

1 **Title:** Increasing co-occurrence of fine particulate matter and ground-level ozone extremes in the
2 western United States

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18
19 **Abstract:**

20 Wildfires and meteorological conditions influence the co-occurrence of multiple harmful air
21 pollutants including fine particulate matter (PM_{2.5}) and ground-level ozone. We examine the
22 spatiotemporal characteristics of PM_{2.5}/ozone co-occurrences and associated population exposure
23 in the western U.S. The frequency, spatial extent, and temporal persistence of extreme
24 PM_{2.5}/ozone co-occurrences have increased significantly between 2001-2020, increasing annual
25 population exposure to multiple harmful air pollutants by ~25 million person-days/year. Using a

26 clustering methodology to characterize daily weather patterns, we identify significant increases
27 in atmospheric ridging patterns conducive to widespread PM_{2.5}/ozone co-occurrences and
28 population exposure. We further link the spatial extent of co-occurrence to the extent of extreme
29 heat and wildfires. Our results suggest an increasing potential for co-occurring air pollution
30 episodes in the western US with continued climate change.

31 **One-Sentence Summary:** More frequent atmospheric ridging and wildfires increase exposure of
32 western US population to co-occurring air pollutants.

33

34 **Main Text:**

35

36 INTRODUCTION

37 Air pollution is an urgent global health problem, and one that has gained additional attention
38 during the COVID-19 pandemic due to the exacerbating effects of pollutant exposure on
39 infectious disease spread and mortality (1-3). Two main air pollutants - fine particulate matter
40 (PM_{2.5}, defined as particulate matter with diameter of ≤ 2.5 μm), and ground-level ozone
41 (hereafter, 'ozone') - are linked to significant human health concerns including cardiovascular
42 and respiratory illnesses and mortality (4-7). PM_{2.5} and ozone have also been linked to negative
43 ecosystem impacts via their detrimental effects on plants and the broader environment (8-10).
44 Although few studies have quantified the compounding health impacts of co-occurring PM_{2.5} and
45 ozone, existing research indicates that simultaneous exposure to both pollutants can have
46 disproportionately more severe health impacts beyond the individual effect of either pollutant
47 (11, 12).

48 Wildfires can cause simultaneous increases in both pollutants through the direct emission
49 of PM_{2.5} (13, 14) and ozone precursor compounds (15-18) in smoke plumes, and recent research
50 has shown that ozone concentrations in urban areas in the western United States (US) can be
51 enhanced in the presence of wildfire smoke (19-21). During years of limited wildfire activity,
52 most of the western US experienced annual maximum PM_{2.5} concentrations during the cool
53 season when stagnant air conditions are typically prevalent (22). This seasonality would typically
54 minimize co-occurrence risk with high ozone concentrations, which peak during the warm
55 season when hot and dry conditions facilitate the formation and build-up of ozone (23).
56 Summertime wildfires therefore present a mechanism for PM_{2.5} extremes to occur at a time of

57 year when ozone levels are seasonally high, leading to increased chances of elevated
58 concentrations of both air pollutants occurring simultaneously.

59 Smoke from the unprecedented wildfire activity in the western US during August and
60 September of 2020 contributed to several weeks of extremely hazardous air quality over a large
61 area (3, 24). Similar conditions, although on a smaller scale, occurred during the 2015, 2017 and
62 2018 wildfire seasons (25, 26), and extensive wildfire smoke affected the region again in 2021.
63 Such widespread hazardous conditions acutely affected vulnerable communities in the region –
64 those at enhanced risk due to socio-economic or demographic factors and underlying health
65 conditions – contributing to an increased burden on the healthcare system through increased
66 hospitalizations and emergency department visits (26). In addition, recent research has linked
67 wildfire smoke in 2020 to higher risk and mortality associated with COVID-19 in many western
68 states (3). As recurrent and prolonged exposure to air pollution can exacerbate the public health
69 impacts of wildfire smoke (27–31), recent wildfire seasons have thus raised significant concerns
70 regarding the trajectory of air quality in the region.

71 Historical and projected climate and wildfire trends in the western US both point toward
72 increasing risk of exposure to poor air quality. Increased wildfire activity has already contributed
73 to rising extreme PM_{2.5} concentrations in fire-prone regions of the western US (32, 33), offsetting
74 national-level air quality improvements following the Clean Air Act. Indeed, wildfires have
75 contributed up to 50% of annual PM_{2.5} in parts of the western US in recent years (34). Annual
76 burned area across the western US has experienced exponential growth in recent decades (35,
77 36), partially due to drying of vegetation in the region tied to anthropogenic climate change (37–
78 39). These observed trends are projected to continue in a warming climate (24, 40–42).

79 Long-term climate and daily-scale meteorological conditions both influence the
80 formation, accumulation, and transport of air pollutants. Large-scale high-pressure systems (or
81 ‘ridges’) during the summer enhance surface temperatures, promote air stagnation, and can
82 contribute to both increased wildfire activity and ozone production in the western US (23).
83 Previous work has shown that such high-pressure systems are expected to increase in frequency
84 and persistence due to climate change (43, 44), raising the potential for increased warm-season
85 co-occurrence of PM_{2.5}/ozone extremes in the future. These conditions are amplified in the
86 western US by topography that promotes air stagnation in populated regions adjacent to fire-
87 prone lands (e.g. the Los Angeles Basin and the Willamette Valley near Portland).

88 Despite rising public health and air quality concerns, the influence of increasing wildfire
89 activity and changing meteorology on widespread hazardous air quality conditions across the
90 geographic extent of the western US has not yet been investigated. Schnell and Prather (45)
91 systematically demonstrated the influence of meteorology on the co-occurrence of PM_{2.5}, ozone,
92 and temperature extremes over eastern North America. Western North America, however, has
93 fundamentally different seasonality and drivers of these pollutants, and previous studies
94 investigating air pollutant co-occurrences have been restricted to urban areas [e.g. (19-21)].
95 Understanding how regional factors influence air pollutant characteristics and contribute to their
96 changing risks is critical for assessing their public health impacts and anticipating future trends
97 associated with climate variability and change.

98 Given the compounding human health impacts of air pollutant co-occurrences, we
99 investigate the influence of wildfires and meteorological factors on the spatial and temporal
100 characteristics of extreme PM_{2.5}/ozone co-occurrences across the western US and assess the
101 associated population exposure. Using gridded 1° x 1° datasets of observed PM_{2.5} and ozone

102 developed by Schnell et al. (46) and atmospheric reanalyses, we (1) quantify trends in the
103 frequency, persistence, and extent of widespread co-occurrence of PM_{2.5}/ozone extremes across
104 the western US in the past two decades, (2) identify the large-scale atmospheric patterns
105 associated with widespread co-occurrences and population exposure, (3) examine trends in
106 atmospheric patterns that amplify or mitigate co-occurrence risk across the region, and (4)
107 investigate the relationship between the geographic extent of co-occurrence, wildfire activity and
108 extreme heat during and preceding widespread PM_{2.5}/ozone co-occurrences. We also investigate
109 these factors in the context of the exceptional widespread and long-lasting co-occurrence episode
110 during the record-breaking 2020 wildfire season.

111

112 **RESULTS**

113 **Increasing trends in the spatial and temporal characteristics of PM_{2.5}/ozone co-occurrence**

114 Extremes in individual air pollutant concentrations are defined at each grid cell as exceedances
115 of their annual 90th percentiles (~37 days each year). We find that the simultaneous, spatially co-
116 located occurrence of local PM_{2.5} and ozone extremes (hereafter ‘co-occurrence’) has become
117 significantly more frequent over large areas of the western US during the late-summer wildfire
118 season - July-September - between 2001-2020, driven largely by the changing seasonality of
119 extreme PM_{2.5} concentrations (Fig. 1). High PM_{2.5} concentrations typically peaked during cool-
120 season months across much of this region during the early 2000s (Fig. S1A). However, the
121 fraction of the annual PM_{2.5} extremes occurring during July-September has increased
122 significantly in the past two decades (Fig. 1B). Parts of the region experienced a >80% increase
123 in this fraction, indicating that in these grid cells a majority of PM_{2.5} extremes are now
124 concentrated during this season that previously rarely experienced PM_{2.5} extremes. In contrast,

125 ozone concentrations typically peak during warm-season months (Fig. S1C-D) and the fraction
126 of annual ozone extremes occurring during July-September remains largely unchanged with the
127 exception of small decreases over parts of the Rocky Mountains, High Plains, and coastal
128 California (Fig. 1C). Therefore, the increased occurrence of PM_{2.5} extremes during a time of year
129 when ozone concentrations are seasonally high has largely driven the observed increases in
130 PM_{2.5}/ozone co-occurrence during the late-summer wildfire season across the western US (Fig.
131 1A).

132 In addition to occurring more frequently, local PM_{2.5}/ozone co-occurrences are
133 increasingly occurring across a larger geographic region simultaneously. The maximum daily
134 fraction of western US grid cells with simultaneous PM_{2.5}/ozone co-occurrence during July-
135 September has more than doubled (from 18.9% to 44.6%) over the past two decades, with an
136 increasing trend of ~1.35% per year (*p-value* = 0.02) (Fig. 1D). The largest spatial extents of co-
137 occurrence were observed in 2015, 2017, 2018 and 2020 – coincident with hot, dry summers and
138 widespread fire activity, including the largest burned areas across the western US wildland-urban
139 interface (20, 24, 25, 29, 36, 47). Increases in the frequency and spatial extent of co-occurrences
140 are associated with an increasing trend in July-September population exposure of ~24.9 million
141 person-days per year (*p-value* < 0.001) in the western US during 2001-2020 (Fig. 1E).
142 Cumulative population exposure over the season to PM_{2.5}/ozone co-occurrences exceeded 600
143 million person-days during the 2017, 2018, and 2020 wildfire seasons (Fig. 1E). Daily
144 population exposure exceeded 35 million people during the most widespread air pollution
145 conditions in these three seasons, peaking at ~46 million people (>50% of the western US
146 population) on August 21st, 2020 (Table S1).

147 Widespread PM_{2.5}/ozone co-occurrences, defined as days on which at least 25% of grid
148 cells covering the western US simultaneously experience local PM_{2.5}/ozone co-occurrence, have
149 occurred almost exclusively during July-September (72 of 75 total days; Fig. S2). Widespread
150 co-occurrences have become significantly more frequent and persistent (Fig. 2A-B), with an
151 increase of ~12.4 widespread co-occurrence days over 2001-2020 and the longest consecutive-
152 day occurrence persisting for an additional ~6.2 days. The frequency of widespread co-
153 occurrences was highest during the recent active wildfire seasons (Fig. 2A; Fig. S3). Of the 72
154 July-September widespread co-occurrence days during 2001-2020, 59 occurred during 2015,
155 2017, 2018, and 2020. In addition, the longest persistence (12 consecutive days) of widespread
156 PM_{2.5}/ozone co-occurrences on record occurred in 2020 (Fig. 2B), during which the daily
157 maximum extent of co-occurrence peaked at ~68.5% of the western US on August 24th, 2020
158 (Fig. 1D; Table S1).

