Can carbon dioxide be used as a tracer of urban atmospheric transport?

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[1] Carbon dioxide is a stable constituent of the atmosphere that has no major terrestrial sinks other than atmospheric transport in the absence of photosynthetic activity by plants. In urban atmospheres, CO_2 mixing ratios are often elevated above ambient by large local sources from combustion. We measured CO_2 mixing ratios and the isotopic composition of CO_2 at four locations in the Salt Lake Valley, Utah, during a persistent cold pool event in the winter of 2004. The results showed a strong influence of atmospheric stability and the height of the capping inversion on CO_2 mixing ratios and suggested that during persistent cold pool events the air mass beneath the capping inversion can be relatively well mixed. Spatial and temporal patterns in the isotopic composition of CO_2 and the relationship between particulate concentrations and CO_2 mixing ratio support this interpretation. These results suggest that CO_2 mixing ratio, which is abundant and relatively easily measured in urban atmospheres, can provide information about complex wintertime atmospheric transport and mixing as well as carbon cycling in urban mountain basins.

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1. Introduction

[2] Measurements of atmospheric CO_2 mixing ratios in cities have become more common as a means of studying local greenhouse gas emissions, urban carbon cycles, and the atmospheric environment for urban plant growth [*Tanaka et al.*, 1985; *Berry and Colls*, 1990; *Aikawa et al.*, 1995; *Reid and Steyn*, 1997; *Idso et al.*, 2001; *Day et al.*, 2002; *Grimmond et al.*, 2002; *Idso et al.*, 2002]. When coupled with measurements of mixing ratio, measurements of the isotopic composition of urban CO_2 can also provide important information as a means of identifying CO_2 sources and distinguishing between biogenic and anthropogenic CO_2 [*Zondervan and Meijer*, 1996; *Meijer et al.*, 1997; *Kuc and Zimnoch*, 1998; *Takahashi et al.*, 2001, 2002; *Kuc et al.*, 2003; *Pataki et al.*, 2003a]. We have shown previously that CO_2 mixing ratio and its isotopic composition has a distinct seasonal trend in the Salt Lake Valley, Utah, due to a combination of varying CO_2 sources and seasonal patterns in atmospheric mixing and transport [*Pataki et al.*, 2003a]. However, these measurements were conducted at a single location at the eastern edge of the valley.

[3] CO_2 mixing ratios can show a high degree of spatial as well as temporal variability, particularly in urban areas, reflecting variability in local sources, atmospheric stability, and measurement location [*Berry and Colls*, 1990; *Idso et al.*, 2001; *Day et al.*, 2002; *Grimmond et al.*, 2002]. In the Salt Lake Valley, anthropogenic emissions dominate CO_2 sources in the wintertime, when CO_2 mixing ratios reach their maximum because of a strong influence of temperature inversions typical of mountain basins [*Pataki et al.*, 2003a]. In the absence of significant biological activity in the wintertime, CO_2 has no major sinks such that mixing ratios are influenced primarily by source strength and atmospheric transport. Atmospheric transport in the Salt Lake Valley was intensively studied in the Vertical Transport and Mixing Experiment (VTMX) and the URBAN 2000 campaigns, which showed the importance of canyon, slope, and down valley flows for atmospheric transport during diurnal nocturnal cold pools [*Doran et al.*, 2002; *Monti et al.*, 2002; *Banta et al.*, 2004; *Fast and Darby*, 2004]. These diurnal cold pools form within mountain basins that have inflow and outflow limited by topography. Diurnal cold pools are characterized by a surface or near surface based temperature inversion that forms each evening and is eroded and destroyed the following morning through the growth of a convective boundary layer. While previous field campaigns provided insight into diurnal cold pools, they did not examine persistent cold pools. Like diurnal cold pools, persistent cold pools occur within mountain basins; however, they form primarily during the winter and are not

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Table 1. Location and Elevation Above Sea Level of the Four Measurement Sites in the Salt Lake Valley, Utah, USA

Site	Coordinate Location	Nearest Cross Streets	Elevation
Eastern foothills	40°45′N, 111°50′W	1400 E, 200 S	1430 m
Western foothills	40°33′N, 112°03′W	Hwy 111, 9000 S	1580 m
Downtown	40°45′N, 111°53′W	200 E, 300 S	1320 m
Residential	40°42′N, 111°51′W	900 E, 2900 S	1330 m

destroyed by daytime sensible heating, and may remain resident within basins for days or weeks [*Whiteman et al.*, 2001]. These cold pools inhibit mixing with the upper atmosphere and in urban areas such as the Salt Lake Valley, are associated with poor air quality. Thus a data gap exists in a critical area of mountain meteorology that strongly influences both climate and air quality in Salt Lake City and other urban basins.

