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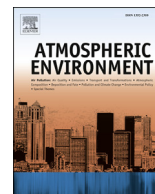


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Transport aloft drives peak ozone in the Mojave Desert



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HIGHLIGHTS

- Ozone exits Southern California through passes and thermal lofting over mountains.
- Ozone far downwind (400–1000 km) exhibits a characteristic time lag.
- Near-downwind sites (up to 400 km) have simultaneous ozone peaks.
- Contradiction is resolved if transport is aloft, not surface.
- Afternoon deep mixing drives Mojave summer ozone peaks.

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ABSTRACT

Transport of anthropogenic pollution eastward out of the Los Angeles megacity region in California has been periodically observed to reach the Colorado River and the Colorado Plateau region beyond. In the 1980s, anthropogenic halocarbon tracers measured in and near the Los Angeles urban area and at a mountain-top site near the Colorado River, 400 km downwind, were shown to have a correlated seven-day cycle explainable by transport from the urban area with a time lag of 1–2 days. Recent short term springtime intensive studies using aircraft observations and regional modeling of long range transport of ozone from the Southern California megacity region showed frequent and persistent ozone impacts at surface sites across the Colorado Plateau and Southern Rocky Mountain region, at distances up to 1500 km, also with time lags of 1–2 days. However, the timing of ozone peaks at low altitude monitoring sites within the Mojave Desert, at distances from 100 to 400 km from the South Coast and San Joaquin Valley ozone source regions, does not show the expected time-lag behavior seen in the larger transport studies. This discrepancy is explained by recognizing ozone transport across the Mojave Desert to occur in a persistent layer of polluted air in the lower free troposphere with a base level at approximately 1 km MSL. This layer impacts elevated downwind sites directly, but only influences low altitude surface ozone maxima through deep afternoon mixing. Pollutants in this elevated layer derive from California source regions (the Los Angeles megacity region and the intensive agricultural region of the San Joaquin Valley), from long-range transport from Asia, and stratospheric down-mixing. Recognition of the role of afternoon mixing during spring and summer over the Mojave explains and expands the significance of previously published reports of ozone and other pollutants observed in and over the Mojave Desert, and resolves an apparent paradox in the timing of ozone peaks due to short-range and long-range transport from the upwind basins.

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1. Introduction

The downwind transport of air pollution from megacities and other major pollution source regions is an important component of regional to global pollution loading in the atmosphere. Regional air quality studies tend to focus on source areas, as they present the greatest risk to human health and because they are the focus of

governmental pollution control efforts. Conversely, long-range transport studies often focus on continental to hemispheric scale impacts, with poor resolution of the details of transport near source areas. Studying the details of transport over moderate distances (100–400 km) can both reveal unexpected downwind atmospheric processes and inform concepts, assumptions, and models at both local and long-range scales, even for long-studied source regions.

The Mojave Desert in southern California and Nevada (USA) (location map in [Supplement Fig. 1](#)) lies downwind of the massive urbanization of the Los Angeles megacity (pop. 15M) in Southern

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California and the intensive farming and petroleum extraction region in Central California's San Joaquin Valley – two of the most polluted and most studied air basins in the United States (CARB, 2014b). These upwind areas have been the subject of periodic large-scale intensive field studies and continuous routine air pollution monitoring since the 1960s, and these studies have consistently shown large pollutant fluxes into the Mojave.

Limited studies have addressed transport along the upwind edge of the Mojave, and large-scale studies have characterized transport downwind of the Mojave, often emphasizing impacts on the Colorado Plateau and beyond, 500–1000 km downwind of Southern California.

No regionally integrated assessment of daily and seasonal air quality dynamics across the Mojave, at ranges from 100 to 400 km from the source regions, is available in the refereed literature. This paper draws on routine monitoring, reports from regulatory and land management agencies, focused studies in sub-regions of the Mojave, and individual scientific reports spanning the past few decades to sketch a unified picture of ozone in the Mojave, with particular emphasis on recognizing the role of transport in elevated layers in modulating surface ozone concentrations, and provides a new perspective on air pollution impacts on the Mojave.

1.1. Major field studies in “upwind” California

Photosynthetic processing of hydrocarbons and NO_x to generate urban ozone and related oxidants was first described in the Los Angeles basin (Haagen-Smit, 1950). Since that time, a large volume of academic research has focused on air quality in Southern California, and the California Air Resources Board (CARB) has sponsored periodic large interdisciplinary field campaigns in collaboration from other government agencies, industry, and the academic community. These campaigns were designed to improve both systematic understanding of atmospheric processes and to refine regulatory strategies to deal with the State's air pollution.

Unfortunately, all this work on the major air pollution source-areas in California largely ignored transport downwind of the study areas. A recent literature search (Watson, 2010) found more than 2300 published papers on air quality in California, but only about 10 percent addressed the desert areas of California in any way.

1.2. Long range transport studies

Beginning in the 1990s, pollutant transport across the North Pacific received increasing attention, beginning with intensive field studies of Asian outflow (e.g. Hoell et al., 1997; Huebert et al., 2003) and early receptor studies in North America (e.g. Jaffe et al., 1999; VanCuren and Cahill, 2002), and maturing into detailed modeling supported by extensive aircraft and surface measurements (e.g. Huang et al., 2010; Lin et al., 2012; Cooper et al., 2011). These studies have demonstrated that there is persistent Asian transport of ozone and other pollutants arriving above the Pacific marine boundary layer, and that it mixes with North American pollution as it encounters surface air lofted by land heating (Neuman et al., 2003; Langford et al., 2010; Neuman et al., 2012; Huang et al., 2013; Langford et al., 2015). Although most of these studies represent short periods, primarily in spring, to focus on the strongest Asian transport period, the dynamical insights from this research have identified fundamental transport pathways and provided an essential paradigm for understanding the complex vertical structure of transport in the western U. S.

1.3. Local air quality studies in the Mojave

Research projects conducted in the Mojave have been largely focused on individual problem areas, such as wind-blown dust from disturbed soil (e.g. Grantz et al., 1998) and desiccated lakes (e.g. Gillette et al., 1997), pollutant exposure for natural resource management (e.g. Burley et al., 2014), or effects on particular receptors (Trijonis et al., 1988) (A detailed map containing the place names mentioned in this paper is provided as Supplemental Fig. 1).

One major effort at tracking the geographic extent of pollutant transport into the western Mojave was undertaken by the U. S. Navy, with the purpose of identifying sources of visibility-impairing particles reaching the China Lake (Navy) and Edwards (Air Force/NASA) flight research areas in the far western Mojave (Trijonis et al., 1988). That study demonstrated routine transport of pollutants into the Mojave from both the San Joaquin and Southern California source areas and delineated a rough boundary between the Los Angeles and San Joaquin influence zones in the Antelope and Indian Wells Valleys in the far western Mojave. No comparable studies are available elsewhere in the Mojave.