159 During widespread co-occurrences the concentrations of both pollutants are elevated
160 relative to co-occurrence conditions of smaller geographic extent. Although co-occurrences are
161 defined as values above the local, annual 90th percentiles for both PM_{2.5} and ozone in each grid
162 cell, average observed concentrations on all widespread co-occurrence days exceeded the 95th
163 percentile for PM_{2.5} and the ~97th percentile for ozone across all grid cells experiencing local
164 PM_{2.5}/ozone co-occurrence (Fig. 2C). These findings are consistent with Schnell and Prather
165 (45), who reported enhancements in PM_{2.5} and ozone concentrations over eastern North America
166 during large, multi-day pollution episodes well above the statistical thresholds used to define
167 individual extremes (e.g. 90th or 95th percentiles). During widespread co-occurrence days in
168 recent seasons (2015, 2017, 2018, and 2020), PM_{2.5} concentrations averaged across all
169 constituent grid cells experiencing PM_{2.5}/ozone co-occurrence exceeded the EPA regulatory limit

170 of 35 $\mu\text{g}/\text{m}^3$ on 13 individual days (Fig. 2D, orange markers), peaking at 47.7 $\mu\text{g}/\text{m}^3$ on
171 September 3rd, 2017 during a period of widespread fire activity and smoke conditions in the
172 western US (25). Ozone concentrations averaged across the same grid cells on these days ($n =$
173 13) ranged from 57-63 ppb (Fig. 2D, blue markers; see also Fig. S4 for average concentrations
174 during all co-occurrences). Although below the EPA regulatory limit of 70 ppb, the fact that such
175 high ozone concentrations were present when averaged over a large geographic area and for
176 prolonged periods in combination with widespread $\text{PM}_{2.5}$ regulatory exceedances illustrates the
177 magnitude of human and environmental exposure to harmful air pollutants during recent wildfire
178 seasons, the health impacts of which are emerging (3, 26, 30, 31).

179

180 **Increasing trends in atmospheric patterns conducive to co-occurrence**

181 Although wildfires are a key source of emissions of $\text{PM}_{2.5}$ and ozone precursor compounds
182 during the late-summer season, the spatial extent, local concentrations, and temporal persistence
183 of their co-occurrences are modulated by a suite of meteorological factors, including surface
184 temperature and atmospheric patterns (45). To understand if and how atmospheric patterns that
185 affect $\text{PM}_{2.5}$ /ozone co-occurrence characteristics are changing, we use a spatial clustering
186 approach known as Self-Organizing Maps (SOMs) (48, 49). Our SOM implementation
187 categorizes daily large-scale weather patterns during July-September into 12 representative
188 clusters (or ‘nodes’) based on 500-hPa geopotential height anomalies from the European Centre
189 for Medium-Range Weather Forecasts (ECMWF) ERA5 reanalysis product (1979-2020; refer to
190 *Materials and Methods*).

191 We quantify the number of widespread co-occurrence days and population exposure to
192 co-occurrence associated with each node (Table 1; Fig. S5), and identify the 6 SOM nodes with

193 the largest (Nodes 5, 9 and 10, hereafter ‘high-exposure nodes’) and smallest (Nodes 2, 3, and 4,
194 hereafter ‘low-exposure nodes’) PM_{2.5}/ozone co-occurrence risk (Fig. 3). High-exposure nodes
195 are characterized by widespread positive geopotential height anomalies (hereafter, ‘ridging’) and
196 high daily maximum surface temperature anomalies across the region, which are largely co-
197 located with those grid cells experiencing the highest number of local PM_{2.5}/ozone co-
198 occurrences during widespread co-occurrence days in that node (Fig. 3A-C). In contrast, low-
199 exposure nodes are characterized by widespread anomalously low geopotential heights, cooler
200 temperatures, and onshore airflow from the Pacific Ocean, providing critical natural ventilation
201 for this region and suppressing widespread co-occurrence risk (Fig. 3D-F) (50).

202 Large-scale atmospheric patterns represented by high-exposure nodes contributed 43 of
203 the 72 widespread co-occurrence days (~60%), despite accounting for only ~29% of all July-
204 September days since 2001, indicating an elevated risk of PM_{2.5}/ozone co-occurrence across the
205 region when such patterns occur. We find robust increases in the frequency and persistence of
206 high-exposure nodes since 1979. These nodes now occur on an additional ~14.2 days per year
207 during July-September (*p-value* < 0.001) and the longest persistence of these nodes is an
208 additional ~4.3 consecutive days longer (*p-value* = 0.008) compared to four decades ago (Fig. 4,
209 orange lines). While the frequency of nodes relates to the frequency of pollutant exposure, the
210 longer persistence of certain nodes can have additional impacts beyond that of single-day node
211 occurrences. For example, previous research has shown that high ozone concentrations are more
212 likely during prolonged, multi-day heat conditions than on single hot days (22, 45). Of the 29
213 remaining widespread co-occurrence days not associated with the high-exposure nodes, 21
214 occurred in conjunction with atmospheric patterns favorable for widespread smoke transport
215 across the region during periods of high wildfire activity (Nodes 1, 7 and 11; see Table 1 and

216 Fig. S5). In contrast to the high-exposure nodes, the combined frequency and multi-day
217 persistence of low-exposure nodes exhibit negative trends during 1979-2020, now occurring on
218 ~12.6 fewer days per year (p -value < 0.001) and the longest consecutive-day occurrence of these
219 nodes persisting for ~4.3 fewer days (p -value = 0.002) compared to four decades ago (Fig. 4,
220 blue lines).

221 Together, these results suggest that atmospheric patterns that are conducive to
222 widespread local PM_{2.5}/ozone co-occurrences and larger population exposure across the western
223 US are becoming more frequent and persistent during July-September. Recent active wildfire
224 seasons have occurred in conjunction with record frequency and persistence of the high-exposure
225 nodes (i.e. ridging), with the highest frequency since 1979 of 44 days observed in July-
226 September 2017 and longest persistence of 17 consecutive days observed from September 3rd-
227 19th, 2020 occurring simultaneously with historic wildfire activity across several western US
228 states (Fig. 4) (24). The observed increase in ridging has co-occurred with and likely amplified
229 increasing aridity and extent of wildfire burned area over the western US at least partially
230 associated with anthropogenic warming, posing compounding hazards to the region (37-39, 51).
231 Further, increased persistence of ridging during wildfire smoke conditions can exacerbate
232 ground-level pollution in topographically-constrained basins, as decreased sunlight and increased
233 atmospheric stability traps smoke and prolongs the air pollution conditions (52, 53). Conversely,
234 atmospheric patterns favoring decreased widespread PM_{2.5}/ozone co-occurrences across the
235 western US (i.e. negative geopotential height anomalies and onshore airflow) are appearing less
236 often and with shorter duration during the late-summer wildfire season.

237

238 **Case study: Widespread co-occurrence episode of August 2020**

239 The ‘exceptional’ 2020 wildfire season featured the second highest number of widespread
240 PM_{2.5}/ozone co-occurrence days across the domain, along with the longest consecutive-day
241 persistence of widespread co-occurrence (Fig. 2A-B), the single most widespread daily co-
242 occurrence extent (~68.5%) across the western US (Fig. 1D), and the highest cumulative
243 seasonal population exposure to all local PM_{2.5}/ozone co-occurrences of nearly 1 billion person-
244 days (Fig. 1E) in the 20-year observed record. Widespread wildfire activity and extreme
245 temperatures associated with atmospheric ridging both contributed to shaping the record multi-
246 day co-occurrence episode observed during the second half of August 2020 (Fig. 5).

247 To examine their influence in shaping the multi-day widespread air pollution episode, we
248 analyze the wildfire and meteorological conditions between August 15th and 29th, 2020. We find
249 a sharp increase in the spatial extent of locally defined PM_{2.5}/ozone co-occurrences immediately
250 following a peak in daily burned area aggregated over the western US and southwest Canada
251 (brown line, Fig. 5A). This increase in burned area was associated with an extremely anomalous
252 dry lightning outbreak that ignited hundreds of wildfires, leading to multiple large fires that
253 burned for several weeks in central and northern California (Fig. 5B) (24). Grid cells in large
254 areas of the interior western US, both near and downwind of fires, observed PM_{2.5}/ozone co-
255 occurrences on a majority of days (>7) during this 15-day period (Fig. 5B, shading). The grid
256 cells that experienced a high number of co-occurrences are largely co-located with areas where
257 wildfire smoke persisted during that period, which is identified by the National Oceanic and
258 Atmospheric Administration’s (NOAA) Hazard Mapping System (HMS) smoke product (Fig.
259 5B, contours). Notably, grid cells in northern Nevada immediately downwind of California fires
260 observed local PM_{2.5}/ozone co-occurrences on at least 12 days and smoke was observed on at
261 least 13 days of the 15-day episode. In addition, many grid cells in the interior western US

262 observed record warmest 15-day average of daily maximum temperatures since 1979, conditions
263 that likely enhanced ozone production and contributed to the widespread extent of PM_{2.5}/ozone
264 co-occurrences (Fig. 5C) (23).

265 Large-scale atmospheric patterns shaped multiple aspects of this air pollution episode,
266 including the high temperatures, wildfires, and smoke transport. Atmospheric ridging across the
267 western US resembling the pattern of the high-exposure nodes contributed to the hot, dry, and
268 stagnant air conditions conducive to wildfire ignition and pollutant accumulation from smoke
269 during the first five days of the episode (August 15th-19th) (Fig. 5A). More than 75% of the
270 western US experienced daily maximum temperature anomalies exceeding 1 standard deviation
271 (σ) on all 5 days. Following the large increase in burned area during this time, a shift to an
272 atmospheric pattern characterized by ridging centered in the interior West (Node 1, see Fig. S5A)
273 developed on August 20th and persisted for three days, resulting in southwest-to-northeast
274 atmospheric airflow in the western part of the domain (Fig. 5A). This pattern transported smoke
275 from California fires across large areas of the interior western US, contributing to an increase in
276 local PM_{2.5}/ozone co-occurrence extent from <30% to ~66% of the western US grid cells by
277 August 21st (blue line, Fig. 5A).