[4] We monitored CO_2 mixing ratios, meteorological variables, and atmospheric particulates during the evolution of a major cold pool event that occurred in January 2004. Mixing ratio measurements occurred simultaneously at four locations that were distributed both horizontally and in vertical elevation around the Salt Lake Valley. These measurements have potential applications for understanding both greenhouse gas emissions and air quality in urban areas where combustion sources dominate the variability in atmospheric CO_2. However, CO_2 is also an inert gas that is present in large, easily measurable quantities in the urban atmosphere. Here we utilize data from the Salt Lake Valley to address the question: can CO_2 act as an inert tracer of atmospheric transport and mixing in urban areas?

2. Materials and Methods

[5] CO_2 mixing ratios and the isotopic composition of CO_2 in air samples were collected and analyzed according to *Pataki et al.* [2003a]. Briefly, mixing ratios were monitored with infrared gas analyzers (LI-COR 7000 and LI-COR 6262, LI-COR Inc., Lincoln, Nebraska, USA) and data loggers (CR23x, Campbell Scientific, Logan, Utah, USA) recording 2-min running averages every 5 min. Air samples were dried with magnesium perchlorate ($Mg(ClO_4)_2$) before sampling, and CO_2-free air and WMO traceable calibration standards were introduced into the gas analyzers every four hours to correct measurements to known values. Measurements were conducted at four locations. The first site was on the roof of a four story building at the University of Utah, located on sloping terrain on the east side of the Salt Lake Valley near the foothills of the Wasatch Mountains (eastern foothills). The site was surrounded by building and trees that covered a wide range of heights (approximately 4–45 m). The second site was on the roof of a five-story building in downtown Salt Lake City located on the valley floor (downtown). The site was surrounded to the east by 3 to 5 story commercial and residential buildings and to the west by the downtown core with buildings with an average height of 15 to 20 m, but as high as 90 m. The third sampling site was on a 4.5 m

