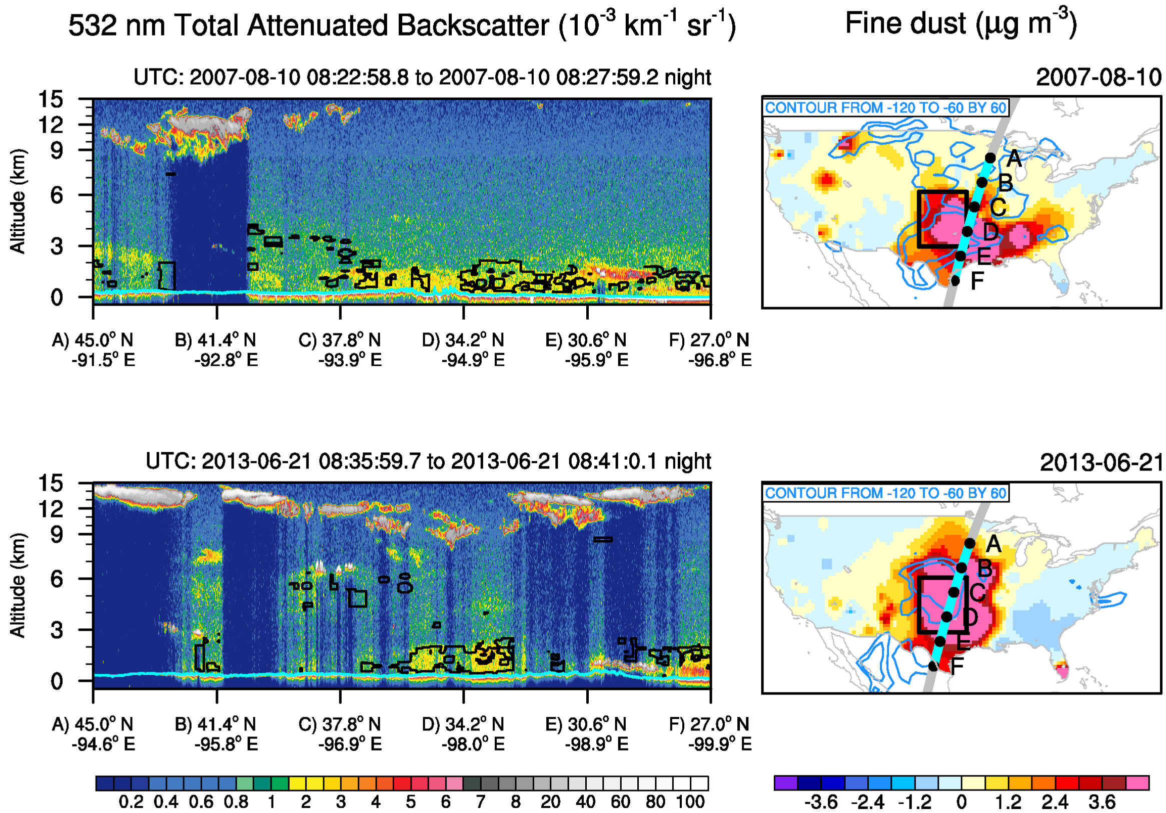

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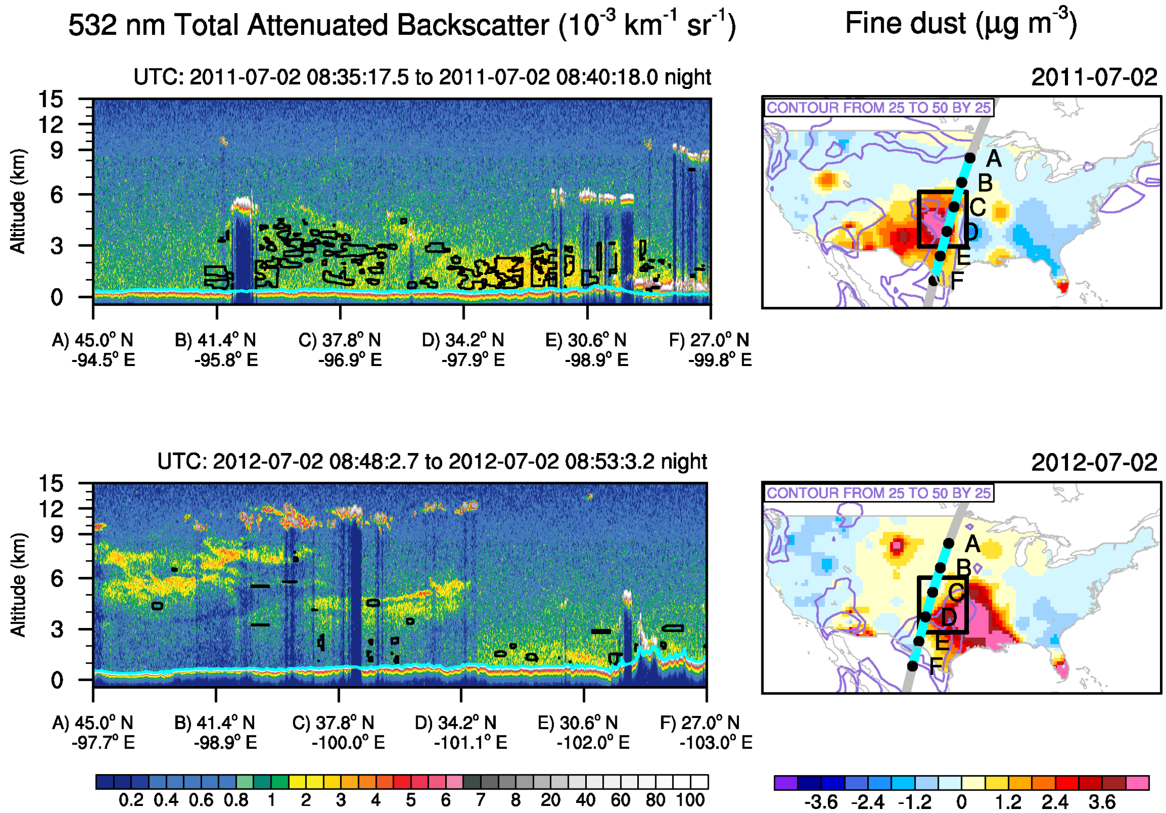
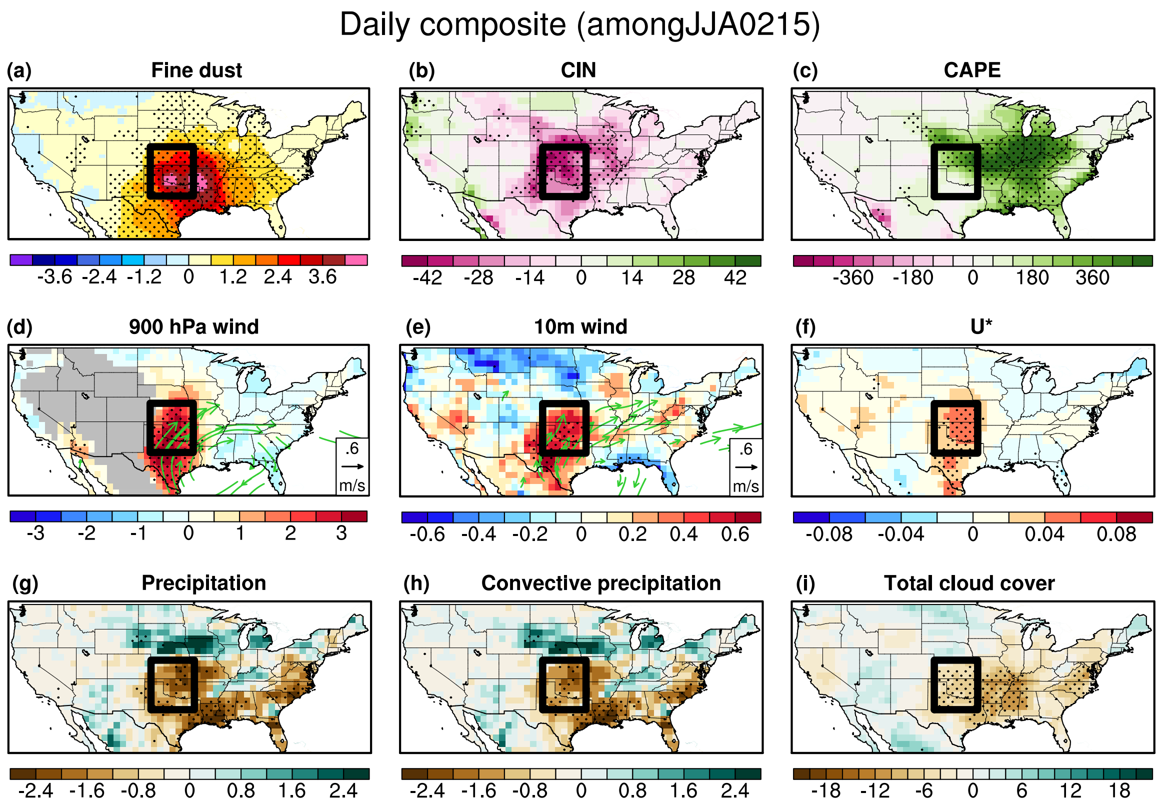


Figure 8. Same as Fig. 7~~10~~ but for July 2nd, 2011 (top) and July 2nd, 2012 (bottom). Only positive CIN anomalies from 25 to 50 J kg^{-1} are shown (light purple contour).