278 The remote transport of wildfire smoke containing multiple pollutants including PM_{2.5}
279 and ozone to areas experiencing record warm conditions and enhanced photochemical ozone
280 production (Fig. 5C) were critical to the widespread extent of this episode. Closer to active fires,
281 dense smoke blocks solar radiation and mitigates ozone production (19). In addition, previous
282 studies have noted that aged smoke is more conducive to downwind ozone production (e.g. [16,
283 20, 21]), promoting local PM_{2.5}/ozone co-occurrences in remote areas where smoke is
284 transported. However, the contribution of wildfire smoke to increased ozone concentrations, and

285 thus increased PM_{2.5}/ozone co-occurrences needs to be further understood. Buysse *et al.* (19)
286 found that the presence of wildfire smoke enhances ozone concentrations in urban areas of the
287 western US, particularly in smoke plumes away from fire sources with PM_{2.5} concentrations
288 below 50 µg/m³. Similarly, Brey and Fischer (21) and Gong *et al.* (20) noted general
289 enhancement of ozone concentrations on smoke days in the western US. However, they also note
290 distinct regional variation with some locations not observing increased ozone concentrations
291 during smoke conditions.

292

293 **Relationships between burned area, heat extremes, and PM_{2.5}/ozone co-occurrence**

294 The dynamics of the August 2020 widespread co-occurrence episode highlight the importance of
295 both meteorology and wildfire extent in shaping the extent of PM_{2.5}/ozone co-occurrences and
296 therefore, exposure. We thus further characterize this relationship between wildfire burned area,
297 meteorology, and the peak spatial extent of all temporally independent widespread co-occurrence
298 periods ($n = 21$; refer to *Materials and Methods*) (Fig. 6). Given its relevance for ozone
299 production, we specifically focus on relating daily maximum temperature anomalies to
300 PM_{2.5}/ozone co-occurrence extent.

301 Similar to the August 2020 episode, we find that the extent of wildfire activity and heat
302 affect the spatial extent of other PM_{2.5}/ozone co-occurrences. The largest extents exceeding 45%
303 of the western US (much larger than the threshold used to define a widespread co-occurrence
304 day) occurred in 2015, 2017, 2018, and 2020 and were associated with extensive wildfire activity
305 in the western US and southwest Canada (maximum daily burned area extent >650 km², Fig. 6A)
306 and widespread positive daily maximum temperature anomalies exceeding 1σ (maximum daily
307 extent ≥55% of the western US, Fig. 6B) in the 7 days preceding the peak spatial extent of

308 temporally independent widespread PM_{2.5}/ozone co-occurrence periods (refer to *Materials and*
309 *Methods*). Analyzing this relationship over these 21 independent co-occurrence spatial extent
310 peaks during 2003-2020, we find robust pairwise correlation between the PM_{2.5}/ozone co-
311 occurrence extent and lagged burned area ($r = 0.66$, $p\text{-value} = 0.001$) as well as co-occurrence
312 extent and 7-day lagged extent of anomalously high ($>1\sigma$) maximum temperature anomalies ($r =$
313 0.49 , $p\text{-value} = 0.02$). These findings emphasize the role of simultaneous widespread heat and
314 wildfire activity in shaping widespread PM_{2.5}/ozone co-occurrences, with high values of this
315 combination of contributing factors in four of the six most recent July-September seasons in the
316 western US.

317

318 **DISCUSSION**

319 **Summary**

320 Our analysis demonstrates an increasing risk of exposure of the western US population to more
321 frequent and persistent extreme PM_{2.5}/ozone co-occurrences, defined at each grid cell as the
322 simultaneous exceedance of the local annual 90th percentile concentrations of both pollutants,
323 during the late summer wildfire season. These trends are largely driven by PM_{2.5} extremes
324 shifting toward the summer associated with increased wildfire activity in recent years (24-26, 32-
325 39) and coinciding with the season of high ozone concentrations. PM_{2.5}/ozone co-occurrences are
326 also affecting larger areas, with more than a doubling of the maximum daily spatial extent
327 (18.9% to 44.6%) of the western US experiencing simultaneous local co-occurrences over the
328 past two decades. We find that increasing widespread pollutant co-occurrences are associated
329 with increasing wildfire activity and increasing occurrence of conducive atmospheric patterns.

330 The increase in widespread PM_{2.5}/ozone co-occurrences during July-September
331 highlights the role of increasingly severe and larger wildfires in contributing to compounding
332 public health hazards in the western US. Although wildfire smoke can be transported to this
333 region from remote areas including Alaska (54) and Siberia (55), we find a robust correlation
334 between burned area in the western US and adjacent southwest Canada and extent of local
335 PM_{2.5}/ozone co-occurrence across the western US (Fig. 6A). Years with the maximum extent of
336 co-occurrence (Fig. 1D, red line) and greatest frequency of widespread co-occurrence days (Fig.
337 2A) also experienced the highest mean burned area in the western US (see Fig. 7 in (35)). The
338 largest spatial extents of co-occurrence in the observed record have all occurred since 2015
339 during particularly active wildfire seasons, with record co-occurrence extent and persistence in
340 2020 coinciding with record-breaking wildfire activity in several western US states. Given its
341 ability to produce PM_{2.5} extremes at a time of year when ozone concentrations are seasonally
342 high, our results imply that increasing wildfire activity is a key mechanism by which
343 simultaneous occurrences of local PM_{2.5}/ozone extremes are increasing in the western US despite
344 declining background levels of these pollutants in response to the Clean Air Act (23, 32, 56, 57).

345 Our results emphasize that atmospheric ridging patterns can affect widespread
346 PM_{2.5}/ozone co-occurrences and associated population exposure by amplifying multiple direct
347 physical drivers and sources of air pollutants. In addition to promoting conditions that are
348 conducive for wildfires that produce multiple harmful air pollutants, persistent ridging results in
349 widespread heat and air stagnation that enhances ozone production. Indeed, we identify a
350 significant relationship between the extent of heat and local PM_{2.5}/ozone co-occurrences. Further
351 emphasizing the importance of meteorology in influencing population exposure to widespread
352 air pollution conditions, large-scale airflow around high pressure ridges can transport smoke and

353 associated pollutants to remote areas. The presence of such favorable meteorological conditions
354 was critical in shaping the 2020 widespread co-occurrence episode via record heat and
355 atmospheric patterns conducive to smoke transport.

356 The increasing frequency and persistence of ridging during the late-summer wildfire
357 season (Fig. 4) suggest an increased likelihood of the type of atmospheric conditions that
358 contributed to the August 2020 co-occurrence episode, if these trends continue. While recent
359 studies have shown an intensification of western US summer ridging since the 1980s using
360 atmospheric reanalysis (58) and tree-ring records (59), identification of trends in ridging
361 frequency and persistence over the western US prior to the present analysis had been restricted to
362 other seasons (60–62). Our findings of changing late-summer atmospheric patterns agree with
363 recent studies that have highlighted the role of increasingly warmer and drier summer seasons,
364 which are strongly favored by atmospheric ridging, across the western US in driving increased
365 wildfire burned area extent and severity (38, 39). Further, drought and extreme heat events
366 associated with persistent ridging can produce widespread dust and photochemical pollution-
367 related health impacts across the western US (63, 64), increasing the likelihood of compound
368 stressors upon human health.

369

370 **Limitations**

371 We note multiple caveats to our findings. First, the derived gridded datasets of PM_{2.5} and ozone
372 used in this study are based on a relatively sparse observational network in some parts of the
373 western US, which might result in uncertainties in identified trends in these areas. Enhancing
374 spatial coverage of the monitoring network is critical to get more accurate and finer-scale air
375 quality information, particularly over rural areas of the western US. While the PurpleAir network

376 is rapidly enhancing the PM_{2.5} observational coverage (65), it has notable measurement biases
377 and a similar low-cost network is not currently available for ozone. Second, we mainly focus on
378 identifying proximal relationships and do not directly link wildfire emissions with local
379 PM_{2.5}/ozone co-occurrences, and do not examine the dependence of pollutant and precursor
380 concentrations on burn severity or types of fuel burned in different landscapes. Although we
381 explicitly link the presence of wildfire smoke to local PM_{2.5}/ozone co-occurrences during the
382 widespread episode of August 2020 using the NOAA HMS product, we do not systematically
383 quantify the climatology of pollutant co-occurrences with or without presence of wildfire smoke
384 due to the limited record, and do not link all individual fires to pollutant co-occurrences. Third,
385 we investigate the relationship between the extent of PM_{2.5}/ozone co-occurrences and two main
386 drivers – widespread heat and wildfire burned area - without explicitly accounting for hot, dry
387 weather promoting further wildfires leading to enhanced co-occurrence extent. Hot temperatures
388 are a common underlying driver of both wildfire activity and ozone production across the
389 western US on different time scales (23), and high-resolution modeling would be required to
390 disentangle the individual contributions of heat and wildfire smoke to local PM_{2.5}/ozone co-
391 occurrences across this region.

392

393 **Implications**

394 In recent years, millions of people across the western US have been affected by
395 hazardous air quality conditions caused by wildfire smoke. Although PM_{2.5} concentrations are
396 greatest in dense smoke plumes near wildfires, we find an increase in local PM_{2.5}/ozone co-
397 occurrences over widespread areas of the western US not limited to the immediate proximity of
398 active fires. These results highlight the potential for increasing population exposure to

399 compounding human health stressors in fire-prone and remote regions, with projected increases
400 in wildfire activity, smoke, and conducive meteorological conditions (51, 66, 67). Although
401 more research is needed to assess the cumulative health outcomes of co-occurrences of
402 PM_{2.5}/ozone extremes as well other pollutants in wildfire smoke, it is very likely that these co-
403 occurring air pollution extremes have compounding public health impacts (29). Their impacts are
404 not only limited to the direct cardiovascular and pulmonary effects, but also extend indirectly to
405 physical and mental health consequences arising from disruptions to outdoor activity, exercise,
406 and normal social activities. Vulnerable communities in the western US that have limited access
407 to healthcare or other resources needed to cope with poor air quality, have livelihoods that
408 involve higher occupational exposure to polluted outdoor air, or have high rates of prevalence of
409 medical conditions that can exacerbate the effects of air pollution exposure are likely to face
410 increasing threats from such co-stressors. Understanding the likelihood and drivers of such co-
411 occurring hazards is, therefore, critical for protecting communities through improved planning
412 and management of human health impacts from projected warming, drying, and increasing
413 wildfire activity in the western US.