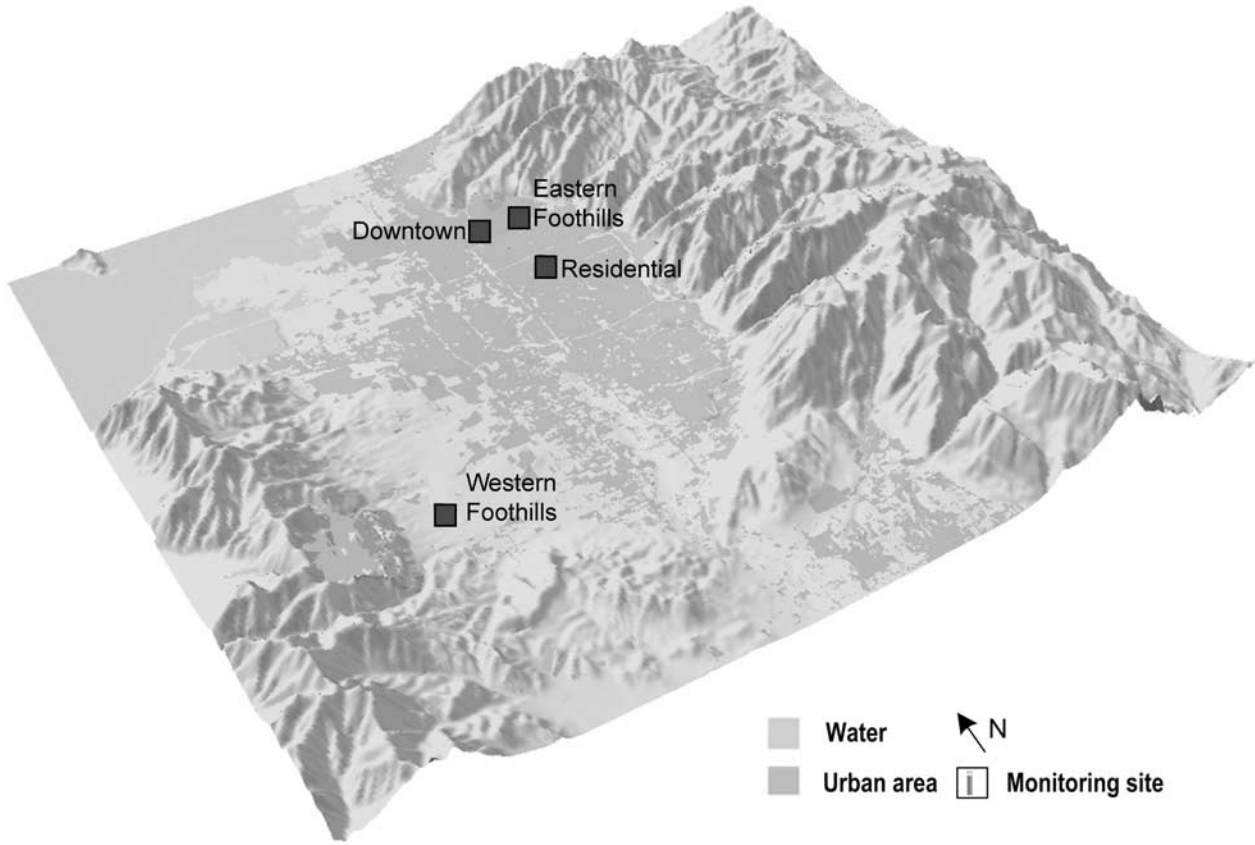

Figure 1. Location of the measurement sites in the Salt Lake Valley, Utah. The area shown is approximately 65 km^2. See color version of this figure in the HTML.

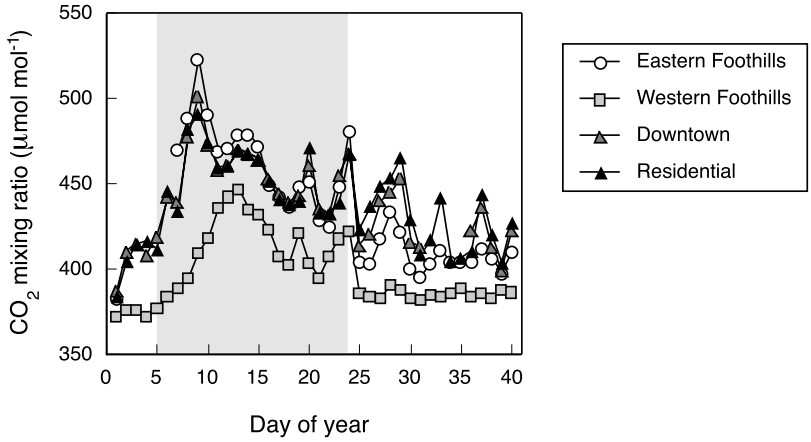

Figure 2. Daily mean CO_2 mixing ratios at the four measurement locations from 1 January to 9 February 2004. The shaded area shows the persistent cold pool period, which began on 5 January and mixed out on 25 January.

antenna pole in a residential neighborhood also located on the valley floor (residential). This site was surrounded by homes and trees with heights of approximately 6–12 m. The last site was a 9 m tower in a nonurbanized area consisting of agricultural and natural sage brush scrub land cover in the western valley foothills (western foothills). The roughness elements at this site were less than 1 m for several kilometers in all directions. The coordinate location, street location, and elevation of each site is shown in Table 1 and mapped in Figure 1.

[6] At three locations: the eastern foothills, downtown and western foothills, air samples were collected for isotope analysis according to *Pataki et al.* [2003a] at the same height as the continuous CO_2 mixing ratio measurements. Using an automated sampler described by *Schauer et al.* [2003], sets of 15 100 mL glass flasks were filled with ambient air periodically for analysis of $\delta^{13}C$ of CO_2 and mixing ratio by continuous flow stable isotope mass spectrometry (Delta Plus, Finnigan MAT, San Jose, California, USA). A minimum of five sets of 15 flasks were collected within a two week period. The precision of these measurements was 0.03‰ for $\delta^{13}C$ of CO_2 and 0.5 μmol mol^{-1} for mixing ratio. Isotope ratios are expressed relative to the V-PDB standard using conventional δ notation. These data were used to estimate the isotopic composition of total CO_2 in the atmosphere originating from urban emissions using the "Keeling plot" method, which removes the effect of background, nonlocal CO_2 on the isotope ratio [*Keeling*, 1958, 1961; *Pataki et al.*, 2003a, 2003b].

[7] Vertical temperature profiles were constructed using conventional twice-daily (05 and 17 LST) radiosonde observations collected by the U.S. National Weather Service Forecast Office at the Salt Lake City International Airport. Mean daily temperatures were based on routine temperature observations collected by the University of Utah Department of Meteorology using a thermistor (HMP 35C, Campbell Scientific, Logan, Utah, USA) mounted on an observing tower on the roof of an eight-story building directly across the street from the eastern foothills site. Regularly acquired wind data from the Department of Meteorology's MesoWest (available at http://www.met.utah.edu/mesowest) data set were also utilized for the western foothill, eastern foothill, and residential sites.