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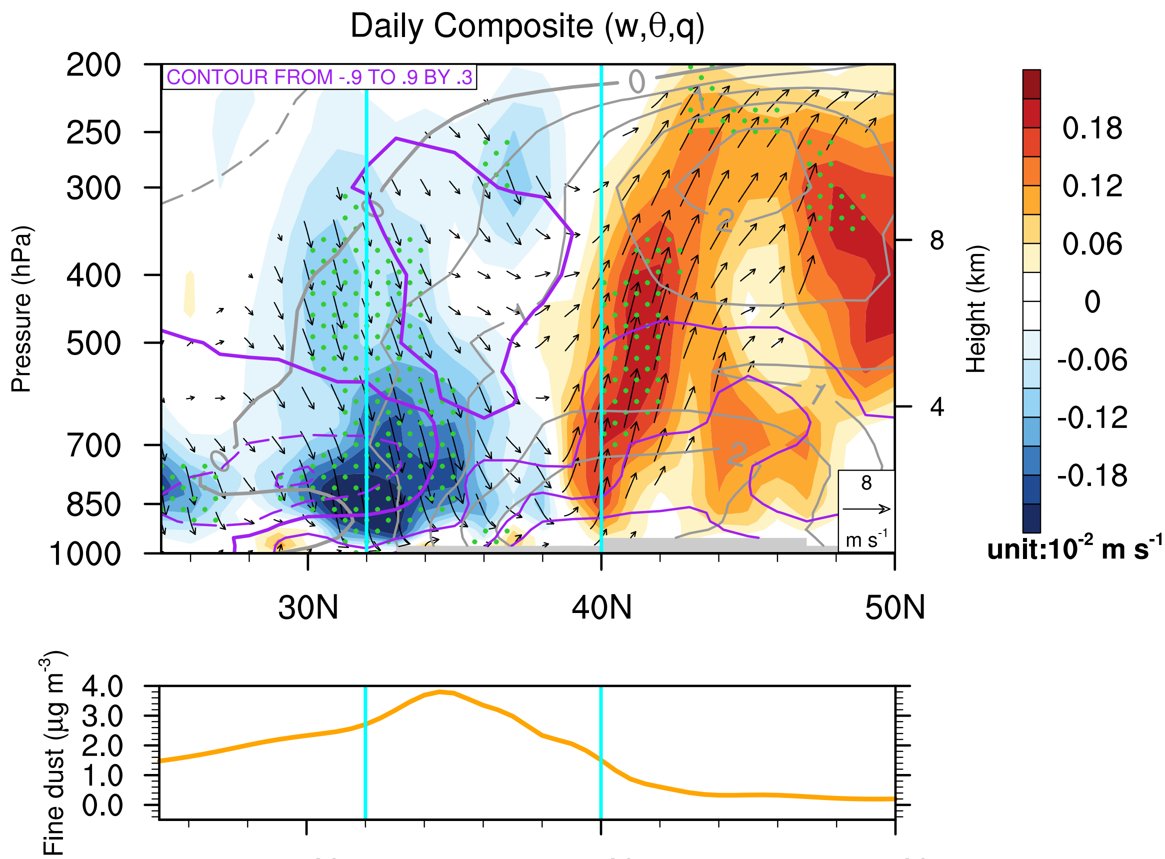
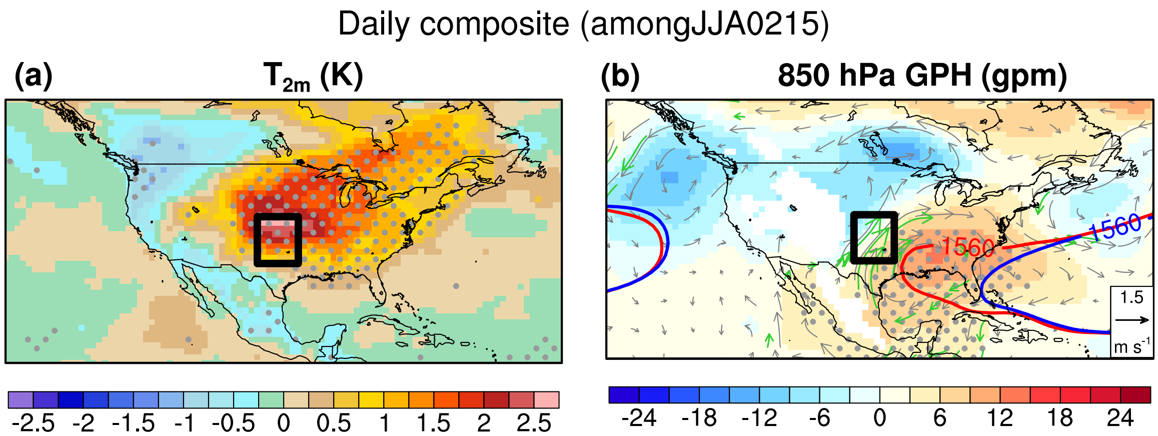


Figure 10. Daily composite of the anomalies of vertical velocity (shading; 10^{-2} m s^{-1}), potential temperature (grey contours; K), and specific humidity (purple contours; g kg^{-1}) from the ERA-Interim, and fine dust concentration anomalies (bottom; orange line) averaged between 95° and 102° W for dusty days in JJA from 2002 to 2015. Dotted area denotes vertical velocity significant at the 90% confidence level. Topography is masked out in grey. Cyan lines denote the domain of the CGP.