In the mid-1980s, a project to track regional haze in the southwest U. S. used anthropogenic halocarbons as “tracers of opportunity.” One significant finding of this study was that methyl chloroform at Spirit Mtn. (1700 m), near the Colorado River in Southern Nevada, 400 km downwind of the Los Angeles urban core, correlated with the weekly cycles of methyl chloroform measured within the urban area and at Cajon Pass, on its downwind edge, with the distant downwind site having a time lag of 1–2 days (Miller et al., 1990; Bastable et al., 1990). Data from this and other sites used in the study demonstrated that the dominant trajectory of the polluted air was eastward into the Colorado Plateau.

Recent research has identified the Southern California urban region as a major source of enhanced ozone at Angel Peak (2675 m) near Las Vegas (Langford et al., 2015) and modeling and aircraft observations also show broad ozone impacts in the Colorado Plateau and the southern Rocky Mountain States (Utah, Colorado, Arizona, New Mexico) at distances up to 1500 km downwind (Cooper et al., 2010; Langford et al., 2010; Huang et al., 2010, 2013).

Measurements at Joshua Tree National Park, located on the upwind edge of the Mojave, have long shown strong ozone and nitrate impacts (Burley et al., 2014; Tonnesen et al., 2007), and recent ozone monitoring by the National Park Service at remote sites in Joshua Tree N. P. and the Mojave National Preserve has shown high concentrations at sites far from the upwind mountain barriers that separate the Mojave from the populous coastal basins. Among these, the modeling work of Tonneson stands out as the only source of detailed spatial distributions of pollutants across the Mojave, albeit focused on long-term pollutant deposition.

2. Air pollution sources affecting the Mojave region

Fig. 1 (adapted from Russell et al., 2010) provides a good regional perspective on the intensity of human activity in and around the Mojave. Satellite-mapped regional NO₂ emissions for 2008 make visible the major air pollution source areas and transport pathways in the region. NO₂ is a good surrogate for total anthropogenic emissions, as it is generated in almost all combustion processes, including motor vehicles, power production, industrial boilers, residential heating, and biomass burning, and it is both a precursor for ozone formation and a dominant source of anthropogenic nitrogen deposited to soil.

Fig. 2 compares aggregate air pollutant emissions for the San Joaquin Valley Air Basin (SJVAB), Coast Air Basin (SOCAB), and Mojave Desert Air Basin (MDAB) in California and the Las Vegas Non-attainment Area (LVNAA) in Nevada (CARB, 2009a,b; Pollack,

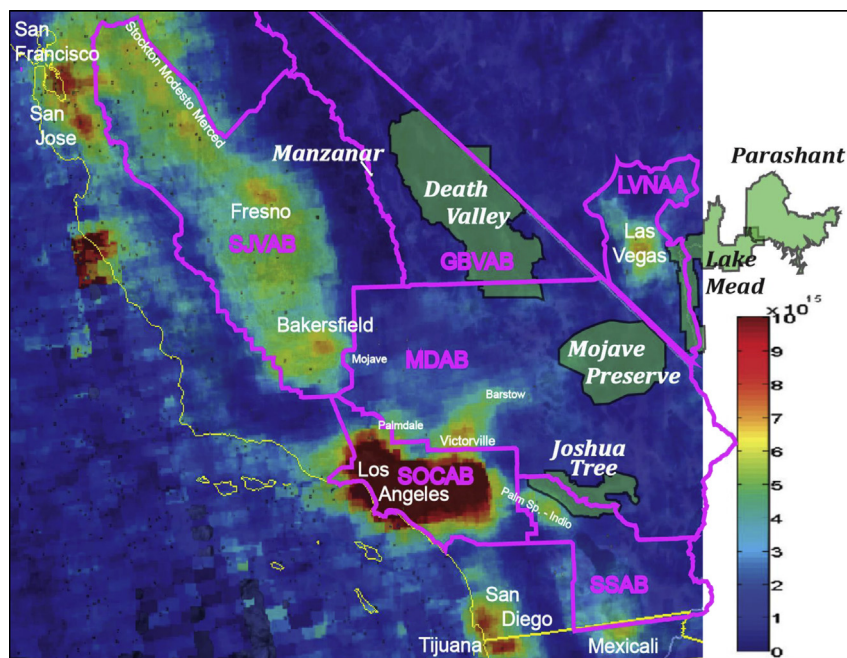


Fig. 1. Regional pollution source areas characterized by aggregate NO₂ column burden (molecules/cm²) observed by the OMI satellite, 2008 (Image base courtesy of Prof. Ron Cohen, U.C. Berkeley; see also: Russell et al., 2010). The major federally recognized air quality planning units in the region are outlined and labeled: SJVAB – San Joaquin Valley Air Basin, GBVAB – Great Basin Valleys Air Basin; MDAB – Mojave Desert Air Basin; SOCAB – South Coast Air Basin; SSAB – Salton Sea Air Basin; LVNAA – Las Vegas Ozone Non-attainment Area. Note the lobes of emissions from urban expansion and heavy transport along the two largest transportation routes out of southern California, the I-15 corridor through Cajon Pass and Victorville toward Las Vegas, and the I-10 corridor through San Geronio Pass and the Palm Springs-Indio area toward Phoenix, and also the NO₂ hotspot around Las Vegas.

2007). Overall, and particularly for ozone precursors (reactive organic gases – ROG – and oxides of nitrogen – NO_x), the total source population is small in the western and central (California) portions of the Mojave, especially when compared to the upwind areas in California.

2.1. Pollution sources internal to the Mojave

2.1.1. Natural ozone in the Mojave

Much of the Mojave is public land, administered by the U.S. Bureau of Land Management and the U.S. National Park Service, or withdrawn from public access for military uses (primarily as buffer space for flight test activities), and consequently is sparsely to totally unpopulated. This limits development and settlement, so that most of the Mojave is relatively undisturbed desert.

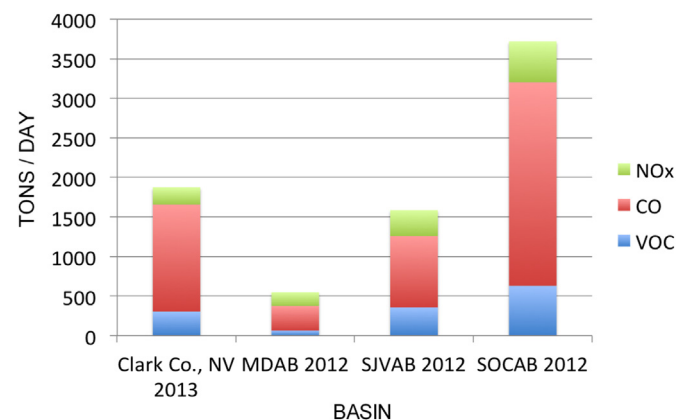


Fig. 2. Air Basin emission inventories, 2008. (Note: Clark Co. Nevada reports Volatile Organic Compounds (VOC), while California reports Reactive Organic Gases (ROG), reflecting slightly different methodologies. The label VOC applies to both in this plot).

