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Interannual variability of ozone and carbon monoxide at the Whistler high elevation site: 2002–2006

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Abstract

In spring 2002, an atmospheric measurement site was established at the peak of Whistler Mountain in British Columbia, Canada to measure trace gases, particle chemistry and physics, and meteorology. This paper uses continuous measurements from March 2002 to December 2006 to investigate the influence of trans-Pacific transport and North American forest fires on both O_3 and CO at Whistler. Annual mean mixing ratios of O_3 and CO were 41 ppbv (monthly means of 35–48 ppbv) and 145 ppbv (monthly means of 113–177 ppbv) respectively with both species exhibiting an annual cycle of late-winter to early-spring maxima and summer minima. The absence of a broad summer O_3 peak differs from previously-reported high altitude sites in the western US. The highest monthly-averaged O_3 and CO mixing ratios relative to the 5-year monthly means were seen in fall 2002 and spring 2003 with increased O_3 and CO of 10 % and 25 % respectively. These increases correspond to anomalously-high values reported at other Northern Hemisphere sites and are attributed to fires in the Russian Federation. Air mass back trajectory analysis is used to associate the mean enhancements of O_3 and CO with trans-Pacific transported or North American air masses relative to the Pacific background. Mean values of the enhancements for March to June were 6 ppbv and 16 ppbv for O_3 and CO respectively. In summers 2002–2006, higher CO and O_3 mixing ratios were always observed in North American air masses and this relative enhancement co-varied for each year with the western US and Canada total wildfire area. The greatest enhancements in O_3 and CO were seen in 2004, a record year for forest fires in Alaska and the Yukon Territory. In August 2004, average O_3 and CO mixing ratios were 13 and 44 ppbv above background values.

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1 Introduction

Factors affecting lower tropospheric ozone are important for the development of policies for local and regional pollution control as well as for evaluating the effectiveness of these controls. Ozone is both a constituent of smog and a greenhouse gas, and it plays a role in controlling the oxidizing capacity of the troposphere. Tropospheric ozone (O_3) is formed primarily by the oxidation of carbon monoxide (CO) and non-methane hydrocarbons (NMHCs) in the presence of nitrogen oxides (NO_x). Emissions of ozone precursors are from both anthropogenic and biogenic sources. NMHCs are emitted from vegetation, fossil fuel combustion, and manufacturing; major sources of NO_x include fossil fuel burning, lightning, emissions from the biosphere, and stratospheric injections. Biomass burning is also a significant source of CO, NMHCs and NO_x (Crutzen and Andreae 1990; Galanter et al., 2000; Simpson et al., 2011) and during the boreal fire season has been shown to influence regional ozone in North America (Wotawa and Trainer, 2000; McKeen et al., 2002).

On the west coast of North America, the contribution to regional ozone of trans-Pacific transported ozone and its precursors also needs to be considered. Over the past two decades, observational and modelling studies have shown that particle and trace gas concentrations throughout the Northern Hemisphere are influenced by intercontinental and hemispheric transport. Trans-Pacific transport of pollutants was first recognized as an issue in the late 1980s and early 1990s (Andreae et al., 1988; Kritz et al., 1990). Since then, numerous studies have documented pollution reaching the west coast of North America resulting in increases in ground level particles, O_3, CO, and pesticides (e.g. Husar et al., 2001; Jaffe et al., 2003; Bailey et al., 2000; Singh et al., 2009 and references therein). Jaffe et al. (1999) showed that transport of pollutants across the Pacific could have an impact on ground-level ozone in the Western US. Modelling studies have furthered the understanding of how transport may affect overall air quality and chemistry on the west coast of North America. Pollution crossing the Pacific Ocean was estimated to increase CO over western US by up to 25 % (Liang et

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al., 2004; Stohl et al., 2002). Anticipated three-fold increases in Asian industrial emissions from 1985–2010 were calculated to result in a 2–6 ppbv increase in ground-level ozone in the western US (Jacob et al., 1999).

To understand the factors affecting O_3 budgets, comparisons of O_3 with CO have been made in North American outflow and also at sites on the west coast of the US (Parrish et al., 1998; Weiss-Penzias et al., 2004; Honrath et al., 2004; Val Martin et al., 2006). The basis for the CO and O_3 comparison is the assumption that CO mixing ratios are directly related to CO and O_3 precursor emissions such as non-methane hydrocarbons and nitrogen oxides, and that changes in CO are indicative of changes in O_3 precursor species and thus would exhibit a relationship with net ozone production (Parrish et al., 1993, 1998; Honrath et al., 2004). Annual and interannual variability in CO is strongly dependent on both fossil fuel emissions and biomass burning (Novelli et al., 2003; Wotawa et al., 2001; Yurganov et al., 2005).

Trends in background ozone have been identified for several sites in western North America. Jaffe and Ray (2007) have examined 20 years of data (1987–2004) and found an average increase of 0.26 ppbv/year of O_3. Parrish et al. (2009) have found a similar trend from looking at west coast marine boundary layer sites (0.34 ppbv/year). Chan and Vet (2010) used a muti-site cluster analysis and also found a significant upward trend in background ozone along the west coast of North America. Similar trends have recently been identified in free tropospheric measurements through a comprehensive integration of data from multiple years and west coast studies (Cooper et al., 2010). Reasons for these trends are unclear and several possibilities have been suggested including regional continental or marine emissions, biomass burning and trans-Pacific transport (Cooper et al., 2010; Jaffe and Ray 2007; Dalsoren et al., 2010; Jaffe, 2010).