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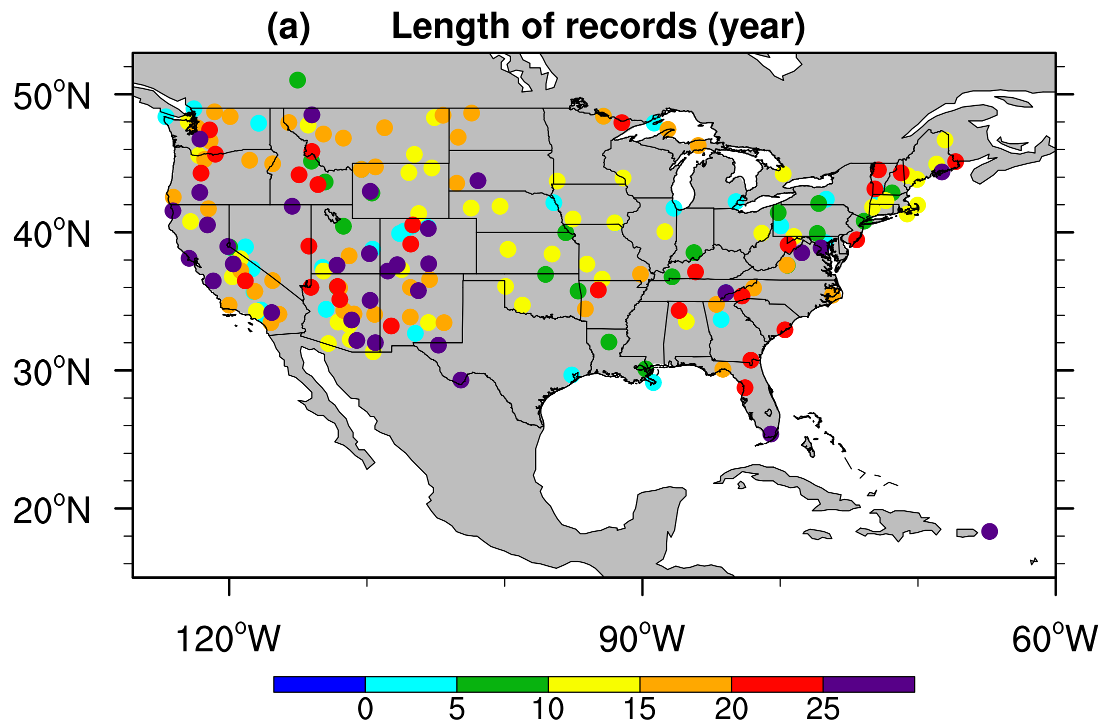
Supplementary figures



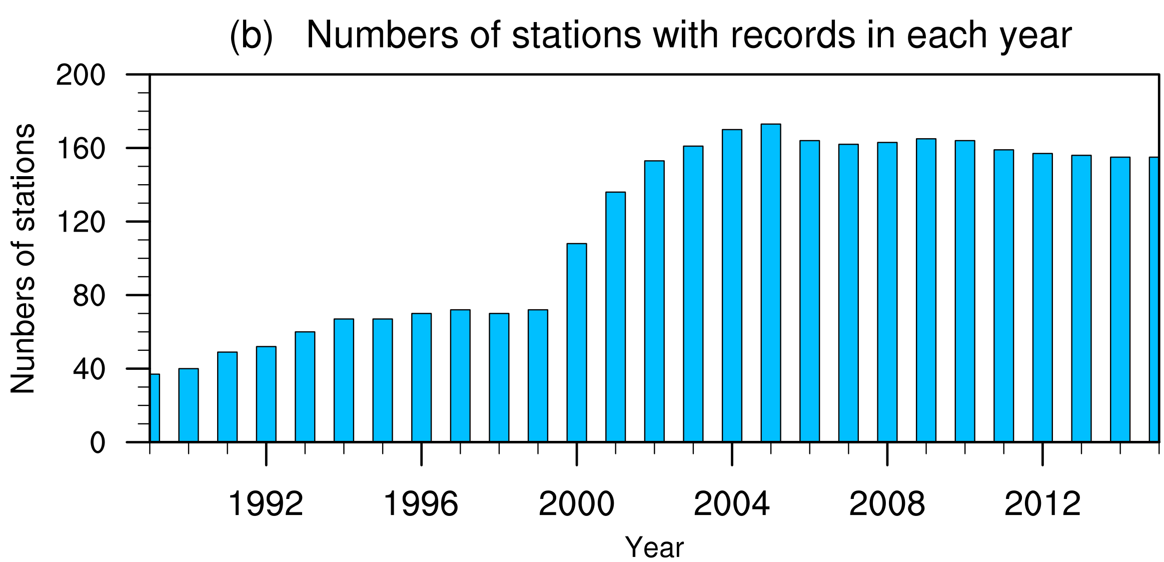


Figure S1. (a) Location of IMPROVE stations in the domain. Color denotes the length (years) of record at each station. (b) Numbers of station with records available in each year from 1989 to 2015.