[8] PM 1 was monitored continuously beginning on DOY 21 at the eastern foothills and western foothills sites using DustTrak™ (simple photometer) monitors (model 8520 DustTrak Aerosol Monitor, TSI, Inc., Shoreview, Minnesota, USA) [*Hitchins et al.*, 2000; *Lee and Chang*, 2000]. The DustTrak™ monitors were calibrated by the manufacturer using standard road dust. Instruments were run side by side for 24 hours prior to the experiments to ensure comparable performance.

3. Results and Discussion

[9] Mean daily CO_2 mixing ratio ranged from 382.3 to 526.7 μmol mol^{-1}. The western foothills site showed the lowest mixing ratios consistently throughout the measurement period (Figure 2). However, from DOY 0 to 13 mixing ratios showed an increasing trend at all sites, including the western foothills (Figure 2). The persistent cold pool that formed during this time was characterized by dense fog throughout the depth of the cold pool. Elevated but variable mixing ratios persisted through DOY 25 when mixing ratios declined at all sites during the "mix-out" of the persistent cold pool that begin on DOY 5. The mix-out was associated with a synoptic scale system with sufficient strength to scour out the pollutant-trapping, persistent cold pool. After the mix-out, mean daily mixing ratios declined to about 380 μmol mol^{-1} in the western foothills and remained relatively constant for the remainder of the study period. In contrast, mixing ratios were consistently elevated at the other three sites, although the pattern shifted from the highest values occurring at the eastern foothills site before the mix-out to the highest values occurring at the downtown and residential sites after the mix-out (Figure 2).

[10] The diurnal pattern of CO_2 mixing ratio was also significantly affected by cold pool depth. During the cold pool on DOY 11–12, mixing ratios were fairly uniform among the four sites (Figure 3), including the western foothills site which is outside the urbanized area of the valley. Soundings at 05 and 17 LST showed that the height

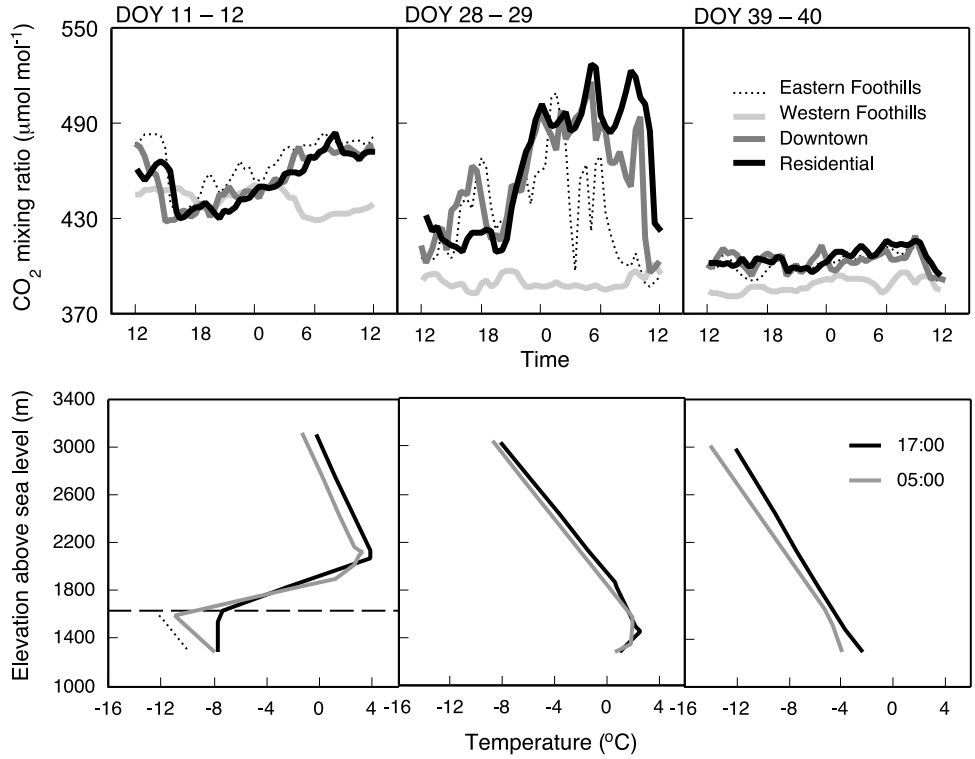

Figure 3. (top) CO_2 mixing ratios at the four measurement locations during three 24 hour periods of contrasting atmospheric conditions. (bottom) Upper air temperature profiles on the same days obtained from U.S. National Weather Service soundings at Salt Lake International Airport. The solid lines show evening soundings collected at 17:00 MST, and the shaded lines show morning data collected at 05:00 EST. On DOY 11–12, which fell during the persistent cold pool, the horizontal dashed line shows the base of the inversion and the dotted line shows the moist adiabatic lapse rate.