The dominant vegetation in the Mojave is the creosote bush scrub plant community. Although the total biomass load appears light in the Mojave, creosote bush (*Larrea tridentata*) is a prodigious producer of biogenic organic chemicals (Geron et al., 2006; Jardine et al., 2010). Local, naturally occurring ozone in the Mojave is created when abundant sunshine drives photochemical processing of biogenic emissions from the desert vegetation and nitrogen produced by pedogenic processes (Diem, 2000; Geron et al., 2006; McCalley and Sparks, 2008; Jardine et al., 2010).

Desert vegetation hydrocarbon emissions track leaf temperature (Jardine et al., 2010), which is closely related to insolation and air temperature, thus the available natural ozone precursors peak around midday. Both ozone concentration and ozone formation rates drop as the precursors' concentrations decline, forcing surface ozone development in the afternoon to compete with increasing mixing and dilution due to solar heating. Thus, desert ozone peaks near midday throughout the year at sites not strongly impacted by anthropogenic ozone (Figs. 7 and 9, Blythe, bottom right).

2.1.2. Anthropogenic pollution sources in the Mojave

The major urban/industrial air pollution sources within the Mojave itself are its two major population centers, located in the far west and northeast. The largest, located in the northeastern Mojave, is the city of Las Vegas, Nevada, which, with its surrounding communities, comprises the largest urban area in the region (pop. 1.9M) (Nevada, 2012). Along the southwestern boundary of the Mojave, groups of smaller cities and unincorporated communities, most clustered near the passes into the Southern California urbanized basins, constitute the other major population concentrations. These clusters include: Antelope Valley (Palmdale, Lancaster and surroundings, approximately 400,000 pop.) north of Soledad Pass; Victor Valley (Victorville, Adelanto, and surroundings, approximately 300,000 pop.) north of Cajon Pass, and the cluster of communities northeast of San Geronio Pass and west of Joshua Tree N.P. (Morongo Valley, Yucca Valley, Joshua Tree,



Fig. 3. Major passes allowing low-altitude (800–1600 m elevation) air flow into the Mojave (labeled arrows), and the approximate boundary between zones of San Joaquin Valley and South Coast influence (dashed line) (Trijonis et al., 1988; Angevine, 2013; Huang et al., 2013).

and Twentynine Palms – pop. about 70,000) (CDOF, 2012; US Census, 2010).

Other, more remote communities in the Mojave are relatively small. The largest of these is Indian Wells Valley (Ridgecrest and Inyokern, serving China Lake NAWS, about 40,000). Others include the communities of Rosamond (18,000) and California City (14,000), north and west of Edwards AFB; Pahrump, Nevada

(36,000), a retirement and bedroom community west of Las Vegas; and the rail and highway transportation hubs of Barstow (23,000) in the central Mojave and Mojave (4300) in the far west (CDOF, 2012; US Census, 2010).

Anthropogenic pollution generated within the Mojave tends to be local in scale, with the exception of a few large combustion facilities. Only the Las Vegas area has a large enough population to

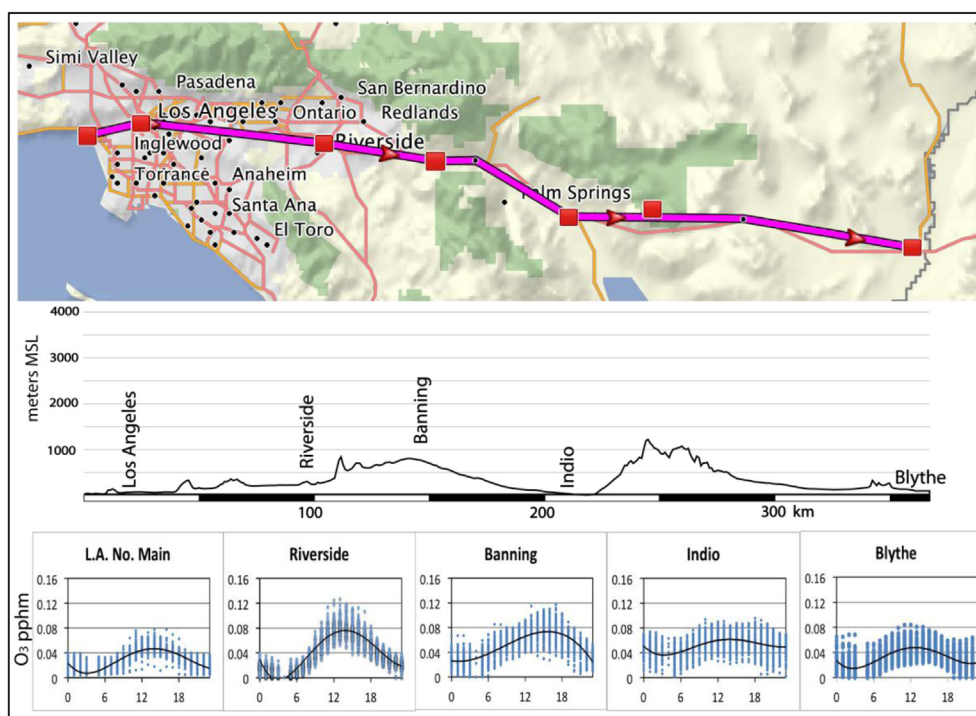


Fig. 4. Ozone diurnal cycles (plot shows all hours June–August 2012; curve is hourly means) along a transect from the Pacific Ocean to the Colorado River through San Geronimo Pass. Top: map of transect. Middle: topographic profile along the transect. Bottom: Composite diurnal ozone cycles for June, July, and August 2012 for selected sites along the transect, illustrating the evolution of the daily ozone “pulse” created by the sea breeze flowing from the Pacific Ocean into the desert.

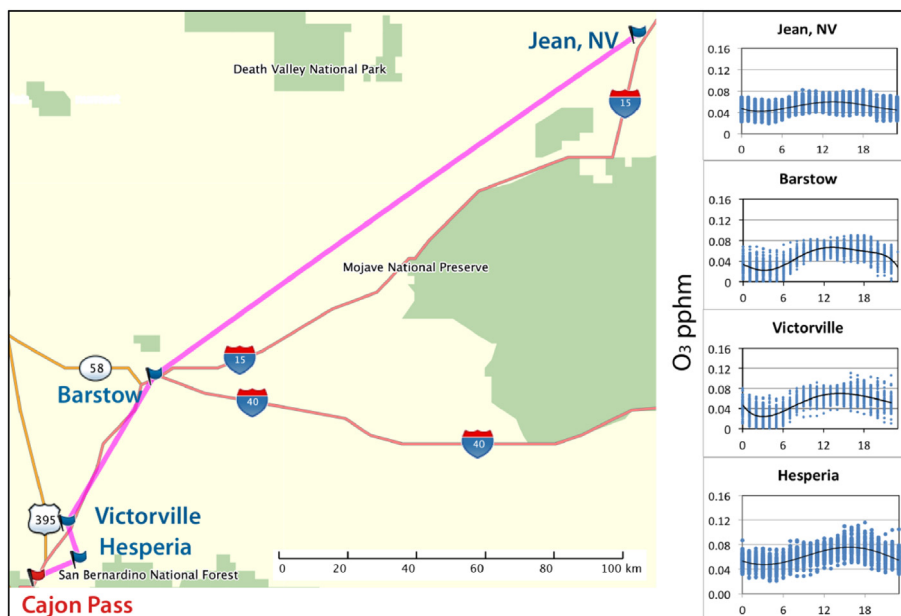


Fig. 5. Ozone diurnal cycles (plot shows all hours June–August 2012; curve is hourly means) along a transect from Cajon Pass to Jean, NV. If surface transport drives peak ozone, peaks must be later farther downwind, however ozone peaks downwind of Cajon Pass are simultaneous.