Relationships between O_3 and CO in North America over multiple years have mostly been made at ground-level sites (e.g. Parrish et al., 1998). Although transport of pollutants is observed at ground-level sites, significant transport occurs in the free troposphere and therefore, the frequency of observable transport events should increase at a high elevation site (VanCuren and Cahill, 2002). In addition, measurements in

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the free troposphere, largely unaffected by local emissions, are valuable to understand the regional and long-range emission and transport influences. Most coupled CO and O_3 data from the free troposphere come from aircraft campaigns which often include a detailed suite of measurements with good spatial coverage but are of limited time duration (e.g. Bertschi et al., 2005; Liang et al., 2007). Integration of satellite measurements with aircraft and ground-based measurements (e.g. Zhang et al., 2008) further the understanding of pollutant transport and transformation.

High elevation surface sites in western North America can provide long-term measurements in the lower free troposphere. Long-term records of ozone are available from several US National Park sites with varying degrees of influence from regional pollution (Jaffe and Ray, 2007). The Mt Bachelor site in central Oregon (Weiss-Penzias et al., 2004) is well situated for background free-tropospheric chemistry and atmospheric measurements began there in spring 2004.

In March 2002, Environment Canada began measurements of particles and some trace gases at the peak of Whistler Mountain, Whistler, British Columbia (hereafter called Whistler Peak). Situated about 100 km from the west coast of Canada, the Whistler Peak site (2180 m a.s.l.) was established to provide a baseline of particles and trace gases in the lower free-troposphere, and to examine incidences of trans-Pacific transport of dust and pollution into western Canada. This paper presents O_3 and CO measurements from 2002–2006, including their annual and interannual variability. Enhancement of CO and O_3 for trans-Pacific air masses is identified for springtime versus summer and compared to the relative enhancement in North American air masses above the Pacific background. The summer enhancements observed in O_3 and CO are then related to the interannual variability of forest fires.

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2 Experimental

2.1 Site details

Whistler Mountain, approximately 100 km north of Vancouver is part of the Coast Mountain range, which is a subset of the Pacific Cordillera extending along the west coast of North America from Alaska to Southern California. This high elevation site (50.06° N, 122.96° W, 2180 m a.s.l.) was chosen because of its relatively high exposure to the background troposphere, low impact expected from local pollution, and year-round accessibility. It is located just above the Whistler glacier, approximately 400 m above the tree line. The mountain peak is usually snow covered except for July to September. Whistler village lies just north of the site at about 650 m a.s.l. elevation. Vegetation in the surrounding valleys is mixed forest, dominated by coniferous trees.

Samplers are located in the chair lift operator's hut. Access is provided through Whistler-Blackcomb mountain operations. The site operates year round although for about one month during each of the late spring and fall, operator-required sampling is suspended because of limited site access. Pollution influences from trucks, snowmobiles or snow grooming equipment are readily identified and have been removed from the dataset.

Trace gases are sampled through a 1/4″ Teflon sample line, with a 5 µm in-line Teflon filter to remove particles and mounted approximately 5 m above the ground. All site measurements were recorded, and are reported here as Pacific Standard Time (PST).

2.2 Trace gases ozone and CO

Carbon monoxide was measured continuously with a Thermo Environmental Instruments Inc. Model 48C-Trace Level analyzer which determines CO by non-dispersive Infrared Spectrophotometry (NDIR). Instrument zeros were done every two hours for the period of March 2002–2005, every half-hour for March 2005–2006, and hourly after March 2006. Installation of a thermostatically controlled fan maintained the room

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temperature to ±0.5 °C. Calibrations were carried out approximately 3–4 times per year with a NIST traceable standard and were stable throughout the four year period, changing by less than 3 %. The detection level was about 19 ppbv and the uncertainty of reported CO concentrations was within ±5 ppbv, taken as one standard deviation of the instrument zero.

Ozone was measured with a Thermo Environmental Instruments Inc. UV absorption monitor (TECO 49C). Ozone zeros were carried out every 48 h with an in-line Koby air purifier cartridge. The instrument was calibrated every 3–4 months with a NIST traceable Dasibi Model 1008 PC ozone calibrator. Over the period March 2002 to March 2006, the calibration factors varied by less than 1 %; the detection level was about 0.5 ppbv and the uncertainty of the measured ozone concentrations is within ±0.5 ppbv.

2.3 Meteorological measurements

Meteorological measurements at the site include temperature, pressure and relative humidity. Pressure is measured with a Vaisala PTB101B pressure sensor. Temperature and relative humidity are both measured with a Campbell Scientific HMP45CF probe. Additional meteorological measurements (wind speed and direction) are made available through Whistler-Blackcomb on an hourly time resolution.

Fog is detected with an optical cloud detector (MCD-05, Associated Weather Services, Inc).

Ten-day back trajectories with the arrival pressure specified at 750 mb (the approximate mean pressure at the site) were used, calculated with the trajectory model of the Canadian Meteorological Centre (D'Amours and Page, 2001). The model uses 3-D analyzed wind fields and trajectories are calculated on an X by Y grid. Trajectories were calculated for arrival time at Whistler every six hours.

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3 Results and discussion

3.1 Meteorology

Monthly average temperatures from all hours of data at Whistler Peak (March 2002 to December 2006) range from −8.3 °C in winter to 9.4 °C in summer (Fig. 1). Maximum temperatures are commonly in July and August, although during 2003 the maximum temperature was in June followed by lower-than-average temperatures during the autumn. The predominant wind direction is westerly to southwesterly with gust velocities reaching 150 km/h during winter storms.