of the capping inversion was greater than 1500 m above sea level, and therefore above all four CO_2 measurement sites (Figure 3). During a much shallower inversion on DOY 28–29, mixing ratios were significantly elevated above ambient at all three sites except the western foothills site, which was located above the height of the inversion. CO_2 mixing ratio at this site remained close to the background, nonurban value, which was approximately 375 μmol mol^{-1} on average in Utah in 2002, the most recent period for which data are available from the U.S. National Oceanic and Atmospheric Administration's Utah station (NOAA Climate Monitoring and Diagnostics Laboratory, Carbon Cycle Group).

[11] While the exact mechanisms governing the evolution of cold pools are uncertain, these patterns appear to be associated with two distinct mixing regimes that occurred during the periods shown in Figures 2 and 3. As shown in Figure 4a, the first cold pool regime consists of a surface based temperature inversion with one or more sites near or above the top of the inversion. Limited vertical mixing occurs below this lid; most of the mixing that does exist is confined to the horizontal direction. Figure 4b shows the second type of cold pool regime with an elevated temperature inversion above a well-mixed layer. In this regime, pollutants emitted within the mixed layer are homogenized but confined to the volume of the cold pool. This regime is characterized by features consistent with cloud-topped mixed layers as shown in Figure 5. The observations on DOY 28–29 were consistent with the first type of cold pool regime (i.e., Figure 4a), while the observations on DOY 11–12 were consistent with the second (Figure 4b). Note that for both types of cold pool regimes, the winds within the Salt Lake Basin were extremely light (typically <2 m s^{-1})

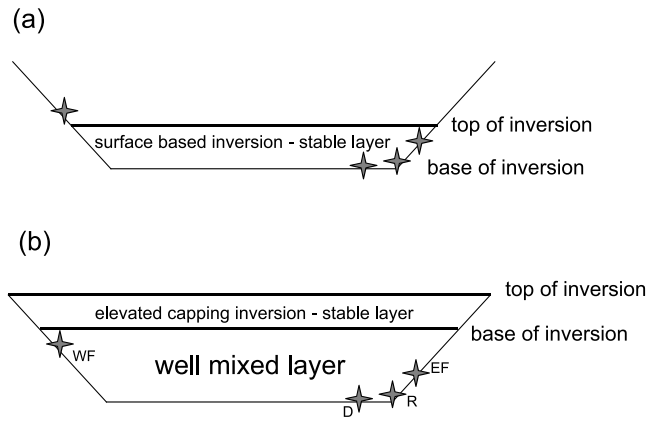

Figure 4. Schematic of two inversion scenarios: (a) a deep, well-mixed cold pool with an elevated inversion and (b) a surface-based inversion with mixing largely confined to the horizontal. WF is western foothills, EF is eastern foothills, R is residential, and D is downtown.

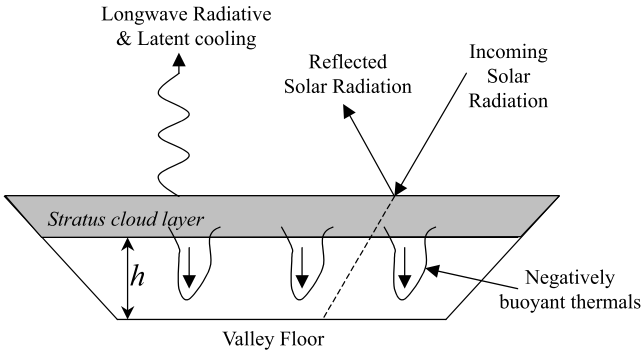

Figure 5. Schematic of the "top-down" convective cold pool mixing theory associated with cloud-topped radiative cooling. Adapted from *Houze* [1992].

and spatially varying depending on the location within the valley and proximity to different slopes. Figure 6 shows the clear diurnal pattern in wind direction associated with three of the sites.

[12] Nocturnal reductions in CO_2 mixing ratio occurred at the western and eastern foothills sites when the capping inversion was located near their elevation. For example, the capping inversion on DOY 11–12 was very near the elevation of the western foothills site, which experienced a slight reduction in CO_2 mixing ratio overnight, in contrast to increasing values at the other three sites. On DOY 28–29, the capping inversion was lower and near the elevation of the eastern foothills site, which showed a decline in mixing ratio after midnight. These mixing ratio changes may be a reflection of downward advection of the capping inversion or the mixing of ambient air into the cold pool by nighttime drainage flows and associated turbulence. Notably, during unstable conditions on DOY 39–40, CO_2 mixing ratios remained between 380–410 μmol mol^{-1} at all sites (Figure 3).