414

415 **MATERIALS AND METHODS**

416 **Datasets**

417 We use 1° x 1° gridded PM_{2.5} and ozone datasets spanning 2000-2020 for the United States
418 developed using the methods of Schnell et al. (46) and subset to the western US domain (125°W-
419 103°W, 31°N-49°N). These gridded datasets are derived from surface monitoring station data
420 provided by the United States Environmental Protection Agency's Air Quality System (AQS;
421 <https://www.epa.gov/aqs>, for PM_{2.5} and ozone), Canada's National Air Pollution Surveillance

422 Program (NAPS; <https://open.canada.ca/data/en/dataset/1b36a356-defd-4813-acea->
423 [47bc3abd859b](https://open.canada.ca/data/en/dataset/1b36a356-defd-4813-acea-47bc3abd859b); for PM_{2.5} and ozone), and the Clean Air Status and Trends Network (CASTNET;
424 <https://www.epa.gov/castnet>, for ozone). Validated AQS data are used for PM_{2.5}/ozone spanning
425 October 2000 - July 2019, with preliminary data sourced from the AirNow online portal
426 (<https://www.airnow.gov>) for August 2019 - September 2020. We use daily averages for PM_{2.5}
427 and the maximum daily 8-h average (MDA8) for ozone, reflecting the measures typically used
428 for regulatory purposes and health impacts. For ozone, the hourly measurements are interpolated
429 and MDA8 is calculated. For PM_{2.5}, daily averages are constructed prior to interpolation from
430 any hourly reporting stations, and the daily average values are interpolated. The interpolation
431 procedure is a hybrid inverse distance-weighted method that includes a declustering component
432 designed to limit the influence of multiple clustered, typically urban observations. Parameters for
433 the interpolation were optimized with a leave N-out cross-validation procedure. These gridded
434 datasets were originally developed for the purpose of evaluating global chemistry models for
435 their ability to simulate large-scale, multi-day air pollution episodes. They have also been used to
436 analyze large-scale PM_{2.5}, ozone, and extreme temperature co-occurrences in the eastern US
437 (45); thus, they are well-suited for similar analysis of PM_{2.5}/ozone co-occurrences across a large
438 geographic region herein. PM_{2.5}/ozone data are analyzed over two seasons - July-September of
439 the given year and October of the previous calendar year through June of the given year.

440 Meteorological data, consisting of 500-hPa geopotential heights and 2-m air temperature,
441 were obtained from the ECMWF ERA5 reanalysis
442 (<https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era5>) on the native 0.25° x
443 0.25° resolution (68, 69). For analyzing the co-location of wildfire smoke and PM_{2.5}/ozone co-
444 occurrence during the August 2020 case study, daily wildfire smoke polygons for August 15th-

445 29th, 2020, were obtained from NOAA’s National Environmental Satellite, Data, and Information
446 Service (NESDIS) Hazard Mapping System (HMS) smoke product
447 (<https://www.ospo.noaa.gov/Products/land/hms.html#data>) (19, 54). For each day, all polygons
448 representing smoke of any density were merged into a single polygon representing total smoke
449 coverage for that day (19), and were overlaid with the 1° x 1° grid of the PM_{2.5} and ozone
450 datasets. Any grid cell spatially co-located with any portion of a smoke polygon is categorized as
451 experiencing a ‘smoke-day’, enabling the computation of the total number of smoke-days during
452 the 15-day episode in each grid cell. For visualization in Fig. 5B, the gridded values of smoke-
453 day frequencies were interpolated to contours and smoothed with a Gaussian filter ($\sigma = 0.2$),
454 allowing for the preservation of large-scale spatial features of smoke-day counts while
455 minimizing visual noise induced by local-scale variation.

456 The Moderate Resolution Imaging Spectroradiometer (MODIS) Aqua+Terra Thermal
457 Anomalies/Fire Locations 1 km dataset (MCD14DL) was retrieved from NASA’s Fire
458 Information for Resource Management System (FIRMS) archive download portal
459 (<https://firms.modaps.eosdis.nasa.gov/download/>). The MCD14DL product is used to identify
460 the presence of wildfires (>95% confidence) in at least 50 1-km grid cells contained within each
461 of the larger 1° x 1° grid cells during the August 2020 widespread co-occurrence episode. The 50
462 km² threshold was chosen to isolate large fire occurrences (70), as these fires are presumed to
463 impact air quality on regional scales. To quantify the spatial extent of burned area in the western
464 US and adjacent southwest Canada (Canadian data subset to <60°N, >115°W), we use the
465 MODIS burned area product (2003-2020) (71).

466 We quantify population exposure to PM_{2.5}/ozone co-occurrences using estimated 2020
467 population counts from the Gridded Population of the World version 4 (GPWv4) dataset,

468 obtained on a 1° x 1° grid from Columbia University’s Socioeconomic Data and Applications
469 Center (SEDAC; <https://sedac.ciesin.columbia.edu>) (72). Western US population is defined as
470 the total population contained in all grid cells ($n = 375$) within the study domain, which includes
471 adjacent parts of the Great Plains and Mexico. We use person-days as a metric to quantify
472 population exposure to local PM_{2.5}/ozone co-occurrence. It is obtained by multiplying the
473 estimated 2020 population in each grid cell by the number of co-occurrences in that grid cell, and
474 then aggregating it across the domain. We consider a fixed population to isolate the influence of
475 changing physical hazards on changing exposure.

476

477 **Defining PM_{2.5}/ozone co-occurrences**

478 We seek to understand changes in simultaneous occurrence of extreme PM_{2.5} and ozone
479 concentrations, as co-occurrences of both pollutants have the potential to induce co-stressor
480 effects on human and environmental health. We therefore define extremes for PM_{2.5} and ozone at
481 each grid cell individually as the exceedances of the local 90th percentiles of their daily
482 concentrations (average daily value for PM_{2.5} and MDA8 for ozone) within each individual year.
483 Therefore, we examine the co-occurrence of the top ~37 PM_{2.5} and ozone extremes in each grid
484 cell for each year. Instead of a fixed threshold to define extremes over the study period, this
485 time-varying definition allows us to identify extremes relative to the overall improving air
486 pollution due to emission reductions and stricter national air quality standards. Further, having a
487 fixed number of individual occurrences of both pollutants in each year enables us to identify
488 years with anomalous temporal co-occurrences driven by factors other than their climatology.
489 Assuming independent distributions, in a given grid cell the joint probability of PM_{2.5}/ozone co-
490 occurrence each with a 10% chance of occurrence is 3.65 days per year, if co-occurrences are

491 truly random. However, nearly 86% of western US grid cells have a higher likelihood of co-
492 occurrence relative to random chance alone (Fig. S6), suggesting the role of common physical
493 drivers of such co-occurrences.

494

495 **Characterizing large-scale atmospheric patterns**

496 To investigate the influence of large-scale atmospheric patterns on local PM_{2.5}/ozone co-
497 occurrences, we use Self-Organizing Maps (SOMs) to cluster daily geopotential height
498 anomalies during July-September, 1979-2020 and identify typical atmospheric circulation
499 patterns. SOMs are a type of artificial neural network commonly used in the climate sciences for
500 spatial clustering of large-scale meteorological variables based on their similarity (48). The
501 number and arrangement of SOM nodes is a subjective choice and depends on the application
502 (49, 60). We test three SOM node configurations comprising 6 (2 x 3), 12 (3 x 4), and 20 (4 x 5)
503 nodes, to identify a configuration that minimizes similarity between clusters while also capturing
504 the range of patterns that occur in this region.

505 To help inform our SOM configuration selection, we examined two sets of spatial
506 correlation coefficients following Gibson et al. (73): 1) between each SOM node pattern and the
507 individual constituent patterns in that node ('node-field' correlation, higher values are optimal),
508 and 2) between every unique combination of node pairs ('node-node' correlation, lower values
509 are optimal). See Fig. S7 for the distribution of both sets of correlation coefficients. We selected
510 the 12-node (3 x 4) SOM configuration as the median node-field correlation is higher than in the
511 6-node configuration and the node-node correlation interquartile range is lower than in the 20-
512 node configuration. The improvement in node-field correlation in the 20-node configuration is
513 small (Fig. S7A), and this configuration qualitatively exhibits overlapping patterns due to the

514 larger number of nodes. While the 6-node configuration does have a larger distinction amongst
515 nodes (based on lower median; see Fig. S7B), it does not adequately represent the range of
516 geopotential height patterns seen in the 12-node configuration. For SOM training, we use 200
517 initial iterations and 800 final iterations, and set the initial neighborhood radius to 3 with a final
518 neighborhood radius of 1. SOM computation was performed using the MATLAB ‘SOM
519 Toolbox.’

520

521 **Examining relationships between wildfires, extreme heat, and co-occurrence extent**

522 Local co-occurrences of PM_{2.5} and ozone extremes are a result of complex interactions between
523 meteorology and wildfire smoke operating on multiple timescales. Our *a priori* assumption is
524 that long-range transport of wildfire smoke can take several days to cover a large geographic
525 extent of the western US. Further, our hypothesis is that multi-day heat waves can influence co-
526 occurrence extent through both promoting wildfire activity that can produce air pollutants in
527 following days, and through widespread photochemical production and accumulation of ozone.
528 To account for such interactions, we examine the relationship between antecedent fire and heat
529 conditions in the preceding week (7-day window) with local PM_{2.5}/ozone co-occurrence extent
530 on a given day. We estimate the correlation between wildfire burned area preceding peak co-
531 occurrence extent, and between the spatial extent of positive daily maximum temperature
532 anomalies preceding peak co-occurrence extent.

533 To isolate conditions antecedent to peaks in the spatial extent of widespread co-
534 occurrence, we extract the largest co-occurrence spatial extent in non-overlapping 15-day
535 windows. This is done iteratively in descending order of co-occurrence extent for all July-
536 September days during the period of overlap with burned area data (2003-2020). Starting with

537 the largest spatial extent (68.5% of the western US on August 24th, 2020), a 15-day window,
538 centered on that day, is used to exclude all other days in this window and this process is repeated
539 for each successive lower extent provided it is outside of all previous 15-day windows. This
540 process yields 21 widespread co-occurrence extent peaks (out of 72 total widespread co-
541 occurrence days; see Fig. S2) during July-September, 2003-2020, that we define as temporally
542 independent and use in the correlation analyses to examine the relationship between the extent of
543 burned area, heat and local PM_{2.5}/ozone co-occurrences.

544 The highest correlation between burned area and local PM_{2.5}/ozone co-occurrence extent
545 ($r > 0.65$) for these 21 peak spatial extents occurs for lags of -3 to -7 days (Fig. S8, blue line),
546 with peak correlation at -4 days ($r = 0.74$). The highest correlation between the extent of heat and
547 co-occurrence ($r > 0.49$) occurs for lags of -6 to -13 days (Fig. S8, orange line), with peak
548 correlation at -11 days ($r = 0.53$). We note that these lags are based on a relatively small number
549 of peak dates and the time of peak extent of local PM_{2.5}/ozone co-occurrences following heat and
550 fire conditions can vary for individual dates. Therefore, our use of the 7-day lagged window in
551 this analysis captures the overlapping period of high correlation of co-occurrence extent with
552 antecedent widespread heat conditions and burned area extent while accounting for differences in
553 the timing of individual extent peaks.