Clouds also impact this high elevation site. Although the fog detector gives no information on cloud amount, it provides qualitative information on cloud frequency. Data are available for an entire year (April 2002–June 2003) and three fall-through-spring periods (September 2003–March 2004; October 2004–May 2005; September 2005–April 2006). The percent of time during each available period that the site was in cloud at each hour of the day ranged from 8–20 % for entirely cloud covered to 15–40 % for greater than 10 min out of an hour. Cloud coverage increased from morning to afternoon with a decrease again at approximately 16:00 PST.

Several approaches have been used to separate boundary layer from free troposphere influenced data. Weiss-Penzias et al. (2006) compared water vapour at the Mt. Bachelor site with that from nearby radiosondes. Kleissl et al. (2006), Kleissl et al. (2007) provided a detailed examination of the effects of buoyant upslope flow or mechanical lifting on the Azores Pico site through the use of observed vertical soundings supplemented by those from a European numerical weather model. A simplified approach is used here to estimate the timing of the boundary layer influence on the mountain peak CO and O_3 mixing ratios. In spring 2005 (May) and 2006 (March–June) Whistler Peak temperature data were supplemented by temperature data from the Environment Canada National Archives Climate Database, and additional temperature records from two temporary sensors (MadgeTech, Temp101) to provide temperature measurements at 300 m vertical intervals up the mountain. The calculated tempera-

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ture lapse rates from 1000 m to 2182 m were compared to the dry and wet adiabatic lapse rates to determine if the air mass was unstable, conditionally unstable, or stable with the assumption that the air mass was mixed under unstable conditions. A value of 1–3 was assigned to each hour based on the stability calculations (1 = unstable; 2 = conditionally unstable; 3 = stable), to estimate of the timing of the transition from stable to unstable conditions. Figure 2 shows the diurnal variation in this "stability index" and in water vapour for the month of May 2006. The calculations confirmed mixing to the site from lower levels during springtime with the transition from stable to conditionally unstable conditions at approximately 08:00–09:00 PST and return to stable conditions by 17:00–20:00 PST. Although this simple technique only addresses one possible reason for boundary layer influence, it provides some time of day estimates for data segregation, agrees well with a more detailed meteorological analysis over an entire year (Gallagher et al., 2011), and supports the time of day approach employed by Andrews et al. (2011) for several mountain-top sites. Very little valley influence is expected in winter. Daytime growth of the boundary layer to encompass the mountain site was also confirmed from aircraft profiles of particles and gases during the INTEX-B campaign of spring 2006 (Leaitch et al., 2009). Early morning profiles showed a decoupling of the boundary layer and free troposphere below the peak while the afternoon profiles most often showed a mixed boundary layer up to 3 km.

3.2 Trace gases

3.2.1 Influence of boundary layer or free troposphere

The extent of this boundary layer influence is considered next by examination of the diurnal cycle in O_3 and CO (Fig. 3). There is little diurnal variation in O_3 (Fig. 3a) during the winter months (DJF) as the snow covered surface suppresses convective lifting of the boundary layer to the Peak level. As the valley warms, during the late spring months and early summer (MJJA), the mountain site becomes more influenced by valley air mixed up to the site. A decrease in averaged ozone is observed at approx-

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imately 08:00–09:00 PST as the nocturnal inversion in the valley breaks up and air with lower ozone reaches the site. By mid-day, ozone begins to rise and often exceeds its nighttime value, peaking at approximately 17:00–18:00 PST. This late afternoon rise to values on average 1 ppbv higher than nighttime may be due to photochemically-produced ozone. The average maximum diurnal change during JJA is 3.5 ppbv. The spring and fall months show similar patterns with the ozone decrease happening later in the morning and with the overall diurnal change reduced; photochemical production is lower than during summer. The largest diurnal variation in ozone was during July 2004 at approximately 10 % change about a daily mean value.

Although CO is a good tracer for pollution and biomass burning, it also increases as a result of the oxidation of biogenic NMHCs (e.g. Hudman et al., 2008; Slowik et al., 2010). Despite these various sources, diurnal variations in CO are more difficult to observe. The maximum change is once again during the summer months with a total diurnal change of up to 6 ppbv (Fig. 3b). This change, however, is close to the sensitivity of the instrument and overall, a diurnal variation is not significant on a monthly average.

To minimize the impact of the boundary layer chemistry on calculating averages for the free troposphere, the monthly mixing ratios are calculated from nighttime data only (20:00–08:00 PST). It is noted, however, that 81 % of the averages showed differences of less than 1 % between all hours and nighttime only; the largest difference in monthly averages calculated with all data and nighttime only data was 2.2 % for O_3 (June 2005) and 1.6 % for CO during (August 2002, August 2004).

3.2.2 Ozone

Ozone for March 2002 to December 2006 at Whistler Peak is given in Fig. 4a as monthly averages of nighttime data with the 5-year average annual cycle superimposed. As also shown in Fig. 5a, the ozone mixing ratios mostly range from 20–60 ppbv (90 % of data are between 28 and 53 ppbv) with greater variability in spring and summer values. Monthly-averaged ozone varied from a high of about 45–50 ppbv in springtime

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to a low of 32–40 ppbv in summer (Table 2). Annual median mixing ratios ranged from 40 to 43 ppbv over the years 2002 to 2006 (Table 2). These are 15–20 ppbv higher than the annual medians (23–27 ppbv) reported at a regional coastal site in British Columbia for 1992–2001 (Saturna Island) but consistent with high elevation sites in the Western United States; annual medians at Lassen National Park, California (1995–2001) were 38–43 ppbv (Vingarzen, 2004). These values are also consistent with annual baseline values determined for the Canadian coastal sites (19 ppbv) and US coastal sites (39 ppbv), by Chan and Vet (2010) who acknowledged that site elevation was a major difference between the two groups.