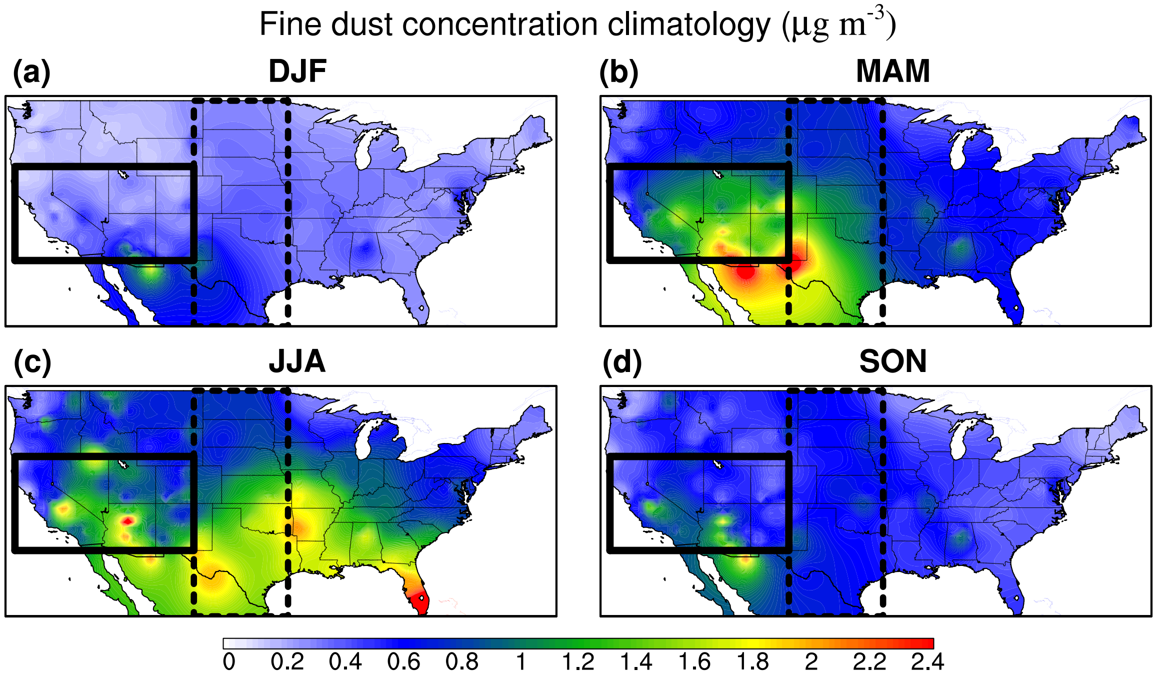


Figure S2. Climatology of fine dust concentration (μg m^{-3}) averaged from 1990 to 2015. Black boxes denote the southwestern U.S. (solid) and the Great Plains (dashed).

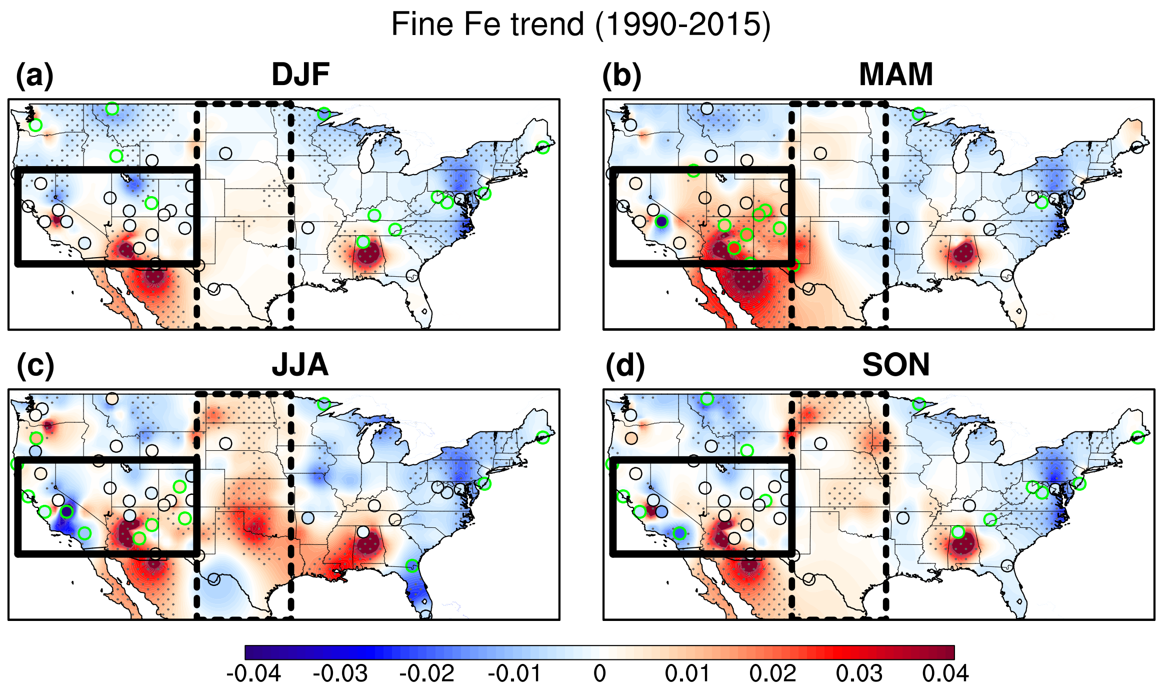


Figure S3. Same as Fig. 1 but for the trend of fine iron (Fe) (10^{-1} μg m^{-3}). Black boxes denote the southwestern U.S. (solid) and the Great Plains (dashed).