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765

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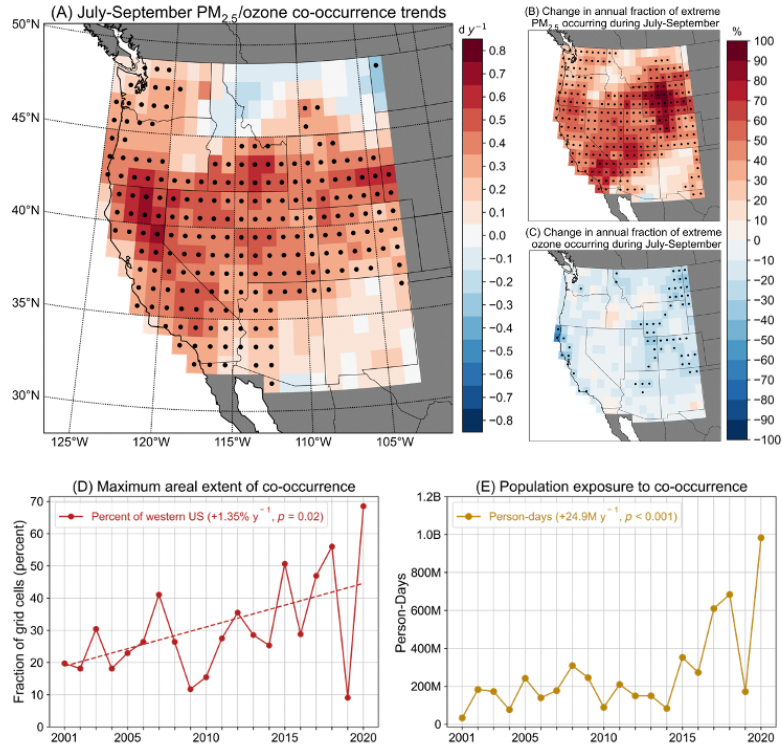
781 **Author contributions:**

782 All authors contributed to designing the research. D.A.K. conducted the analyses; and
783 D.A.K. and D.S. drafted the paper with feedback from all co-authors.

784 **Competing interests:** The authors declare that they have no competing interests.

785
786 **Data and materials availability:** Gridded PM_{2.5} and ozone data used in this study and all
787 other datasets used to perform analyses herein are available for download at the Zenodo
788 open-access repository (<https://zenodo.org/record/5174547#.YRK28O11B0t>). All source code
789 used to perform the analyses and create publication figures can be accessed at a dedicated

790 GitHub repository maintained by the corresponding author
791 (https://github.com/dmitri1357/pm25_o3_paper).



792

793 **Fig. 1. PM_{2.5}/ozone co-occurrence trends during 2001-2020 and population exposure. (A)**

794 Trends in the number of days ($d\ y^{-1}$) with PM_{2.5}/ozone co-occurrences at each grid cell during

795 July-September. Co-occurrences are defined as values of each pollutant exceeding their

796 respective local annual 90th percentile daily concentrations simultaneously. Trends in the annual

797 fraction of (B) PM_{2.5} extremes and (C) ozone extremes occurring at each grid cell during July-

798 September relative to rest of year (October – June). The maximum possible number of co-

799 occurrences is 37 per year in each grid cell, equal to the number of days above local annual 90th

800 percentile daily concentration values for each pollutant. Black dots denote statistical significance

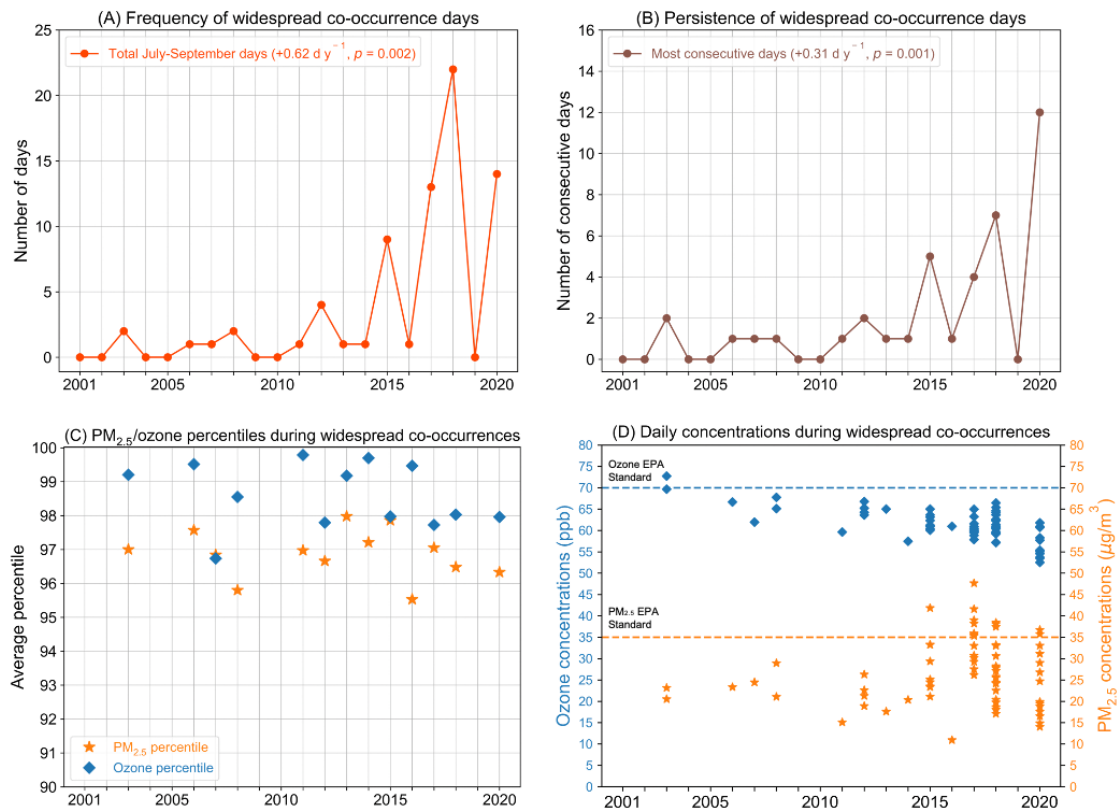
801 of trends at $p < 0.05$ based on a non-parametric permutation test. (D) Maximum daily extent of

802 western US grid cells simultaneously experiencing co-occurrences of local PM_{2.5}/ozone extremes

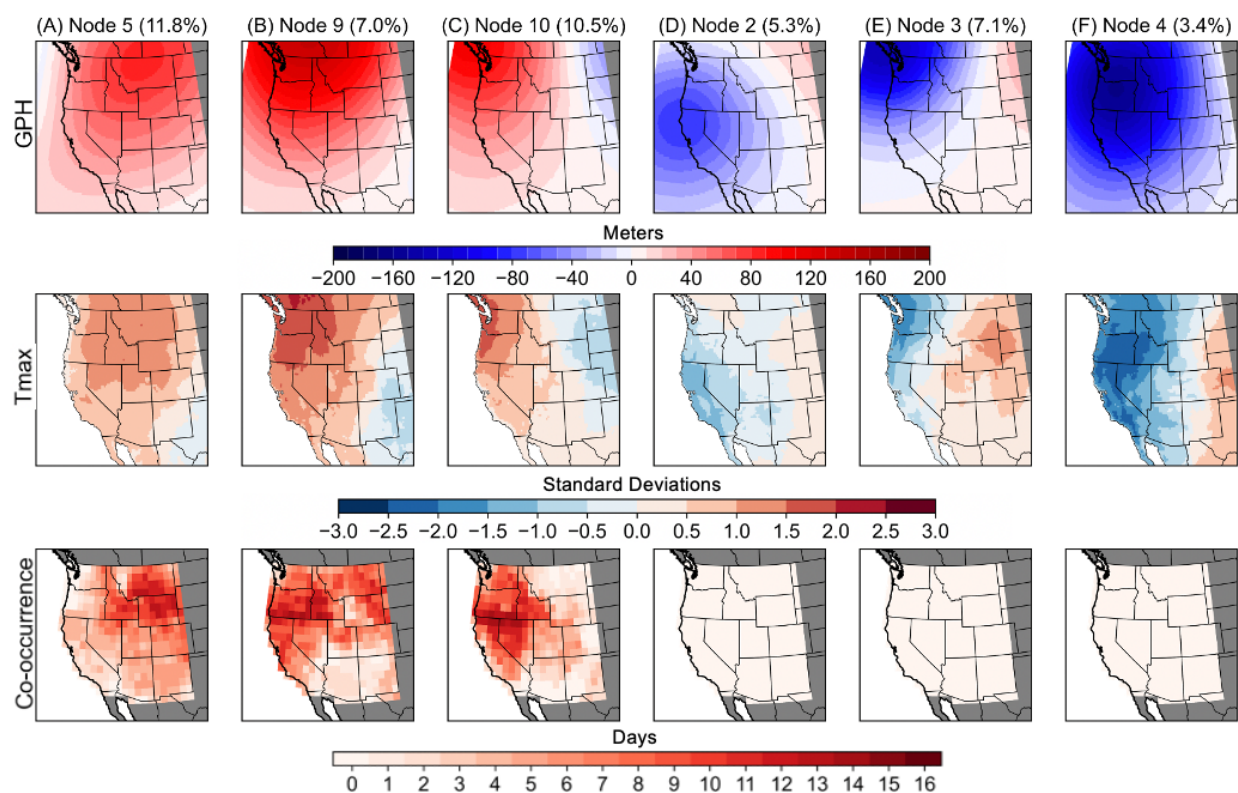
803 during July-September each year. (E) Total population exposure to all local PM_{2.5}/ozone co-

804 occurrences during July-September measured in million person-days (M) per year. Text in panels

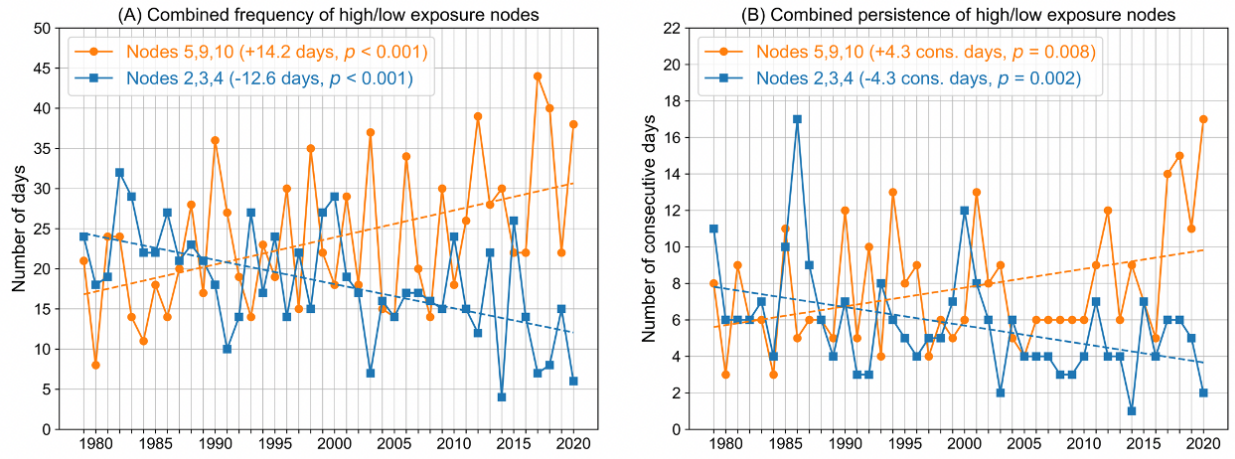
805 (D) and (E) indicates the linear trends and p -values based on a permutation test.