The amplitude of the annual signal (approximately 14 ppbv) is 5 ppbv less than that reported for Cheeka Peak (Weiss-Penzias et al., 2004) or Trinidad Head (TDH), California (Oltmans et al., 2008) which are both lower-altitude marine boundary layer sites. The observed ozone cycle at Whistler differs from other high elevation sites on the west coast such as Rocky Mountain and Lassen National Parks (Jaffe and Ray, 2007; Jaffe, 2011). These sites have the spring ozone peak but also have a significant summer peak, sometimes exceeding the springtime maxima. The Whistler ozone data do not show this broad summer maximum and more closely resemble the annual ozone pattern from the coastal TDH site (Oltmans et al., 2008) although monthly median mixing ratios are 3–15 ppbv higher at Whistler (O_3 WHI = 0.7 (O_3 TDH) +19.8 ppbv; $R^2 = 0.74$, $n = 56$). The small secondary peak often observed in August at Whistler was not observed at TDH, and corresponded with increases in Whistler CO, possibly indicative of regional pollution or influences of biomass burning.

The highest positive anomalies in ozone in relation to 5-year monthly means (Fig. 4a) were in fall 2002 through spring and summer 2003 with ozone up to 10 % higher than the 5-year average. Histograms of O_3 over the summer (JJA) period (Fig. 5a) illustrate the shift in O_3 to higher values throughout summer 2003; the mode has shifted from 36 ppbv to about 45 ppbv. The summer 2004 distribution is bimodal with a high mode at 55-60 ppbv and low mode ~35–40 ppbv, corresponding to the location of the single modes for 2002, 2005, 2006. This higher shoulder mode in 2004 is related to air

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masses influenced by biomass burning and is also reflected in the distributions of CO (Fig. 5b).

3.2.3 Carbon monoxide

Monthly averages of nighttime-only carbon monoxide are shown in Fig. 4b. On average, CO ranged from 100 to 200 ppbv, exhibiting a strong seasonal cycle with the maximum value in springtime and minimum in summer. There is a higher degree of variability in summer than winter (the standard deviation for winter data is 14–19 ppbv and for June–August is 16–27 ppbv) resulting from increased regional emission sources during summer and an increase of boundary layer influence. This annual cycle is consistent with other remote sites on the west coast of North America (e.g. Cheeka Peak; Weiss-Penzias et al., 2004; Reidmiller et al., 2008).

The seasonal cycle in CO has been well documented (Novelli et al., 1998; Novelli et al., 2003) and is attributed to a combination of photochemistry and transport. It is characterized by a slow increase in CO throughout the fall and winter followed by a rapid decline in springtime as OH concentrations increase (Novelli et al., 1998). The main sources of CO in the Northern Hemisphere are combustion of fossil fuels, biomass burning, oxidation of methane and oxidation of non-methane hydrocarbons (NMHCs) whereas the main sinks of CO are oxidation with OH and transport of CO to the Southern Hemisphere (Novelli et al., 1998; Holloway et al., 2000).

Novelli et al. (1998) show that in the absence of biomass burning, not only are the CO concentrations reduced but also the maximum CO is shifted to earlier in the year to coincide with the OH minimum. At Whistler, the annual maxima occur in February or March except for 2003, a year with high Asian biomass burning influence (van der Werf et al., 2006), when the maximum was in April. The variation in monthly-averaged CO from year to year is lowest in February–March (9–12 ppbv) and highest in May (52 ppbv). Highest CO mixing ratios were observed throughout fall 2002 and spring–summer 2003 but did not continue through fall 2003.

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Autumn 2002 and spring–summer 2003 were periods of highest CO anomaly at Whistler Peak; mixing ratios averaged 21 % higher in September–October 2002 and 25 % in April–August 2003. Anomalously high CO during these months has been reported from other Northern Hemisphere sites and is attributed to Siberian biomass burning during the summers of 2002 and 2003 (Yurganov et al., 2005; Jaffe et al., 2004). The anomalies in Northern Hemisphere boundary layer stations calculated relative to 2001-2002 values were a maximum of 35 % in 2002 and 36 % in 2003 compared with total column anomalies of 19 % in 2002 and 25 % in 2003 (Yurganov et al., 2005). The CO anomaly is not observed through the fall of 2003. The maximum in Siberian biomass burning emissions (van der Werf et al., 2006) was in August of 2002 but in May of 2003. The timing of these maxima are consistent with the CO anomalies at Whistler Peak, that is the effect of the late summer 2002 maximum was observed through fall 2002 whereas the May 2003 maximum strongly influenced the spring.

In spring 2003, although CO was highest during periods of direct Asian transport, the anomalies were not caused just by a succession of these transport events. Figure 5b shows the frequency distributions of CO during June to August 2002 to 2006. In 2003, the entire distribution is shifted higher by approximately 20–25 ppbv; in other years, the background CO values are generally similar (10th percentiles for June-August are 99, 121, 94, 93, 94 ppbv for 2002–2006 respectively). The 2004 and 2006 spring and summer seasons had two modes at 110 and 130 ppbv for the June–August period. The summer of 2004 was a high year for biomass burning in Alaska and Yukon Territory (van der Werf et al., 2006) and these fires have been shown to influence pollution levels in eastern North America and across the Atlantic (Pfister et al., 2006; Val Martin et al., 2006; Lapina et al., 2006) A combination of regional fires in British Columbia and the Alaskan and Yukon fires affected the CO mixing ratios seen at Whistler site in June through August 2004. These varying sources are discussed in the following sections.