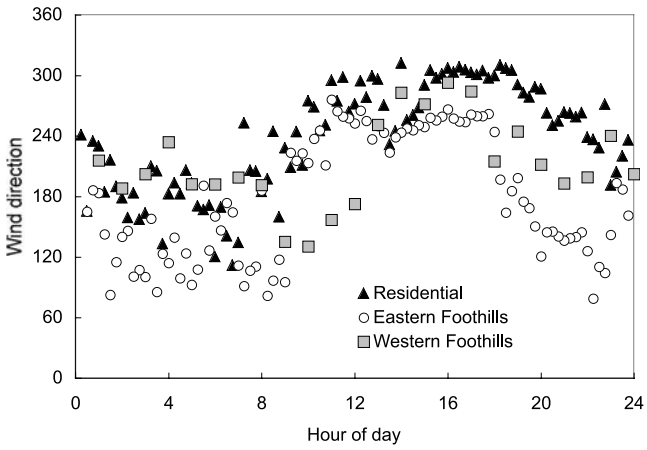

Figure 6. Week-long, ensemble average wind direction (DOY 11–17) for three site locations emphasizing the spatial variability of the winds and diurnal pattern at each site.

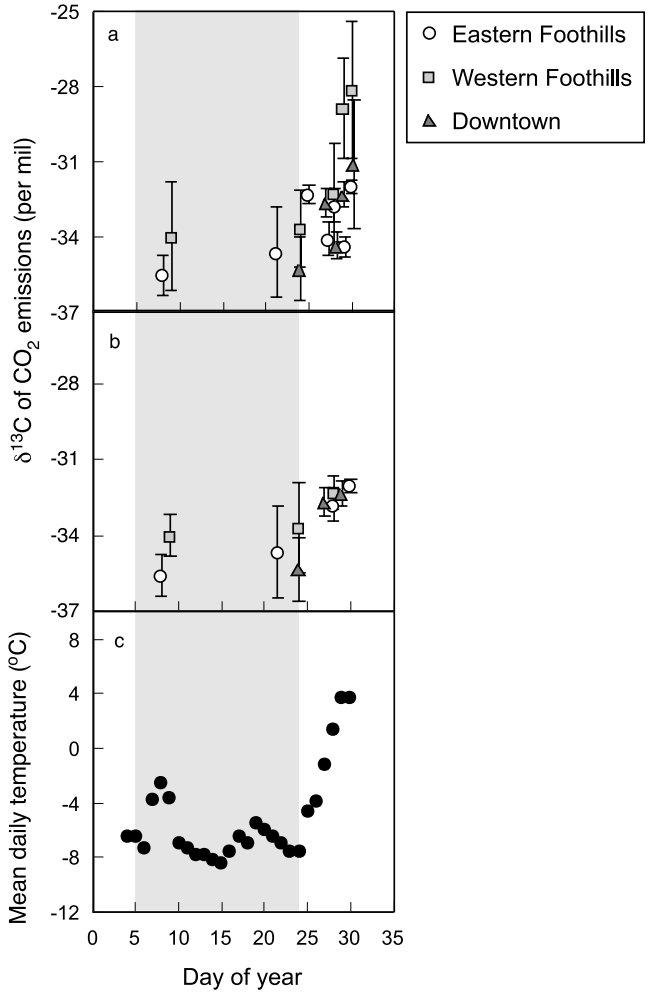

Figure 7. (a and b) Carbon isotope composition of total urban CO_2 emissions in the atmosphere. The influence of background, nonurban CO_2 on the atmosphere was removed from these data using the "Keeling plot" method after *Keeling* [1958, 1961] and *Pataki et al.* [2003a, 2003b]. Figure 7a shows all data for three measurement locations. Figure 7b shows data collected only when the all three measurement locations were beneath the capping inversion. (c) Mean daily temperature measured at the eastern foothills site. The shaded areas show the period of the persistent cold pool.