drive significant urban pollution (Figs. 1 and 2, Supplemental Fig. 1), affecting much of the area between the Spring Mts. and Lake Mead with a distinct, visible particulate haze layer in winter and ozone in summer.

2.2. Upwind pollution sources

Although the Mojave is climatically isolated from the Pacific by mountains that effectively block most precipitation, there are low areas and gaps in the mountains (Fig. 3) that allow low-level air flow into the desert, and, as discussed below, meso-scale flows can loft air over the mountain crests, carrying with it pollutants generated at ground level in the populated upwind areas. These are briefly described here.

2.2.1. Pollution from the Southern California megacity

Sources of particulate and gaseous pollution in Southern California (SOCAB in Fig. 1) include surface and marine transportation, petroleum production and refining, manufacturing, and domestic emissions for a population around 15 million (CDOF, 2012). The State of California and the South Coast Air Quality Management District have imposed strong emission controls throughout the region, and pollution levels have fallen significantly in recent decades, but the EPA continues to classify the SOCAB as non-attainment for both PM_{2.5} and ozone (CARB, 2014a,b).

Much of Southern California's economic activity and the associated emissions are concentrated in the northern portions of the region – Los Angeles, Riverside and San Bernardino Counties – which abut the mountains that separate the Mojave from the coastal lowlands. In the latter decades of the 20th century, significant population has also bled over the mountains into the western Mojave.

2.2.2. Pollution from the San Joaquin Valley

The San Joaquin Valley (SJVB in Fig. 1) is a national hotspot of air pollution, with portions of it being rated by the EPA as severe or extreme for ozone, and the southern counties in the Valley also rated non-attainment for fine particles (PM_{2.5}) (CARB, 2014a,b).

On the far west boundary of the Mojave, the Sierra Nevada and Tehachapi Mts. block low altitude air flow from the San Joaquin Valley into the desert. Two major passes connect the San Joaquin Valley and the Mojave (Fig. 3).

2.3. Pollution from Asia

Most of North America, especially the interior West, is exposed to persistent low-concentration air pollution transported from Asia (VanCuren and Cahill, 2002; Fiore et al., 2002; Lin et al., 2012; Yu et al., 2012). The Asian continental plume is a mixture of dust and combustion products, and it constitutes the single largest component of background aerosol in the free troposphere over North America (VanCuren, 2003; Yu et al., 2012). Although the dust is a useful chemical tracer, long-term average fine particle chemical analysis shows that only 30% is mineral (dust, coal fly ash, etc.), while most of the rest is composed of combustion-related material (VanCuren, 2003). Measurements in the Pacific Northwest have shown the Asian particulate material to be accompanied by gaseous pollutants, including carbon monoxide, ozone, and nitrogen oxides (Jaffe et al., 1999, 2001, 2004; Jaeglé et al., 2003; Fiore et al., 2002).

Asian ozone is regularly transported above the Mojave as it is entrained in stable air isentropically circulating around the east Pacific high-pressure zone (Cooper et al., 2011).

Deep mixing has been identified as a mechanism to deliver Asian ozone to surface sites in California (Parrish et al., 2010). Two case studies demonstrate how this transport occurs. Huang et al. (2010) reported observing Asian ozone over the Pacific Ocean during aircraft sampling in June 2008, and used those observations and a transport model to estimate Asian ozone impact in Southern California. The model showed that the cool marine air over the coastal areas prevented mixing to the ground, but the warm turbulent midday air over Joshua Tree NP drove strong mixing to the surface. Recently, Langford et al. (2015) combined observations and models to estimate the fractional Asian, North American, and Stratospheric ozone components at Angel Peak (2675 m) in Southern Nevada. For the high ozone days during the period of the study, the non-local ozone budget was dominated by stratospheric

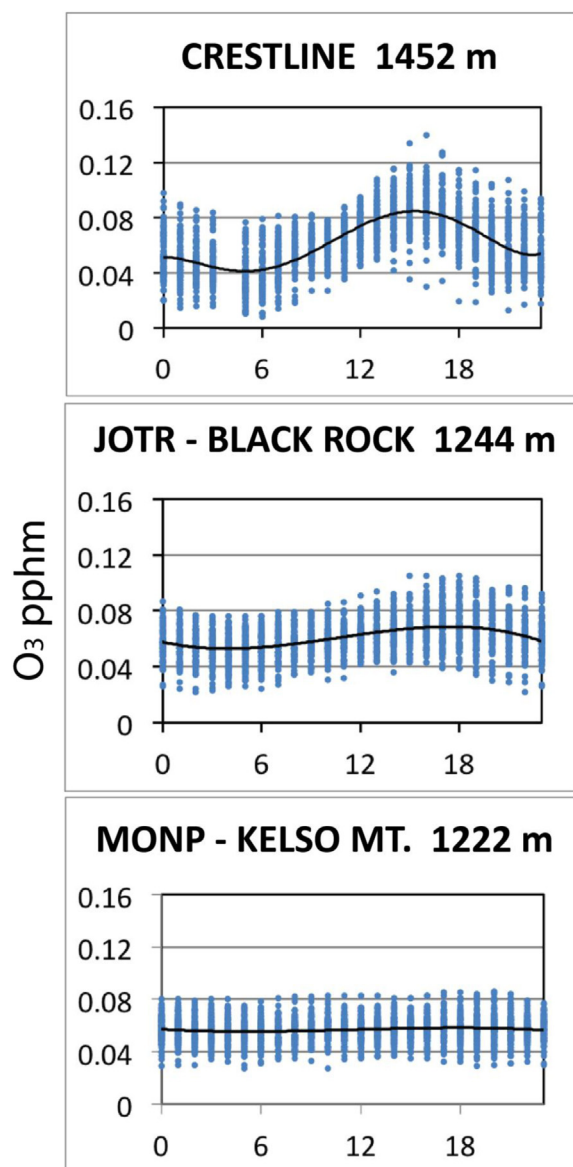


Fig. 6. Diurnal cycles of ozone (plot shows all hours June–August 2012; curve is hourly means) at elevated sites: Crestline (1390 m), on the coastal side of the San Bernardino Mts., exhibits the expected diurnal cycle. Black Rock (Joshua Tree N.P.) is in the lee of the San Bernardino Mts., and Kelso Mt. (Mojave N.P.) is a solitary peak. All show suppressed diurnal cycles, consistent with elevated air masses having limited contact with the ground.

down-mixing, contributing about 30 ppbv to total ozone, while Asian sources provided only about 10 ppbv. Recent assessments of rural ozone across the western U. S. show a rising trend of about .5 ppbv/yr, largely attributed to Asian ozone (Cooper et al., 2010; Gratz et al., 2015).