807
 808 **Fig. 2. Widespread PM_{2.5}/ozone co-occurrences.** Timeseries of (A) the total number and (B)
 809 longest consecutive-day persistence of widespread July-September co-occurrence days, defined
 810 as days with simultaneous local PM_{2.5}/ozone co-occurrence in $\geq 25\%$ of western US grid cells.
 811 Text in panels (A) and (B) indicates linear trends (d y^{-1}) with p -values based on a permutation
 812 test. Characteristics of the individual pollutants during widespread co-occurrences are shown
 813 through (C) percentiles of PM_{2.5} and ozone daily concentrations averaged across all affected grid
 814 cells, and (D) pollutant concentrations averaged across affected grid cells on widespread co-
 815 occurrence days ($n = 72$). Note percentiles in panel (C) are calculated based on the distribution
 816 of concentrations in each year (refer to *Materials and Methods*). Dashed lines in panel (D) show
 817 concentrations corresponding to the EPA regulatory health standards for each pollutant (70 ppb
 818 for ozone and $35 \mu\text{g}/\text{m}^3$ for PM_{2.5}).



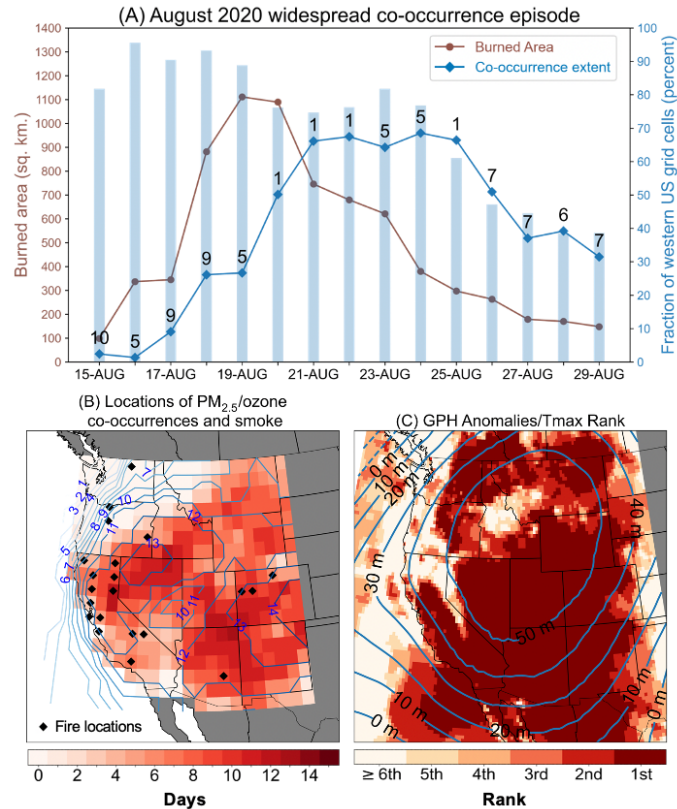
819
 820 **Fig. 3. The 6 SOM nodes with the largest (left three columns) and smallest (right three**
 821 **columns) PM_{2.5}/ozone co-occurrence risk. (Top) Geopotential height (GPH) anomalies for each**
 822 **SOM node trained over 1979-2020. (Middle) Composite standardized anomalies of daily**
 823 **maximum temperatures (Tmax) on all days associated with each node during the 2001-2020**
 824 **period. (Bottom) Number of times each grid cell experienced local PM_{2.5}/ozone co-occurrences**
 825 **during all widespread co-occurrence days associated with that node. The maximum possible**
 826 **number of co-occurrence days in a given grid cell is equivalent to the total number of widespread**
 827 **co-occurrence days associated with that node (Node 5: 14 days, Node 9: 16 days, Node 10: 13**
 828 **days; Table 1). Values in parentheses on top row indicate the frequency of each SOM node**
 829 **relative to all July-September days during the period of overlap with air pollution data (2001-**
 830 **2020).**
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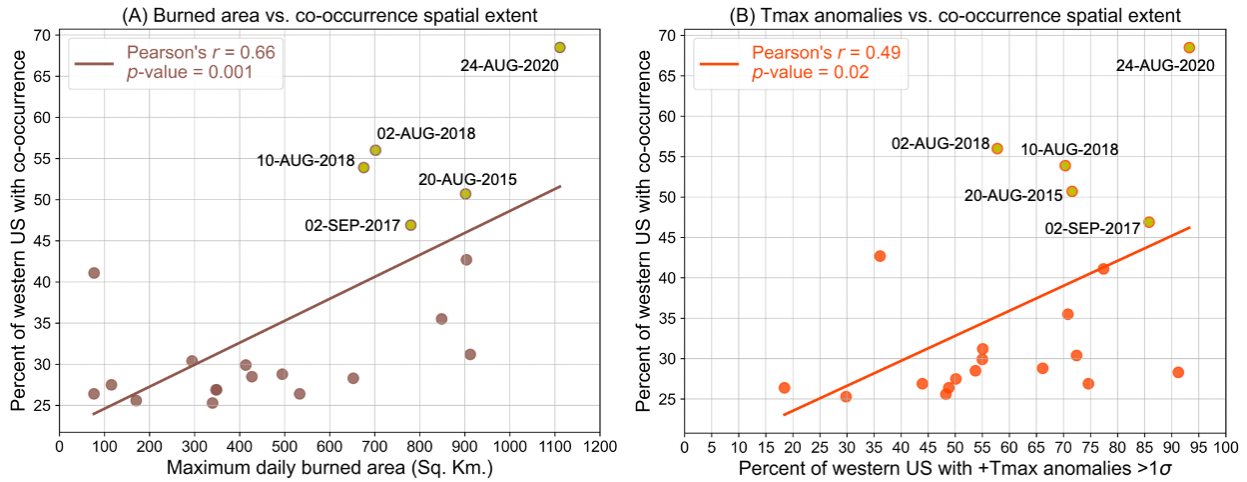
833 **Fig. 4. Frequency and persistence of high/low exposure nodes.** Timeseries of combined (A)
 834 total number of days and (B) longest multi-day persistence for high-exposure SOM nodes 5, 9,
 835 10 (orange lines) and low-exposure SOM nodes 2, 3, 4 (blue lines), during July-September 1979-
 836 2020. In both plots, dashed lines show linear trends with numbers indicating corresponding
 837 changes over the 42-year period and p -values of the linear trends based on a permutation test.

838



839
 840 **Fig. 5. Widespread co-occurrence episode during August 15th-29th, 2020.** (A) Timeseries of
 841 daily burned area from MODIS in the western US and southwest Canada (brown line), fraction
 842 of western US grid cells with local PM_{2.5}/ozone co-occurrence (blue line), and fraction of
 843 western US grid cells with daily maximum temperature (Tmax) anomalies exceeding 1 standard
 844 deviation above local daily climatology (blue bars). Numbers on the blue line indicate the best-
 845 matching SOM node for that day's atmospheric pattern. Note that widespread co-occurrence
 846 conditions begin on August 18th and persist through the 29th. (B) Total number of local
 847 PM_{2.5}/ozone co-occurrence days at each grid cell (shaded) and total number of days with
 848 presence of smoke from NOAA's Hazard Mapping System (contours) between August 15th-29th.
 849 Black markers indicate presence of wildfires from MODIS in at least 50 1-km grid cells
 850 contained within each of the 1° x 1° grid cells of the PM_{2.5}/ozone data. (C) Average geopotential
 851 height (GPH) anomalies (contours every 10 meters, as in Fig. 3 top row) and rank of the average

852 Tmax during August 15th-29th, 2020, compared to all other similar two-week periods during
 853 1979-2019 (shaded). The darkest red shading indicates that in 2020 those grid cells experienced
 854 their hottest average Tmax within the 42-year ERA5 dataset.



855 **Fig. 6. Relationship between widespread PM_{2.5}/ozone co-occurrence extent, wildfire burned**
 856 **area and daily maximum temperatures.** Scatterplots showing the spatial extent of the western

857 **US affected by PM_{2.5}/ozone co-occurrence with (A) 7-day lagged MODIS burned area in the**
 858 **western US and southwest Canada, and (B) 7-day lagged spatial extent of positive maximum**
 859 **temperature (+Tmax) anomalies >1 standard deviation above local daily climatologies in the**
 860 **western US during the period of overlap with available burned area data (2003-2020). For both**
 861 **burned area and +Tmax, the values represent the maximum daily extent in the 7 days preceding**
 862 **the peak spatial extent of PM_{2.5}/ozone co-occurrences. Only temporally independent widespread**
 863 **co-occurrence extent peaks during July-September are included ($n = 21$; refer to *Materials and***
 864 ***Methods*). Dates for the top 5 largest extent peaks are shown. Text in panels indicates Pearson**
 865 **correlation coefficients (r) and p -values for the pairwise relationships.**

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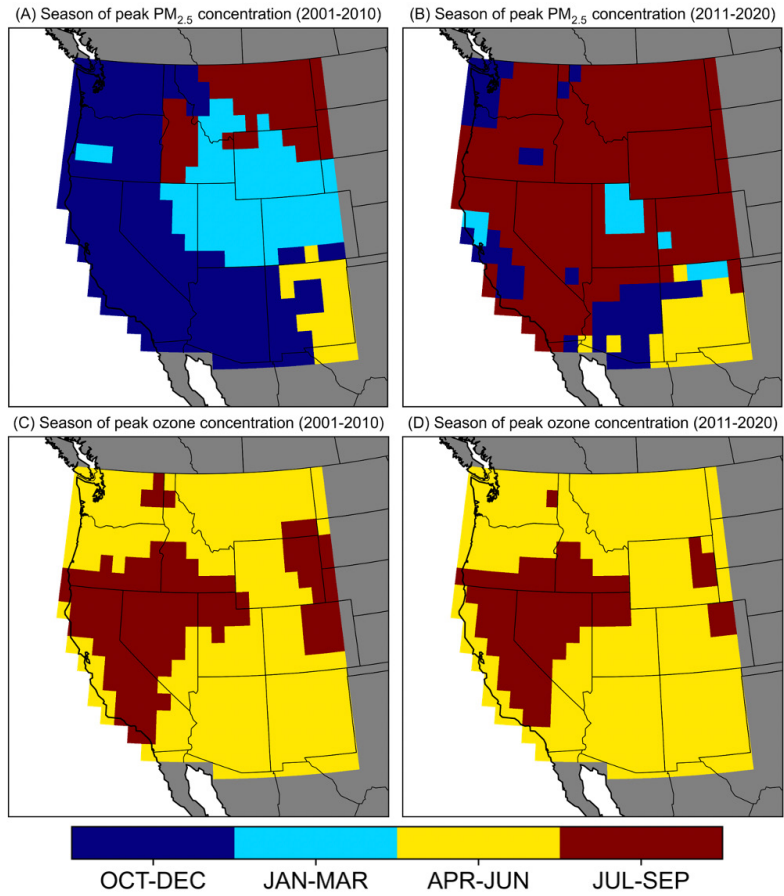
870 **Table 1. Summary statistics for all 12 nodes of the Self-Organizing Map (SOM).**