[13] We have described previously the application of the "Keeling plot" method for determining the isotopic composition of urban CO_2 emissions in order to identify CO_2 sources [*Keeling*, 1958; *Keeling*, 1961; *Pataki et al.*, 2003a, 2003b, 2005]. This method applies a simple mixing model to estimate the isotopic composition of source CO_2, as opposed to background, nonlocal CO_2, in air samples. We utilized this method during the current study at three of the four measurement sites. The carbon isotope composition of CO_2 sources showed both temporal and spatial variability, with a general trend toward isotopically enriched CO_2 at all sites after the mix-out of the cold pool on DOY25, a period that marked a warming trend with increasing mean daily temperatures (Figure 7c). We have described that the major

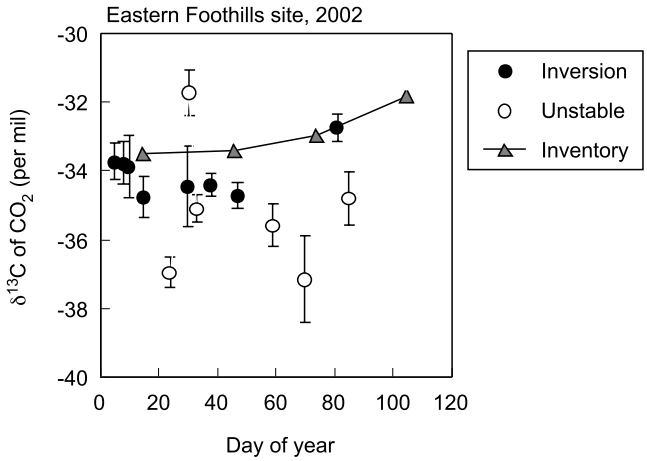

Figure 8. Carbon isotope composition of total urban CO_2 emissions at the eastern foothills measured in winter 2002. The circles show data reported by *Pataki et al.* [2003a]. Open circles show data collected during unstable conditions, or when the eastern foothills site was above a shallow capping inversion. Solid circles show data collected when the eastern foothills site was below a capping inversion. The triangles show the isotopic composition of CO_2 emissions estimated with monthly state fuel use statistics and census data as described by S. E. Bush et al. (Sources of variation in $\delta^{13}C$ of fossil fuel emissions in an urban region, submitted to *Geochimica et Cosmochimica Acta*, 2005).

local sources of CO_2 in the wintertime are gasoline combustion (isotopically enriched) and natural gas (isotopically depleted) [*Pataki et al.*, 2003a, 2005]. Therefore, as indicated by Figure 7a, the carbon isotope ratio of total CO_2 emissions became more enriched during the warming trend as the proportion of gasoline combustion increased relative to natural gas combustion; the latter is the primary source of residential heating in Utah (Energy Information Administration, U.S. Department of Energy, available at http://www.eia.doe.gov).

[14] The large degree of temporal and spatial variability in the isotopic composition of source CO_2 was eliminated when data collected at stations above the capping inversion were excluded (Figure 7b). These results suggest that during certain times in the evolution of a persistent deep cool pool, the air mass beneath the capping inversion is relatively well mixed throughout the valley. In other words, the measurement footprint is very large during these events, obscuring local effects on sources and emission rates. The uniform mixing ratios and isotopic composition of CO_2 at the western foothills site relative to the other, lower-elevation sites during the deep inversion (Figures 1 and 2) strongly support this hypothesis, as this site is not urbanized and is remote from large local CO_2 sources. In addition, as shown schematically in Figure 5, the presence of near-moist adiabatic lapse rates at low levels (an indicator of a well-mixed atmosphere), surmounted by stratus and stratocumulus clouds near the base of the capping inversion, is consistent with cloud-topped mixed layers, which feature turbulent mixing driven by cloud top radiative cooling [*Houze*, 1992].

[15] With this interpretation, we reanalyzed data collected in 2002 at the eastern foothills site, which showed a high degree of temporal variability in the carbon isotope ratio of source CO_2 during the winter [*Pataki et al.*, 2003a]. Although deep cold pools were uncommon in 2002, we identified eight measurement periods in which the eastern foothills site was below the capping inversion (Figure 8). In comparison to values collected during shallow inversions or unstable conditions, the deep cold pool values were more similar to the integrated isotopic composition of county-wide CO_2 emissions estimated by monthly state fuel use statistics and scaled to Salt Lake County with population and employment data from the U.S. census (Figure 8). This lends support to the hypothesis that mixing ratio and isotopic measurements taken below the capping inversion during deep cold pool events were representative of a well-mixed air mass with a large spatial footprint.