Asian ozone can be qualitatively inferred to be regularly present at surface sites in the Mojave since Asian aerosols have been recognized as a very frequent component of IMPROVE aerosol samples collected at Death Valley (VanCuren, 2007).

On a broad scale, Asian ozone is modeled to be, at present, of minor importance to surface ozone in the U.S. (Fiore et al., 2002; Reidmiller et al., 2009; Lin et al., 2012), however, the models used in these and similar studies lack the spatial resolution and density of measurement sites to properly deal with the sharp terrain gradients in the western U. S., and particularly in California (for example Reidmiller et al. use one station, Death Valley, to represent

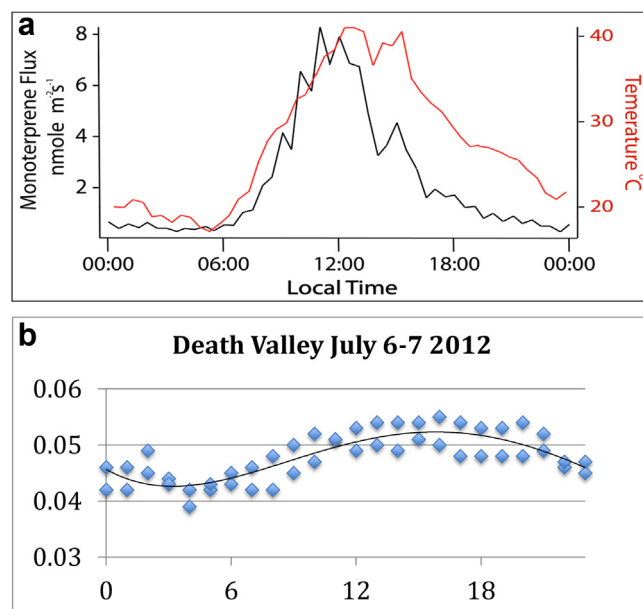


Fig. 7. Ozone production from local biogenic precursors. a. Daily cycle of temperature and monoterpene emissions for creosote (Data from Jardine et al., 2010). b. Ozone at Death Valley, July 5–6, 2012 (no transport).

the entire Mojave region), leaving considerable uncertainty as to effects at particular locations.

3. Transport dynamics in Southern California

3.1. Low level transport

Because ozone concentrations are highest in both the San Joaquin Valley and Southern California under strong temperature inversions, efforts to detect the effects of ozone transport on neighboring air basins has focused on surface advection during periods of high ozone (CARB, 2001). This has led to a focus on transport through the passes shown in Fig. 3.

Low-level air flow from the coastal regions into the Mojave is driven by temperature differences between the cool Pacific marine layer and the warmer air inland, particularly in the southern San Joaquin Valley and in the Mojave Desert. Marine air moves eastward across Southern California, driven by the Pacific Ocean sea breeze. Similarly, Pacific Ocean marine air enters the northern end of the San Joaquin Valley through San Francisco Bay and the Sacramento-San Joaquin Delta and is advected south toward the warmer southern end of the Valley. There, assisted by afternoon heating, it can then flow over the lower crests of the extreme southern Sierra Nevada and through Tehachapi Pass into the northwest Mojave.

As each daily pulse of air moves across the Los Angeles coastal plain or through the San Joaquin Valley, ozone precursors (NO_x and VOCs) are added to the air while sunlight drives ozone production, resulting in progressively higher ozone peak concentrations as the air moves downwind.

In the extreme western Mojave, air flow out of the San Joaquin Valley comes through Tehachapi and Walker Passes (Fig. 3), and spreads eastward and somewhat northeast toward Las Vegas.

On the southern edge of the far west Mojave, air flows out of the South Coast Air Basin through Soledad Pass (Fig. 3). This flow appears to be constrained to primarily move eastward along the northern flanks of the San Gabriel Mountains. Farther east, this flow is joined by air moving through Cajon Pass, where it then spreads

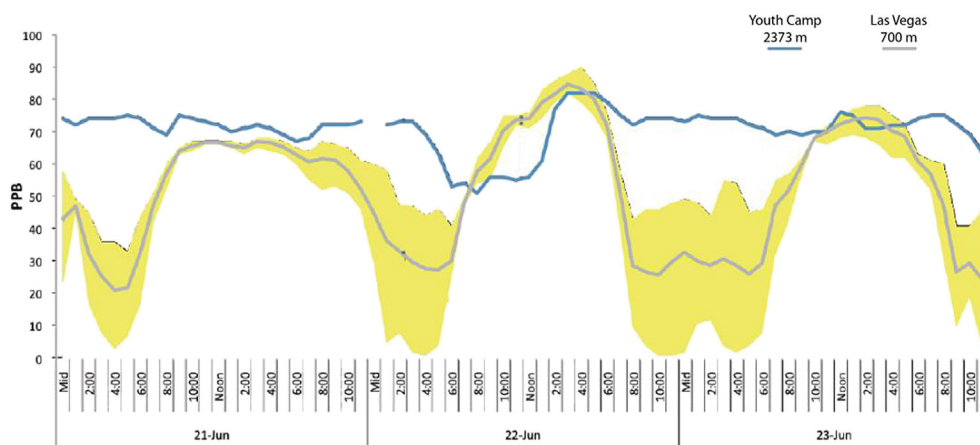


Fig. 8. Diurnal structure of ozone fumigation from aloft: a. Timelines of ozone at an elevated site in the Spring Mts. (SMYC) and an urban valley site in Las Vegas, summer 2011 (Huys, 2012). Afternoon heating mixes ozone from aloft to the surface, equalizing the concentrations late in the afternoon on June 22.

east and northeast farther downwind (Trijonis et al., 1988; Angevine, 2013, Fig. 8; Huang et al., 2013, Fig. 3). In the far eastern SOGAB, air exits the coastal basins through San Geronio Pass (also known as Banning Pass), flowing into the southern Mojave and Colorado Deserts (Fig. 3).

The daily summer ozone pattern in the SOGAB and the southern Mojave is illustrated in Fig. 4, which shows ozone diurnal cycles for

sites along a transect from the Pacific Ocean to the Colorado River. The transect follows a low altitude route, passing through San Geronio Pass (780 m), then skirting the southern edge of Joshua Tree National Park and reaching the Colorado River at Blythe.