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3.3 Effect of trans-Pacific transport

The influence of various source and transport regions on O_3 and CO measured at Whistler was investigated by segregating the data by 750 mb air mass back trajectories as calculated by the CMC trajectory model (D'Amours et al., 2001). Boxes were defined to represent different air mass origins and pathways (Fig. 6). CO and O_3 data were averaged over 6-h periods centered on the arrival time of the air mass. To reduce possible local boundary layer influences and problems with trajectories over mountainous terrain, CO and O_3 associated with trajectories having spent more than 12 of their final 48 h at altitudes less than 1000 m a.s.l. were eliminated from the analysis. The defined boxes are of different sizes and thus there is an inherent bias in the total amount of time a given trajectory can spend in a particular box. Thus the assignment of a six-hour mean CO or O_3 value to a back trajectory box is based on the residence time of the trajectory above a seasonally varying threshold residence time. For a given trajectory, at least 40 % of the total residence time within a box region was determined as the optimal threshold. This threshold maximized the largest number of samples that could be kept for subsequent analyses and minimized the number of trajectories that were attributed to multiple box regions. Mean values of O_3 and CO calculated for threshold values of 20 % and 60 % of the seasonally varying maximum are also shown for comparison.

Seasonal and interannual differences in the relative transport through the boxes reflect the variations in the synoptic scale climatology controlled by the Aleutian Low and the Pacific High (Klock and Mullock, 2001). The composite 700 mb vector winds for Jan and July 2002–2006 illustrate the climatological flows. (NCEP Reanalysis data provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA, from their Web site at http://www.cdc.noaa.gov/ (Kalnay et al., 1996). In winter, zonal flow is generally stronger and the Aleutian low results in a slight southwesterly component of the flow over BC (Fig. 7a). In summer, the intensifying Pacific High and weakening Aleutian Low cause a more northwesterly flow over the south coast of British Columbia (Fig. 7b).

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Normalized transport in representative boxes is shown in Fig. 8 along with the 5-year mean CO at Whistler Peak. The frequency of trajectories originating in the Asian box (Box 7) is highest during (November–April) and lowest during (May–August) reflecting the stronger zonal transport during winter and spring. Transport via the northerly route and passing over Northern British Columbia and Alaska is stronger in spring and early summer (Box 4). In contrast, air masses passing along the west coast of the US (Box 2, not shown) are more often seen during summer. The frequency of transport to the site from the South part of the North Pacific (Box 8) remains relatively constant throughout the entire year.

Mean values of O_3 and CO for Boxes 7 and 8 are given in Fig. 9a and b. Box 7 represents air masses with Asian origin and trans-Pacific transport (t-P) and Box 8 represents the south part of the North Pacific, also defined here as representing the Pacific background. For O_3, differences between Asian-influenced (Box 7) and background (Box 8) air are less than 1.5 ppbv during November–January and July–August. The greatest differences are seen in March–June and September–October with an average of 6±2 ppbv more O_3 in t-P air masses above the clean background for the entire March–June period. For CO, mixing ratios in Boxes 7 and 8 are similar during July and August, but for all other months, the CO in t-P air masses ranges from 4 to 25 ppbv more than is seen in the clean Pacific air masses. In March–June, the mean CO enhancement is 16 ±2 ppbv. The similarities in Boxes 7 and 8 in July–August may be a reflection of the reduced zonal transport. The CO and O_3 values for Boxes 7 and 8 in July and August are lower than the mean values for all air masses.

Highest values of CO and O_3 in summer are dominated by local and regional events with transport from the North American boxes, i.e. either up the coast from California or from the north and influenced by Alaska, Yukon or Northern British Columbia. This may be due both to the influence of forest fires or also of urban centers south of Whistler, throughout the Pacific Northwest and northern California. The next section investigates the relative influences of trans-Pacific transport or biomass burning during the spring and summer.

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3.4 Effect of North American boreal fires

Influence of trans-Pacific transport and of North American sources varies from spring to summer as well as from year to year. Data from May and August are shown as representative of spring transport and biomass burning seasons respectively (Fig. 10). The pattern for June is similar to May while the July pattern is similar to August. Air masses from Box 8 are selected to represent the Pacific background. The O_3 and CO enhancements for trans-Pacific (t-P) (Boxes 6,7) or North American (NA) (Boxes 2, 3, 4, 5) air masses are relative to the Pacific background (Box 8) May 2002–2006. In May, there is no significant difference between O_3 in t-P or NA air masses in 2002, 2003, 2005 (i.e. no significant difference and each 3-6 ppbv higher than background (Fig. 10a). The exception is in 2006 when t-P air masses averaged approximately 11 ppbv above background whereas the NA boxes were lower than background. This implies an enhancement of 10–15 ppbv in ozone for t-P transport events during May 2006 relative to NA air masses. This estimate is in line with ozone enhancements measured during the spring 2006 INTEX-B period (Leaitch et al., 2009; Walker et al., 2010). For CO (Fig. 10b), values in t-P or NA air masses were up to 5–15 ppbv higher than the background but this enhancement was not consistently dominated by one sector or the other. In 2004 and 2005, the North American influence was greater but in 2003 and 2006, CO was higher in t-P influenced air masses.