871 PM_{2.5}/ozone co-occurrence data represents all July-September days from 2001-2020. Asterisks

872 indicate statistical significance of node trends (1979-2020) at $p < 0.05$.

| SOM Node | Number of Days | Cumulative PM _{2.5} /ozone co-occurrence exposure in million person-days | Number of widespread PM _{2.5} /ozone co-occurrence days ($n = 72$) | 42-year change in SOM node frequency (days/year) | 42-year change in maximum SOM node persistence (consecutive days/year) |
|-----------|----------------|---|---|--|--|
| 1 | 151 | 441.7 | 5 | -1.1 | -0.6 |
| 2 | 98 | 78.2 | 0 | -6.4* | -3.5* |
| 3 | 130 | 77.9 | 0 | -2.0 | -0.2 |
| 4 | 63 | 4.4 | 0 | -4.2* | -1.7 |
| 5 | 217 | 835.7 | 14 | 5.8* | 1.2 |
| 6 | 198 | 529.2 | 6 | 0.0 | -0.1 |
| 7 | 251 | 650.8 | 8 | 2.7 | 0.7 |
| 8 | 110 | 122.6 | 2 | -2.2 | -1.0 |
| 9 | 129 | 866.7 | 16 | 2.2 | 0.6 |
| 10 | 194 | 847.2 | 13 | 6.2* | 2.1* |
| 11 | 179 | 739.5 | 8 | 2.8 | 0.6 |
| 12 | 120 | 141.0 | 0 | -3.7 | -0.7 |

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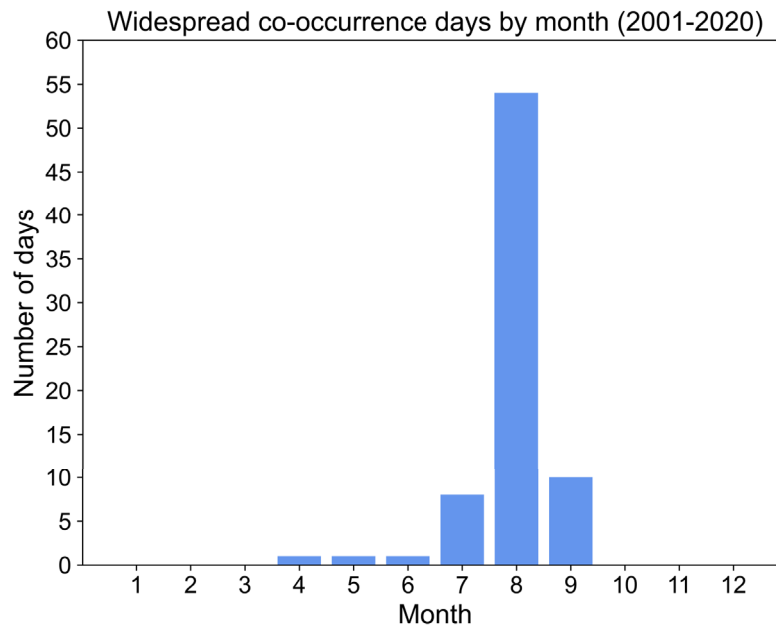
875 **Fig. S1. Peak seasons of mean $PM_{2.5}$ and ozone concentrations.** Seasons of peak average

876 concentrations of $PM_{2.5}$ (top) and ozone (bottom) during 2001-2010 (left) and 2011-2020 (right)

877 at each grid cell.

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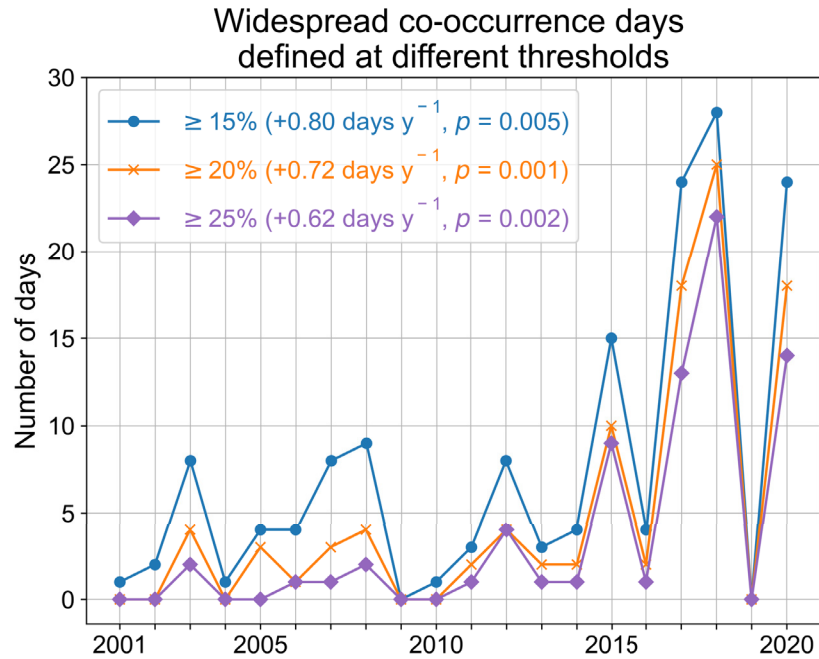
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882 **Fig. S2. Seasonality of widespread co-occurrence days.** Total number of widespread
883 PM_{2.5}/ozone co-occurrence days ($\geq 25\%$ of the western US) during each calendar month, 2001-
884 2020.

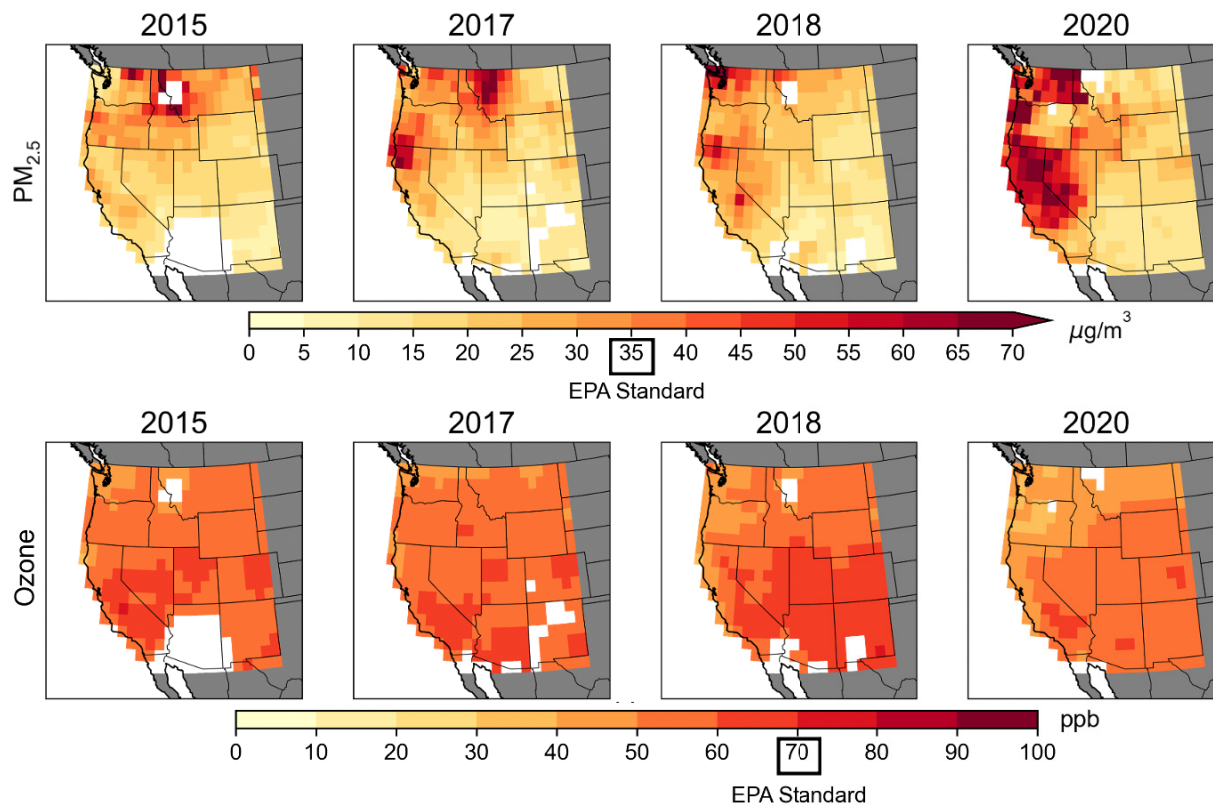
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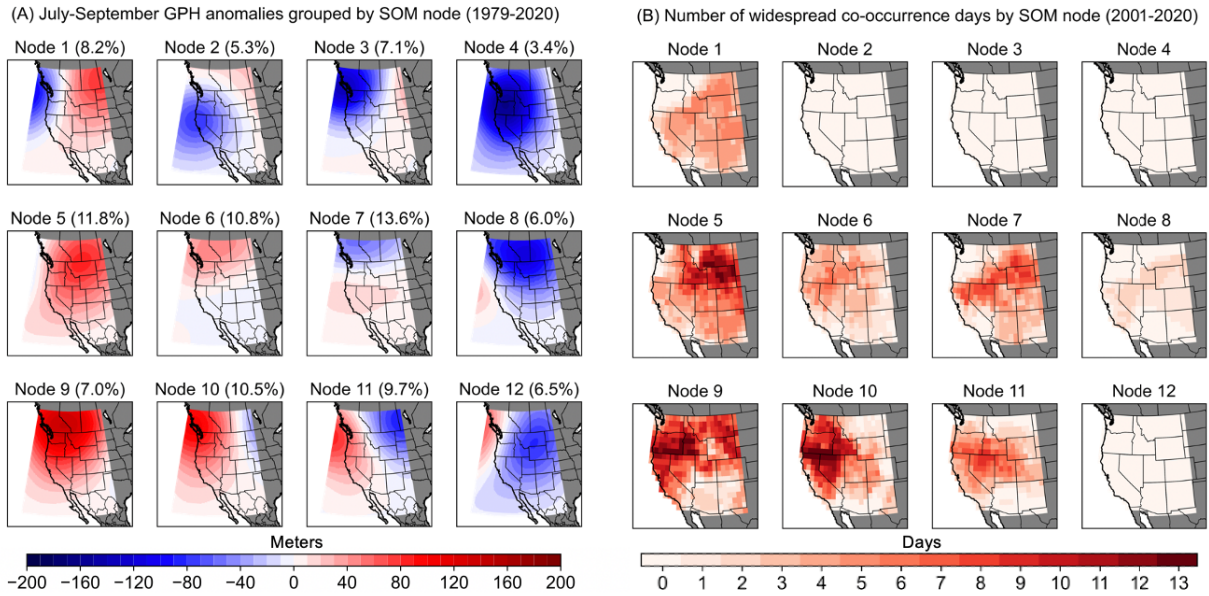
887 **Fig. S3. Widespread PM_{2.5}/ozone co-occurrence days during July-September at different**
 888 **thresholds.** Days with simultaneous local PM_{2.5}/ozone co-occurrences affecting ≥15% (green),
 889 ≥20% (brown), and ≥25% (blue) of western US grid cells, 2001-2020. Text indicates annual
 890 linear trends and *p*-values based on a non-parametric permutation test.