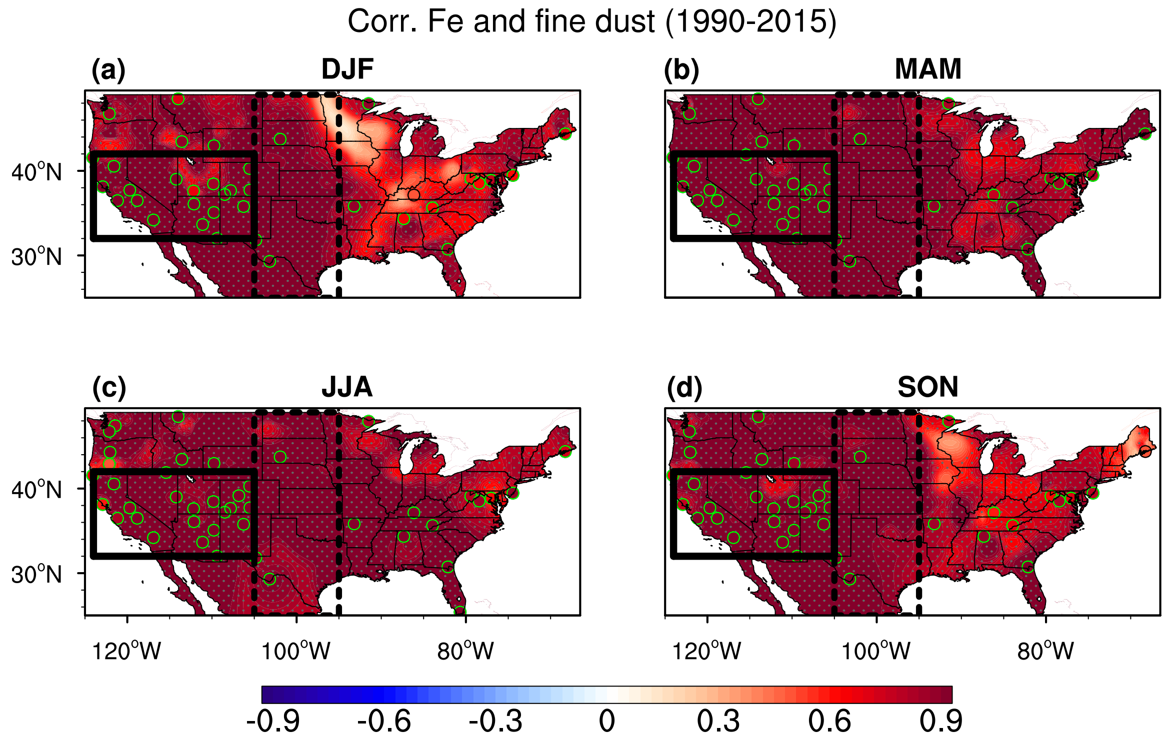


Figure S4. Correlation between fine Fe and fine dust concentrations from 1990 to 2015. Areas significant at the 99% confidence level are dotted in grey. The colored circles show correlations at IMPROVE stations with consecutive records for at least 23 years during 1990-2015. Circles with green outlines denote correlations are significant at the 99% confidence level. Black boxes denote the southwestern U.S. (solid) and the Great Plains (dashed).

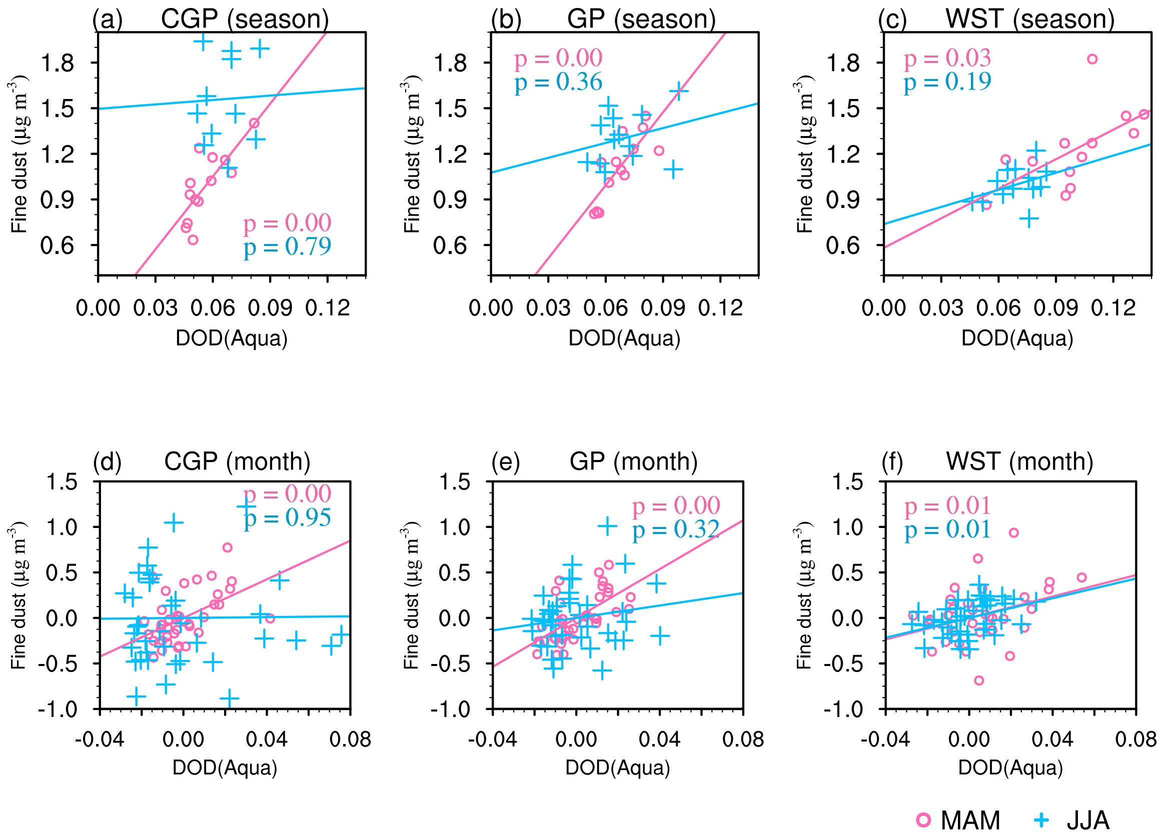


Figure S5. Scatter plot of (a)-(c) seasonal mean and (d)-(f) monthly mean fine dust concentration versus DOD during spring (pink circles) and summer (light blue plus) over the central Great Plains (CGP), Great Plains (GP) and the southwestern U.S. (WST). p-values of the linear fitting are shown at the corner of each plot.