During the summer biomass burning period (as represented by August), the amount of ozone in the t-P boxes relative to background is approximately constant at 3–5 ppbv over all of the years (Fig. 10c). For the NA boxes, however, the greatest ozone enhancement relative to the background occurred in 2004 at about 13 ppbv, followed by an 8 ppbv increase in 2005. Enhancement of CO in NA air masses is always greater than in t-P air masses (Fig. 10d). For 2004, the corresponding enhancement in CO was 44 ppbv for the North American air masses whereas the trans-Pacific enhancement was only 12 ppbv.

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These largest differences in O_3 and CO are attributed to the North American boreal fires. Total estimates of area burned (Table 1; Sources: US National Interagency Fire Center; Canadian Interagency Forest Fire Centre) in western North America are also plotted on Fig. 10c–d. Notable regional influences in 2002 were late spring and early summer fires in Alberta. As noted previously, 2003 was a high year of boreal fires in Siberia which resulted in increased O_3 and CO at Whistler. However, because the entire CO distribution was shifted higher in 2003, the incremental differences in average NA CO above the defined background mixing ratios (Table 2) were not as great as those in 2004–2005. The most notable North American forest fires over these latter years were those in Alaska (AK) and Yukon Territory (YT) in summer of 2004 and in AK in 2005. The 2004 AK and YT fires were the largest on record in terms of area burned (Global fire Monitoring Centre; http://www.fire.uni-freiburg.de/) and they influenced much of Eastern North America (Pfister et al., 2006) and across the Atlantic (Honrath et al. 2006; Val Martin et al., 2006). In summer 2005, the total area burned in British Columbia was only about 50 % of the 10-year average but it was once again another very active burning season in Alaska; the estimated area burned reached almost 70 % of the 2004 season. In 2006, fire plumes from California and the US Northern Rockies were both observed at Whistler.

The August enhancement in both CO and O_3 for North American air masses relative to the background, co varies with the total area burned (Fig. 10c–d; see solid circles and red line). For several sites in western North America, Jaffe et al. (2008) have shown an increase in summer mean ozone (1998–2004) with area burned in a 10×10 degree region around each measurement site. This relationship is also evident for the Whistler site and relates increases in ozone to CO, a tracer for combustion. Boreal fires, in addition to their significance in controlling the CO budget, are substantial sources of NO_x and NMHCs (Wotawa et al., 2001) and thus additional precursors for ozone formation. These emissions have been shown to have an effect on regional ozone concentrations (Jaffe et al., 2004; Val Martin et al., 2006; Honrath et al., 2004; DeBell et al., 2004; Bertschi and Jaffe, 2005). Pfister et al. (2006) estimated the ozone production from

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Alaskan and Northern Canadian wildfires during summer 2004 to increase the ozone burden from the surface to 300 mb (50–70° N, 180° E–60° W) by approximately 7–9 %. The ozone increase at Whistler for August 2004, is about 40 % over the background. This analysis does not separate any NA anthropogenic from forest fire sources. However, when the ΔO_3(NA) for 2004 is compared to the ΔO_3(NA) for 2002 as an estimated ΔO_3 from anthropogenic sources, the resulting increase is still 30 % higher than baseline levels.

3.5 Relationship between ozone and CO

The relationship between O_3 and CO has been examined at various background sites to estimate the degree of photochemical ozone production in the atmosphere (e.g. Parrish et al., 1993; Parrish et al., 1998). The close covariance of O_3 and CO at Whistler is shown in Fig. 4c with the springtime maxima in CO sometimes preceding that for O_3. For several North Atlantic sites (Parrish et al., 1998) O_3 and CO were positively correlated in summer with O_3 versus CO slopes of approximately 0.3–0.4. Honrath et al. (2004) analyzed 2001 and 2003 O_3 and CO from the high altitude Azores site (Pico) and found slopes often closer to unity, in part attributed to longer transport times from North American source regions to the Azores. On the west coast of NA, Weiss-Penzias et al. (2004) found slopes of about 0.2–0.3 for O_3 to CO relationship in summer 2001. At Whistler, the slopes of the hourly O_3 to CO relationship as a function of month are shown in Fig. 11. The seasonality of the slopes introduced by Parrish et al., 1998 is also seen at Whistler, with positive slopes in spring and summer and negative slopes in winter. Summertime (JJA) slopes of O_3 to CO at Whistler range from a low of 0.21 in August 2005 to 0.62 in July 2005 (average slope JJA, 2002–2006 = 0.45). The reduction in slope from July to August 2005 is primarily driven by the mixed influence from CO produced by regional and local forest fires in August 2005 and their impact at the site. The correlations between O_3 and CO at Whistler, however, are lower than those found by Parrish et al., 1998 and closer to those observed at Cheeka Peak (maximum R^2~0.4, Weiss-Penzias et al., 2004). This greater degree of scatter

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between O_3 and CO in the Whistler measurements may be due to the proximity of regional boreal fires and the variation in the chemical processing time of the measured air masses.