[16] During the deep well-mixed cold pool regime, CO_2 mixing ratio was well correlated with PM 1.0 at the eastern foothills site (Figure 9). While CO_2 and particulates both originate from combustion, a consistent correlation is not expected as the proportion of gasoline versus natural gas combustion should influence the slope of the relationship, i.e., more particulates are emitted in combustion of gasoline then natural gas relative to CO_2. Following mix-out of the cold pool the change in the CO_2-PM 1.0 relationship is opposite of what was expected on the basis of the known CO_2 sources. As daily temperatures warmed after mix-out, the reductions in natural gas combustion from residential heating relative to gasoline combustion should have caused an increase in the slope of the relationship between CO_2 and PM 1.0, as natural gas combustion produces fewer particulates than gasoline. However, the opposite trend was observed. We speculate that there are two possible explanations for the reduction in the slope: (1) Residential wood-burning declined during the warming period and resulted in a decline in local particulate sources or (2) local sources remained relatively constant and the decline in slope was dominated by local conditions rather than valley-wide air

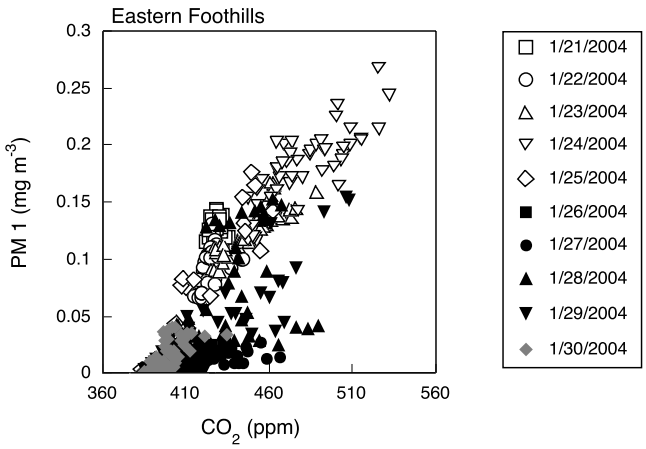

Figure 9. Relationship between PM 1 and CO_2 mixing ratio at the eastern foothill site. Points show half hourly values with symbols indicating different days from 21 to 30 January. The open symbols show data collected during the persistent cold pool, which ended on January 25.

masses because the site was above the shallow cold pools that occurred in late January. The decrease in slope may reflect a shift to samples influenced more strongly by local conditions with lower PM sources after the persistent cold pool ended. This hypothesis is supported by Figure 8, which shows that during unstable conditions or shallow inversions, CO_2 at the eastern foothills site is often more depleted in ^{13}C than the valley as a whole; i.e., more influenced by natural gas rather than gasoline combustion. Notably, on 28 and 29 January the relationship between CO_2 and PM 1.0 showed the greatest variability (Figure 9). Soundings suggest that the eastern foothills site may have been in and out of the cold pool on these days (data not shown), and at times may have seen turbulent bursts. Increases in PM relative to CO_2 may have occurred periodically as valley cold pool air masses advected up to the eastern foothills site, creating a high degree of scatter in the PM-CO_2 relationship.

4. Conclusions

[17] In this study the observed temporal and spatial variability in CO_2 mixing ratios in the Salt Lake Valley was consistent with a dominant influence of atmospheric transport and mixing, particularly in relation to the formation and mix-out of a persistent cold pool. During the deep cold pool, mixing ratios beneath the capping inversion were fairly uniform at four measurement locations, including a remote, nonurbanized location with few local CO_2 sources. This result is consistent with the presence of stratus and stratocumulus clouds and near-moist adiabatic lapse rates beneath the capping inversion associated with a cloud-topped mixed layer. Isotopic measurements supported a well-mixed air mass during this period, and largely reflected the proportion of CO_2 derived from gasoline versus natural gas combustion on a valley-wide scale. Following mix-out of the inversion, changes in the relationship between CO_2 and PM 1.0 were also observed to due changes in pollution sources and the measurement footprint.

[18] Under more idealized terrain and atmospheric flow conditions than those considered here, footprint analysis techniques can provide useful estimates of sources of pollutants and fluxes [*Schmid*, 2002]. We suggest that CO_2 mixing ratios, which are not commonly monitored in most urban areas at present, can provide useful information regarding atmospheric transport and mixing in complex terrain such as mountain basins. Coupled with measurements of the isotopic composition of CO_2 and measurements of other atmospheric constituents correlated with combustion sources, these measurements can enhance our understanding of the interactions between human activities, atmospheric processes, and emissions of greenhouse gases and atmospheric pollutants in urban environments.

[19] **Acknowledgments.** We thank A. Schauer, T. Dance, S. Bush, and C. Cook for their assistance in data collection; P. Torrens for providing a local land cover map; and the Salt Lake City Corp. and Kennecott Utah Copper Corp. for access to their facilities. This study was supported by grant ATM 02157658 from the U.S. National Science Foundation.

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