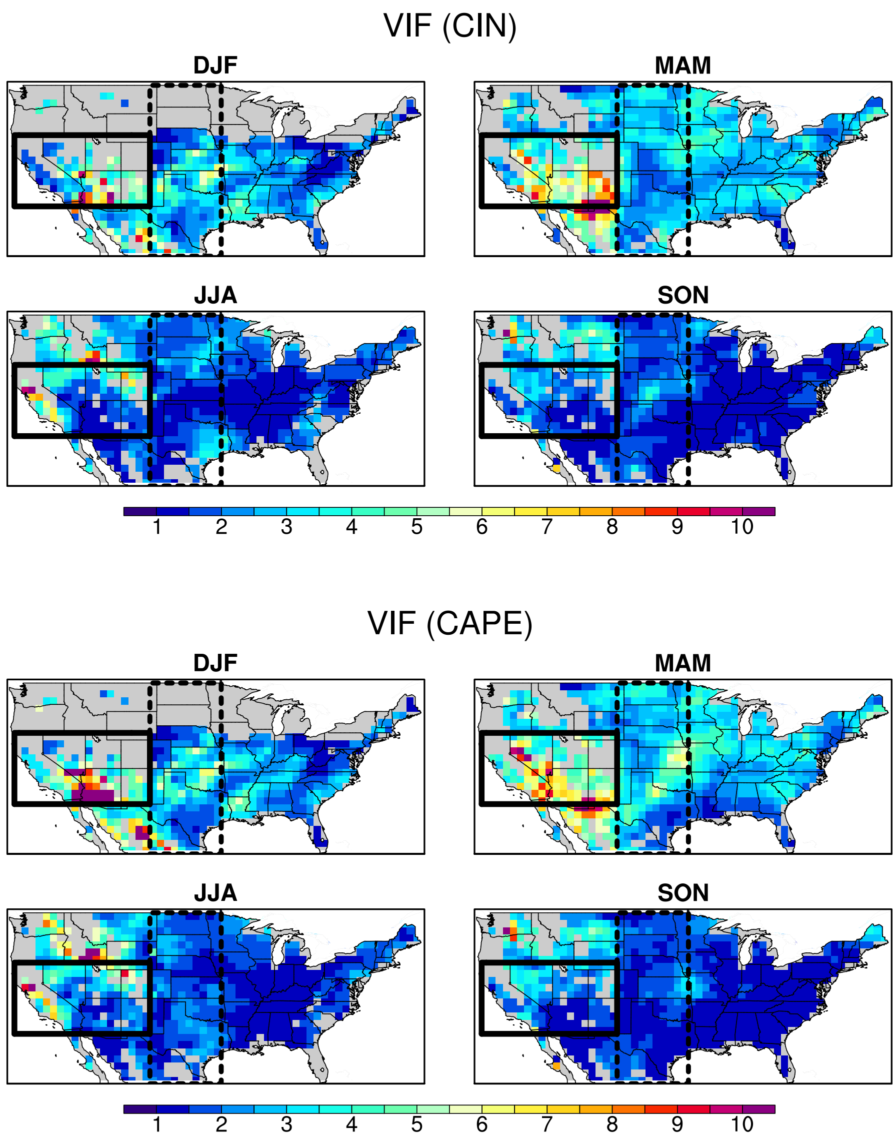


Figure S6. Variance inflation factor (VIF) for CIN (top) and CAPE (bottom) in five-factor multiple-linear regression with fine dust. Black boxes denote the southwestern U.S. (solid) and the Great Plains (dashed).