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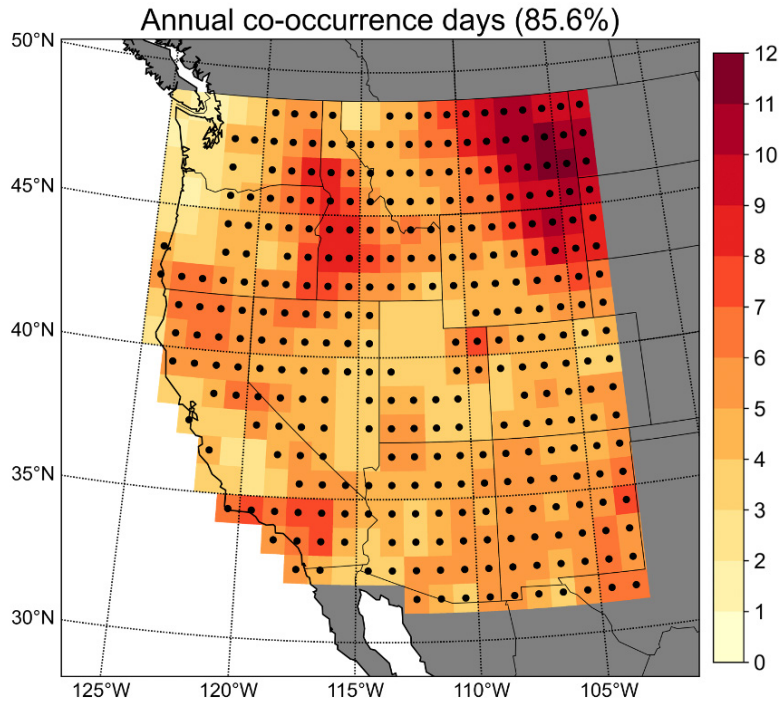
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 894 **Fig. S4. PM_{2.5} and ozone concentrations on co-occurrence days.** Average concentrations of
 895 PM_{2.5} (top row) and MDA8 ozone (bottom row) on all local co-occurrence days during July-
 896 September of 2015, 2017, 2018, and 2020. Corresponding EPA regulatory health standards are
 897 35 $\mu\text{g}/\text{m}^3$ for PM_{2.5} and 70 ppb for ozone. White shading indicates that no co-occurrence days
 898 were recorded in those grid cells during July-September of that year.

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906 **Fig. S5. All 12 nodes of the Self-Organizing Map (SOM).** (A) Geopotential height (GPH)
 907 anomalies for each SOM node trained over 1979-2020. (B) Number of widespread PM_{2.5}/ozone
 908 co-occurrence days (2001-2020) associated with each node. Values in parentheses in plot (A)
 909 indicate the frequency of each SOM node relative to all July-September days during the period
 910 of overlap with air pollution data (2001-2020).

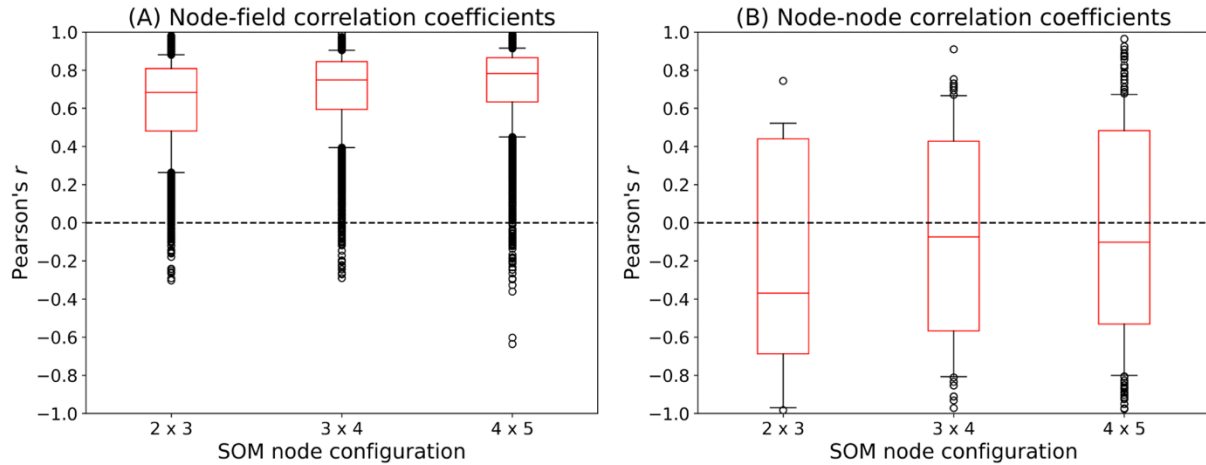
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914 **Fig. S6. Annual frequency of co-occurrence days.** Average number of local annual
 915 $PM_{2.5}$ /ozone co-occurrence days at each grid cell. Black dots denote grid cells averaging more
 916 than 3.65 days/year, the number expected by random chance from a joint probability distribution.
 917 Value in parentheses indicates the percentage of western US grid cells averaging more co-
 918 occurrence days than expected by random chance.

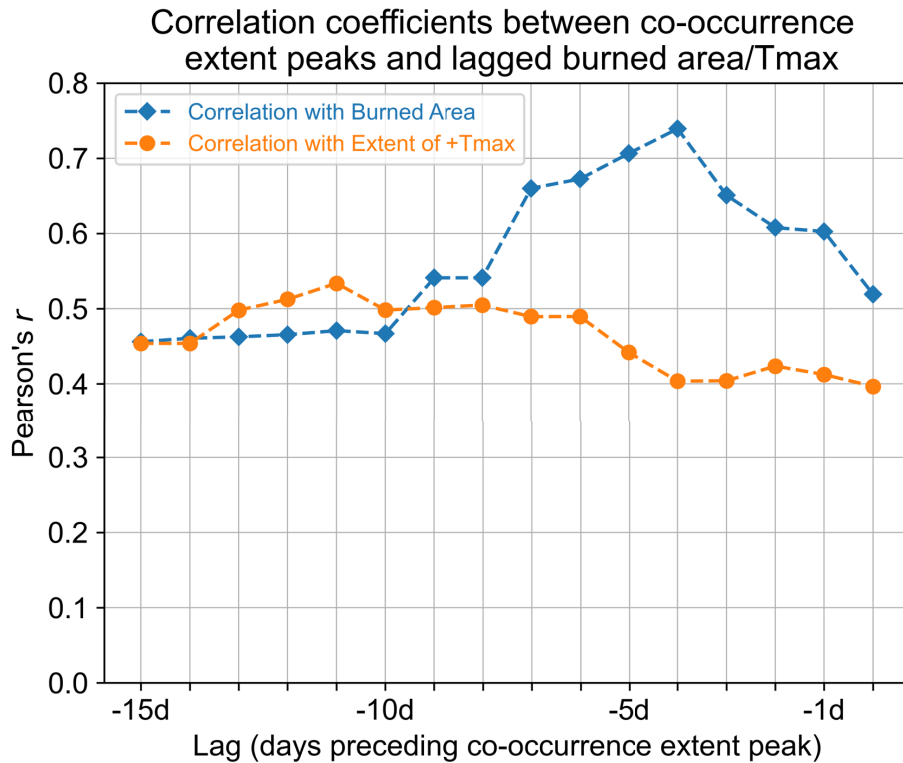
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921 **Fig. S7. Spatial correlation coefficients for three SOM node configurations.** Correlation
 922 coefficients (A) between each SOM node pattern and the individual constituent patterns in that
 923 node, and (B) between every unique combination of node pairs. Higher correlation coefficients
 924 in plot (A) indicate that individual days are well-represented by the node pattern into which they
 925 are assigned, and in plot (B) indicate greater redundancy of nodes in the SOM.

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927

928 **Fig. S8. Lagged relationship widespread co-occurrences, burned area, and maximum**

929 **temperatures.** 7-day lagged correlation coefficients between temporally independent peaks in

930 widespread PM_{2.5}/ozone co-occurrence spatial extent ($\geq 25\%$ of western US, $n = 21$) and peaks in

931 daily burned area in the western US and southwest Canada (blue dashes), and between peaks in

932 widespread co-occurrence extent and peaks in the extent of positive maximum temperature

933 (+Tmax) anomalies >1 standard deviation above local daily climatologies in the western US

934 (orange dashes) for all lags between 0-15 days preceding PM_{2.5}/ozone co-occurrence.

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943 **Table S1. Top 15 days with the largest extent of local PM_{2.5}/ozone co-occurrences in the**
944 **western US (2001-2020).**

945

| Rank | Date | Percent of western US grid cells | Population exposed, in millions |
|-----------|------------|----------------------------------|---------------------------------|
| 1 | 2020-08-24 | 68.5 | 36.7 |
| 2 | 2020-08-22 | 67.5 | 42.6 |
| 3 | 2020-08-25 | 66.4 | 27.9 |
| 4 | 2020-08-21 | 66.1 | 46.3 |
| 5 | 2020-08-23 | 64.3 | 36.7 |
| 6 | 2018-08-02 | 56.0 | 19.7 |
| 7 | 2018-08-01 | 54.4 | 20.4 |
| 8 | 2018-08-10 | 53.9 | 19.5 |
| 9 | 2018-08-09 | 51.7 | 35.5 |
| 10 | 2020-08-26 | 50.9 | 22.0 |
| 11 | 2015-08-20 | 50.7 | 30.1 |
| 12 | 2020-08-20 | 50.1 | 41.3 |
| 13 | 2017-09-02 | 46.9 | 42.4 |
| 14 | 2015-08-21 | 46.9 | 25.8 |
| 15 | 2018-08-08 | 46.1 | 34.7 |

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