The enhancement ratios of O_3 and CO above background values over specific periods also describe the regional O_3 production relative to precursor emissions. The ratios will depend on the values chosen to represent the background. Honrath et al. (2004) used the low modes of the CO and O_3 distributions; Pfister et al. (2008) chose average values of a non-fire impacted air masses; Val Martin et al. (2008) used the 20th percentile of all data for each season and year. The enhancement of O_3 relative to CO in NA air masses is shown in Fig. 12 where the background air masses for each month are defined here as the O_3 and CO mixing ratios in (a) Box 8, and (b) the 30th percentile of all data (Table 2). Although there is scatter, Fig. 12 shows largest enhancement of O_3 and CO in August relative to May. For months when ΔCO was greater than 10 ppbv, ratios of ΔO_3 to ΔCO range from about 0.2 to 0.6 with the median value of 0.31. The lowest ratio was in June 2004 when the site was influenced by a regional fire. This enhancement ratio is in the range of that calculated in Pfister et al. (2006), of approximately 0.26 ppbv/ppbv downwind of the 2004 fires. Mauzerall et al. (1998) reported different $\Delta O_3/\Delta$CO ratios depending on the age of the air mass ranging from 0.15 to 0.74 for fresh to aged plumes during the TRACE-A experiment. While CO is emitted from fires, O_3 is produced from the fire-emitted precursors as the air mass moves downstream. Pfister et al. (2008) found that the areas of maximum O_3 and CO are not necessarily co-located. The relationship between O_3 and CO varied and depended on the location of the fire source area relative to Whistler. Specific events observed at Whistler May–September 2004, 2005, 2006 are shown in Fig. 13 to illustrate the variability in the O_3 to CO relationship at Whistler. For all of these events, the rise in CO was greater than 20 ppbv above background values. Source regions are identified based on back trajectories and satellite analyses NOAA Hazard Mapping System Fire Analysis (http://www.osdpd.noaa.gov/ml/land/hms.html). For these examples, fire source areas were southern British Columbia, northern Washington State,

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Alaska, Yukon and California.

In 2004, fires in Alaska and the Yukon Territory (YT) were a major influence from approximately mid-June to end of July. In addition to this large source, a fire at Lonesome Lake (52.15° N, 125.44° W), approximately 300 km northwest of Whistler burned from approximately 21 June to early Aug with an estimate area burned of 22 745 ha (BC Fire Service). The highest observed CO mixing ratio of 414 ppbv on 24 June 2004 was most likely the Lonesome Lake fire superimposed upon the Alaska fire signal. Although both O_3 and CO are elevated throughout this period, during the large CO plumes associated with the Lonesome Lake fire, O_3 and CO are negatively correlated (Fig. 13a) (slope = −0.21, $R^2 = 0.85$).

Examples from 2005 show the change in the O_3 to CO relationship throughout the course of the event (Fig. 13b–c). During 9–12 August, CO and O_3 are anti correlated with a slope of −0.11 ($R^2 = 0.53$); for 14–17 August, the period of maximum CO (440 ppbv), there is a slight positive slope (0.07) although the correlation is weak ($R^2 = 0.19$). The NOAA emission tracker clearly shows the site in smoke this entire period. For the third period (24 August 03:00–27 August 2000), back trajectories once again show the air mass as coming from Alaska but the smoke analysis shows that the smoke plume was no longer directly over the Whistler area. In this period, O_3 and CO were positively correlated (slope of 0.29, $R^2 = 0.53$).

The final example (Fig. 13d) is from the end of the 2006 fire season. For this case, (1–6 September 2006), the source of this fire was southwestern US. Over 1–6 September period, CO rose from a baseline value of ∼110 ppbv to peak at 248 ppbv on 4 September. Ozone peaked at 65 ppbv, approximately 35 ppbv above the baseline level of 30 ppbv. The calculated O_3 to CO ratio for this episode is 0.25.

Although these examples (Fig. 13) are just a subset of the fire events influencing the Whistler high elevation site, they illustrate the diversity in the CO to O_3 relationship with an anti-correlation between O_3 and CO in fresh plumes (or plumes with a major source close to the sampling site); slight positive correlations in plumes accompanied by smoke but with estimated travel times of 1–2 days from the source, and positive

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slopes of 0.4 ppbv/ppbv for the plumes estimated with travel times of 3–5 days.

4 Summary

Year-round measurements of CO and O_3 from the Whistler high elevation site from 2002 to 2006 provide lower free tropospheric mixing ratios on Canada's west coast. Both O_3 and CO exhibit a seasonal cycle with a spring maximum and summer minimum similar to other background sites throughout the Northern Hemisphere. The annual cycle of O_3 is more similar to clean marine boundary layer sites on the west coast such as Trinidad Head and Cheeka Peak than to inland US high elevation sites that exhibit a broad summer maximum, which can possibly be attributed to regional pollution. O_3 mixing ratios were higher at Whistler than at the marine boundary layer sites by 3–15 ppbv. A boundary layer influence is observed at Whistler during warmer months throughout the daytime hours. The maximum diurnal cycle for O_3 is in JJA and found to be 3.5 ppbv about a daily mean.

Highest monthly-averaged CO was found in fall 2002 and spring 2003, coincident with increased CO found throughout the Northern Hemisphere as a result of elevated burning in Siberia. Background values were approximately 20–25 ppbv higher in 2003, values about 25 % above the five year mean.

The influence of different transport and source regions on measured values was examined through a back trajectory analysis. Largest differences in CO and O_3 for air masses originating in Asia relative to the south part of the north Pacific were in fall through spring. The mean enhancements in CO and O_3 for March–June were 11 ppbv and 4 ppbv respectively. During summer, very little difference was observed between trans-Pacific air masses and the clean background and highest CO and O_3 mixing ratios were found for air masses originating in North America. During the May transport season, the relative importance of North American or trans-Pacific contributions to increased CO and O_3 varied from year to year. In August, enhancements in CO and O_3 in North American air masses were always greater than in trans-Pacific air masses.

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The greatest effects were observed in 2004 and 2005 for both CO and O_3. This corresponded to higher estimates of forest fires throughout the western US and Canada.