The diurnal plots at the bottom of Fig. 4 illustrate the prevailing conceptual model (e.g. CARB, 2001; CARB, 2014b) for surface pollutant transport into the deserts. The plots show composite

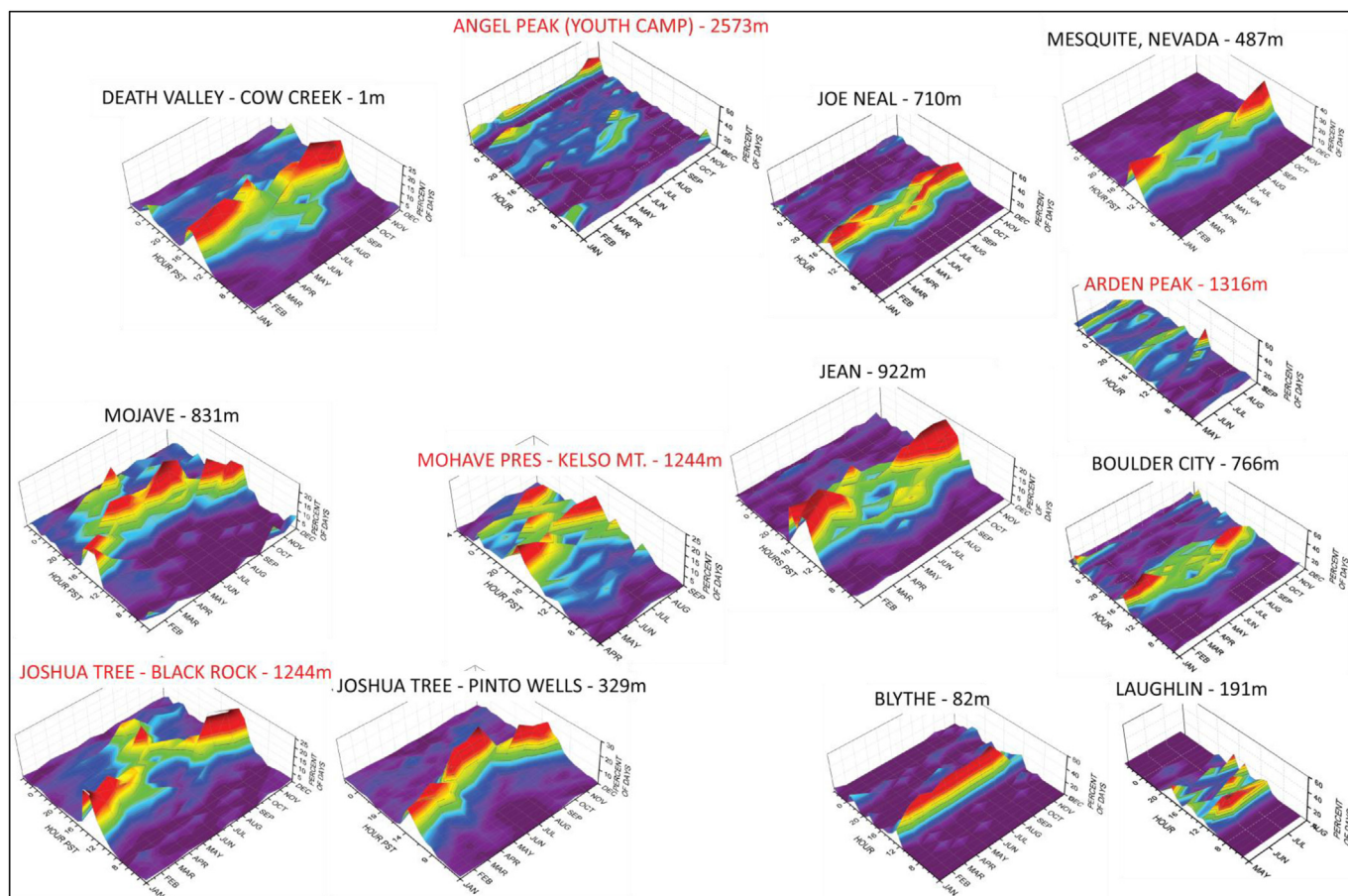


Fig. 9. Frequency distributions by month for hour of peak ozone concentration based on hourly ozone data 2007–2010. Natural in situ ozone production peaks near noon due to dependence on solar uv flux. Elevated sites (Black Rock and Mojave Preserve) are regularly in contact with the deep layer of transported ozone in summer; low altitude sites in the path of the ozone cloud leaving southern California (Pinto Wells, Mesquite) show late afternoon peaks caused by thermally driven vertical transport of ozone from aloft.

diurnal ozone cycles for sites along or near the low altitude route, based on all hours for June, July, and August 2012 (CARB, 2014a). The progression of ozone production and its transport by the sea breeze are evident in the low, early ozone peak in downtown Los Angeles, the later and higher ozone peak at Riverside, due to accumulated pollution in transit, then prolonged evening ozone as the coastal air streams over San Geronio Pass (Banning, 680 m). As the air leaves the pass, it begins to spread out and mix vertically and horizontally, showing up at Indio (sea level) as sustained ozone long after dark, and eventually dissipating and having little, if any, impact at Blythe (82 m). Common to all these sites is the early morning dip in ozone due to chemical destruction and deposition of ozone over-night, and fresh buildup of ozone each afternoon, generally interpreted as driven by daily pulses of fresh ozone and precursors from the upwind Air Basin.

This pattern has been assumed to operate near the other passes shown in Fig. 3. Similar diurnal patterns were cited by CARB in designating the ozone violations in the Mojave Air Basin as due to transport from upwind areas (CARB, 2001, 2014b).

3.2. Contradictions in the low level conceptual model

The low-level flow, or “pass spill-over” conceptual model described above appears valid for the transect shown in Fig. 4. Implicit in this conceptualization is the idea that ozone peaks will be successively later at sites farther downwind from the passes. Unfortunately, this is not the case more broadly across the Mojave. Analysis of peak hour ozone timing in summer shows that summer ozone peaks in the Mojave are broadly simultaneous, regardless of distance from the mountain passes, as illustrated by Fig. 5.

3.3. Transport aloft

Although the cool marine air provides a strong and persistent inversion over the coastal lowlands of Southern California, air is routinely vented from the boundary layer by thermal lofting along south- and west-facing mountain fronts bounding the Southern California coastal zone (Supplemental Fig. 2; Lu and Turco, 1996; Langford et al., 2010). Although the venting of ozone to higher altitudes as the coastal air encounters the mountains has been known since the 1980s (Lawson, 1990), studies at that time focused on its role in driving return flow aloft back toward the coast. This lofting has largely been treated anecdotally, and a literature search did not discover any information on its frequency or spatial extent, however it appears as a strong driver of ozone export from the SOCAB in the month-long simulation presented by Huang et al. (2013). The evidence discussed below indicates it is widespread and frequent throughout the warm months.