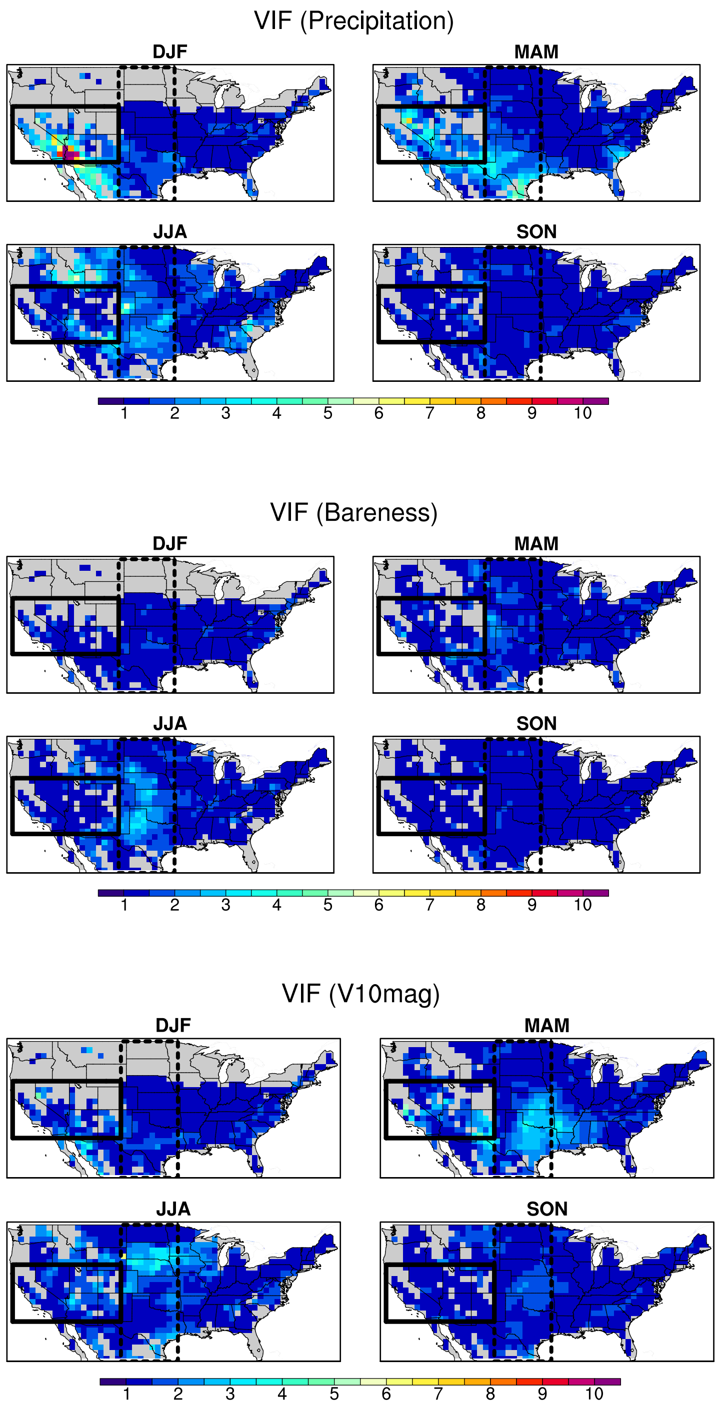


Figure S7. Same as Fig. S6 but for precipitation (top), surface bareness (middle), and surface wind (bottom).

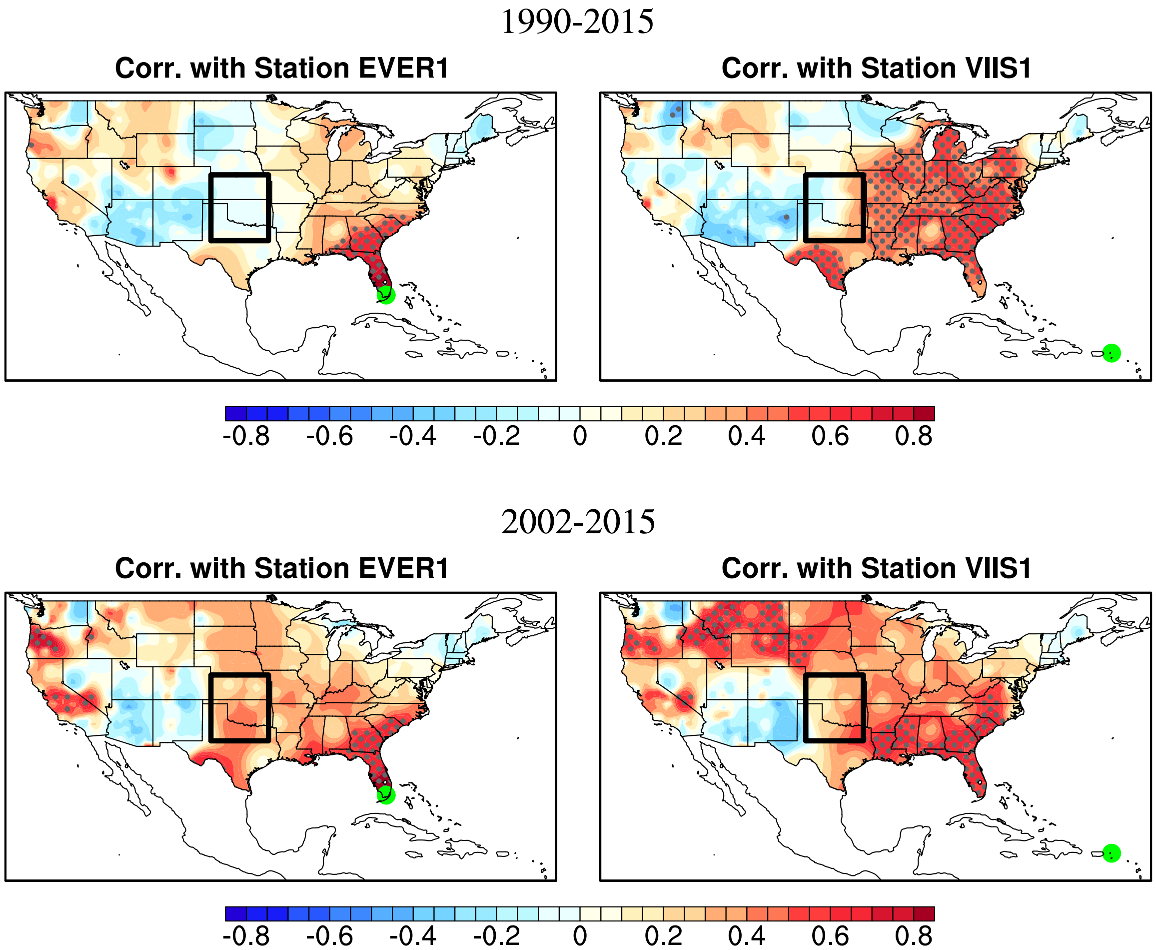


Figure S8. Correlations between fine dust concentration at EVER1 (25.4°N, 80.7°W, in the Everglades National Park) and VIIS1 (18.3°N, 64.8°W, in the Virgin Islands National Park) stations (denoted by green dots) with gridded fine dust concentration in the U.S. for JJA during 1990-2015 (top), and 2002-2015 (bottom). Areas significant at the 95% confidence level are dotted. Black box denotes the CGP area.