3.3.1. Direct evidence of transport aloft

The mountain barriers block low-level flow, but, under strong spring and summer sun, they create an additional pathway for pollutants to escape the upwind basins. Published reports from 1996 to 2010 document thermal lofting of air up the sunny south-facing slopes of the San Gabriel Mts. (Supplemental Fig. 2) (Lu and Turco, 1996; Langford et al., 2010; Cooper et al., 2011). This “chimney effect” lofts polluted air from the basin floor to 4 km altitude, more than a kilometer above the average height of the crest, and likely occurs along other sunny slopes in the South Coast Air Basin. Air lofted above the crest can be entrained in regional flow, carrying Southern California pollutants long distances – in the case described by Langford et al. (2010), ozone transport was tentatively traced 1000 km east into southwestern Colorado, and Huang et al. (2013) model persistent transport up to 1000 km for the entire month of May, 2010.

While the frequency and seasonality of such strong lofting events is uncertain, air from the South Coast regularly tops the Transverse Ranges in summer, as indicated by daily cycles of transported pollutants observed at Mt. Wilson (1730 m) in the San Gabriel Mts. (Gorham et al., 2010).

Fig. 6 shows summer diurnal ozone cycles for elevated sites in and downwind of the mountains bounding the Southern California urban area. Crestline (1390 m), in the San Bernardino Mts., experiences strong diurnal cycling as upslope flow delivers ozone in the afternoon and evening, then ozone drops as flow reverses over night. Unlike the low altitude transport, however, ozone at downwind elevated sites, represented here by the diurnal plots for Black Rock (1215 m) on the western edge of Joshua Tree National Park, and Kelso Mt. (1225 m), an isolated high point in the Mojave National Preserve, experience sustained ozone concentrations that do not track with the strong nocturnal drop in ozone observed at the lowland sites shown in Fig. 4 and at Crestline. This difference can be explained if outflow from the basin is sustained at and above the levels of the passes for many hours each afternoon and evening.

Just such flow as a coherent elevated layer was observed at Cajon Pass with a vertical-looking lidar operated during the Southern California Ozone Study (SCOS) in 1997 (Philbrick, 2001). The lidar observations (Supplemental Fig. 3) show a brief strong peak of ozone transiting the pass in late afternoon, and a long sustained flow of particles, and, presumably, ozone precursors and NO_x reservoir species such as PAN, propagating in a layer about 1.5 km thick throughout the night and next morning. This sustained nighttime flow pattern was also reported to occur regularly at Cajon Pass during the South Coast Air Quality Study field program in summer 1987 (Bastable et al., 1990).

Isentropic transport of ozone lofted up to about 4 km over the mountain crests and a low level persistent flux at 1–2 km elevation through the passes is consistent with the observed long-term structure of the Mojave atmosphere found in two months of daily ozonesondes launched at Joshua Tree National Park in May and June of 2010 (Cooper et al., 2011), which show a weak ozone layer at 1–3 km and a strong layer from 4 to 5 km.

The presence of such persistent transport layers, combining with the isentropic lower troposphere flow off the Pacific described and observed by Cooper et al. (2011), and modeled by Huang et al. (2010), argue strongly for a persistent flow aloft over the Mojave.

Three recent studies have also reported this phenomenon at particular locations in the Mojave.

Burley et al. (2014) argues, based on extensive data from Joshua Tree National Park, that ozone exposure in the western portion of the Park must be due to flow over the crest of the San Bernardino Mts.

Much farther downwind, studies in the Las Vegas area (Huys, 2012), comparing ozone measured at Angel Peak (also SMYC) at 2675 m elevation in the Spring Mts. paired with a low altitude surface site in Las Vegas Valley, indicate that a persistent ozone-rich layer was present over the valley (Fig. 8). In this case, the low altitude site had the typical “local ozone” diurnal signature of a peak near noon on June 21, and began the next day (June 22, center of plot) following the same pattern, but late in the day, solar heating of the ground caused growth of the turbulent boundary layer to reach the ozone aloft, at which time the ground level concentration jumped as the aged ozone from above was rapidly mixed to the surface, causing a sustained afternoon – evening high ozone period. Finally, on June 23, the local ozone diurnal pattern returns with its midday peak.

Most recently, Langford et al. (2015) describe aloft mixing driving peak ozone days in Las Vegas, confirming the observations of Huys.

The combination of the flows reported from Cajon Pass, the dynamics of the events reported by Langford and Huys, and the season-long persistence of ozone aloft shown by the monitoring data, particularly at Kelso Mt., suggest that transport aloft is a significant component of total ozone transport into and across the Mojave.

3.3.2. Indirect evidence for widespread ozone aloft

The limited number of elevated (above 1 km) ozone sampling sites in the Mojave makes direct observation of the spatial extent of the suspected regional layer aloft impossible, but the afternoon mixing process observed by Huys (2012) and described by Huang can be used to detect ozone fumigation due to deep afternoon atmospheric mixing at lower altitude sites below the transported air.

Efficient ozone production requires both strong sunlight and weak mixing, keeping both precursors and ozone confined and concentrated in a shallow boundary layer (Seinfeld and Pandis, 1998). The daily cycle of local (“natural”) ozone production in the Mojave is tied to the availability of biogenic hydrocarbons released by plants, which tracks temperature and sunlight (Fig. 7, top) (Jardine et al., 2010; Geron et al., 2006; Martin et al., 1999; LaFranchi et al., 2011). The diurnal ozone cycles for Death Valley for two days during a low-wind period (regional high pressure) in July 2012 (Fig. 6, bottom) illustrate the resulting ozone diurnal cycle. Ozone on low wind days at Death Valley tracks the biogenic emissions from creosote, rising rapidly toward noon, then flattening as mixing begins to overpower ozone production, and driven a drop in ozone through the later afternoon. In such an environment diurnal cycles can be used to distinguish local ozone production from vertically transported ozone, since local ozone must usually peak near midday, while ozone derived from deep mixing, which cannot occur during cool morning hours, must be more commonly observed as late afternoon or early evening peaks.

These considerations lead to the hypothesis that timing of ozone peaks can be read as an indicator of ozone sources – midday for local, later in the day for vertically transported ozone. Unfortunately, individual days show large variations in ozone timing and concentrations, making it difficult to see such a pattern in a conventional time series. However, if the pattern is stronger than the day-to-day variation in numeric ozone concentrations, then analysis of many days can detect the pattern.

The results of testing this hypothesis are shown in Fig. 9. Using all ozone data from the years 2007–2010, each plot presents the frequency of occurrence of daily peak ozone for each hour of the day (percent of days). In the plots, hours are on the left baseline axis, with the daily cycle (local standard time) plotted from 04:00 to 03:00 the next day to match the observed daily ozone cycle, which bottoms at 04:00 most days. The z-values for the plots are the percent of days in that month when ozone peaks at that hour (months along the right-side base axis).

Transformed in this way, it is easy to visualize the springtime transition from local (“natural”) ozone production in the cool months (December to March) to the strong transport effects in the warm months (April to November).

The plots in Fig. 9 show the overwhelming transport impacts at elevated sites such as Black Rock and Kelso Mt. (red labels), where no firm diurnal cycle is visible, and also pick up the subtler seasonal shifts at the far downwind low altitude sites (black labels) such as Pinto Wells and Mesquite, NV.

Fumigation from transport aloft also helps explain why the onset of the afternoon peak does not migrate ever later in the day at the far downwind sites, as would be expected if the daily pulses through the passes flowed at ground level.

Finally the plots in Fig. 9, which are arranged in a quasi-geographical placing, suggest the spatial extent of significant

ozone impact in the Mojave is concentrated along a corridor from Southern California northeastward toward Las Vegas and the Utah–Arizona border. The lack of evidence of transported ozone at Blythe and the minimal transport impact at Death Valley suggest the lateral limits of strong transport. This spatial pattern is supported by a set of back-trajectories from Las Vegas (Fig. 10; Huys, 2012), computed for high ozone periods in 2011, which show both the eastern Pacific coastal flows and the transport pathways from the San Joaquin Valley are strongly associated with high ozone concentrations in Las Vegas in Spring, while the Southern California source areas dominate in Summer.

4. Conclusion

Ozone in the Mojave Desert is influenced by ozone production in upwind areas of California as well as by both stratospheric down-mixing and Asian ozone export over the Pacific Ocean. Although mixing from aloft has been recognized as a large scale process to deliver Asian and Stratospheric ozone over the Mojave and the Colorado Plateau, a second, lower-layer effect, driven by lower altitude isentropic flows leaving Southern California, appears to be at work as well. The presence of such a lower layer can explain the otherwise apparent contradiction between timing of ozone maxima at low altitude sites in the Mojave Desert and observed transport times at larger scales. Fig. 11 is a graphic representation of this alternative, 2-layer transport process.

The plots in Fig. 9 show that, in winter, local ozone produced by sunlight and biogenic precursors dominates throughout the region.

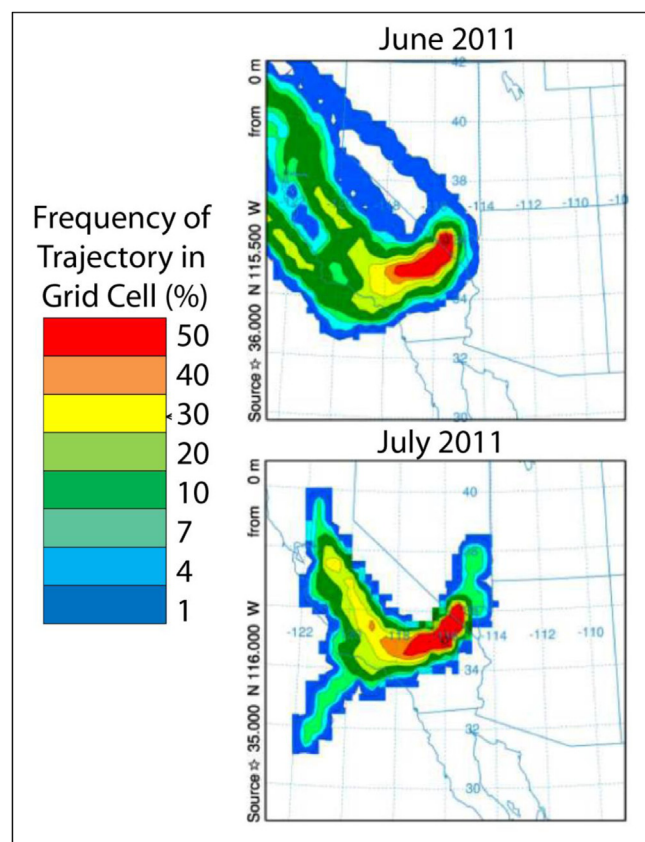


Fig. 10. Back-trajectories from Las Vegas (Huys, 2012), computed for high ozone periods in 2011. The June trajectories include the eastern Pacific flow described by Cooper et al. (2011), while the July trajectories match the apparent ozone corridor suggested by the plots in Fig. 9.

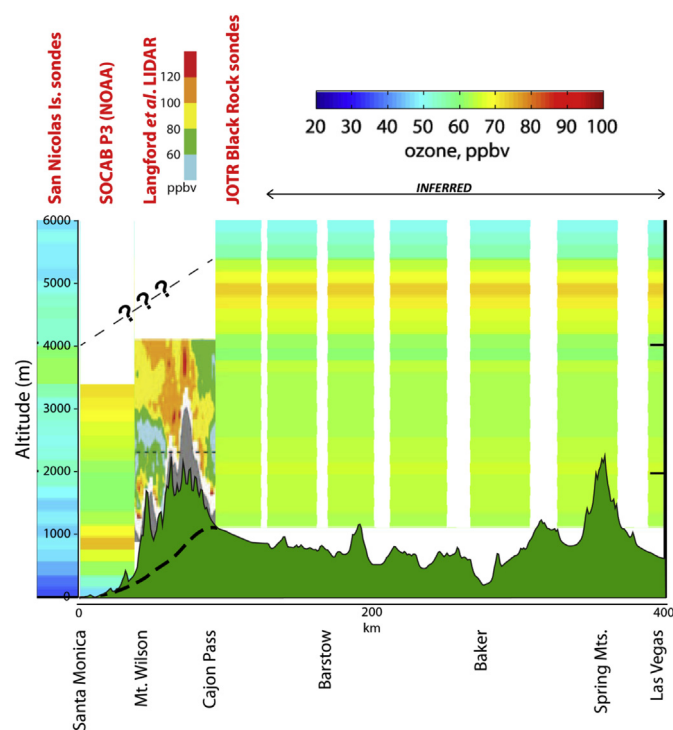


Fig. 11. Graphic “cartoon” representation of ozone transport from the SOCAB across the Mojave Desert. The figure combines the ozonesonde data from Cooper et al. (2011) with the lidar observations from Langford et al. (2010) with a terrain profile showing the elevation variation from Cajon Pass to Las Vegas Valley.

In summer, when transported ozone is expected to be significantly more important than local in-situ ozone production, strong continuous ozone impacts are observed, as expected, at elevated sites (red labels). In summer at lower elevations (<1 km), the local noon-linked ozone peak is not the daily peak; rather, deep afternoon mixing delivers polluted air from aloft, driving the ozone peak to be coordinated with maximum mixing later in the day. These diurnal patterns, unrelated to distance from the upwind source areas, are not consistent with transport delivered by surface flows. The more plausible process is transport aloft, which is compatible with the widespread simultaneous late afternoon ozone peaks observed in the Mojave Desert and the lagged peaks observed and modeled for elevated terrain far downwind.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.atmosenv.2014.09.057>.

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