Monthly correlations of O_3 with CO for 2002–2006 were positive in summer and negative in winter. In summertime, these slopes range from 0.21 to 0.62 with the lowest slope in 2005. Correlation coefficients were lower than those reported by Parrish et al., 1998 presumably because of the varying influence of regional forest fires and natural sources during summers at Whistler. The enhancement in O_3 relative to CO was estimated for May–September by calculating the average CO and O_3 above monthly background levels. For months when ΔCO exceeded 10 ppbv, the enhancement ratios ranged from about 0.2 to 0.6 ppbv/ppbv. Relationships between O_3 and CO from individual summer periods from 2003–2006, all influenced by biomass burning, showed a range of slopes from less than zero to 0.4. Periods with smoke impacting the site from relatively near sources frequently showed an anti-correlation between CO and O_3. When the impact of smoke on the site was from more distant sources, O_3 exhibited a positive correlation with CO.

The data show the importance of North American biomass burning on both the CO and O_3 mixing ratios measured at this lower free-troposphere site. Although trans-Pacific transport plays a role during winter through spring, it is biomass burning that appears most important during summer in contributing to periods of elevated ozone above the hemispheric baseline. This important source needs to be carefully considered in the development and evaluation of ozone mitigation strategies for Canada's west coast.

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Table 1. Estimates of area burned in western North America and for the Russian Federation for 2002–2006. (Sources: US National Interagency Fire Center; Canadian Interagency Forest Fire Centre; Goldammer et al., 2007).

	British Columbia	Yukon Territory	North West Territories	Alberta	Alaska	US Northwest[1]	US Northern Rockies[2]	Russian Federation
	Ha	Ha	Ha	Ha	Ha	Ha	Ha	MHa
2002	8586	35 669	27 089	496 515	880 865	446 648	153 909	10.6
2003	264 736	48 785	127 821	74 874	226 354	145 973	479 700	17.9
2004	220 516	1 720 324	515 621	236 090	2 689 528	49 629	27 035	4.4
2005	34 664	170 694	218 133	60 763	1 796 862	138 030	232 197	9.3
2006	139 201	95 034	53 398	118 762	107 754	386 851	920 240	13.1

[1] Northwest is Oregon and Washington;
[2] US Northern Rockies is the sum of Idaho, Montana, North Dakota, and Wyoming.

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Table 2. Monthly background values defined for CO and O_3 as average mixing ratios from Box 8. The 30th percentiles of all data are also shown in parentheses.

	O_3					CO				
	2002	2003	2004	2005	2006	2002	2003	2004	2005	2006
May	43 (42)	48 (46)	45 (44)	44 (42)	48 (43)	146 (140)	184 (179)	150 (148)	142 (132)	142 (149)
June	39 (38)	43 (39)	40 (37)	36 (34)	40 (38)	124 (123)	156 (153)	130 (125)	109 (112)	114 (123)
July	37 (32)	38 (33)	36 (33)	29 (29)	34 (29)	118 (102)	132 (126)	109 (106)	90 (93)	100 (94)
August	37 (31)	38 (37)	35 (30)	37 (33)	38 (34)	118 (104)	134 (132)	97 (95)	117 (109)	104 (107)
Annual Mean (Median)	41 (41)	43 (43)	41 (42)	41 (40)	42 (41)	153 (151)	160 (160)	141 (143)	138 (138)	135 (137)

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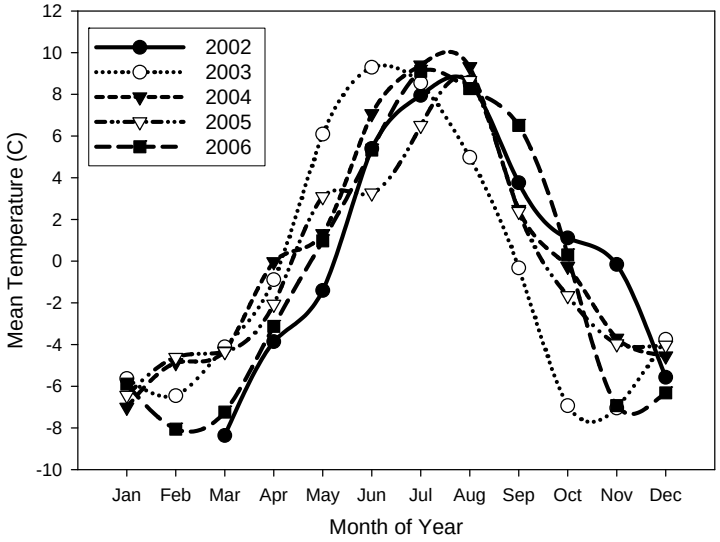

Fig. 1. Monthly averaged temperature at Whistler Peak site for March 2002 to December 2006.

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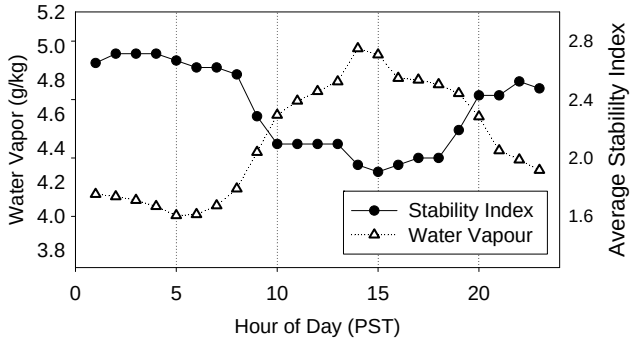

Fig. 2. Diurnal variation in water vapour and stability at Whistler Peak for May 2006. The stability Index of 3 describes stable conditions and 1 is unstable or assumed mixed conditions.

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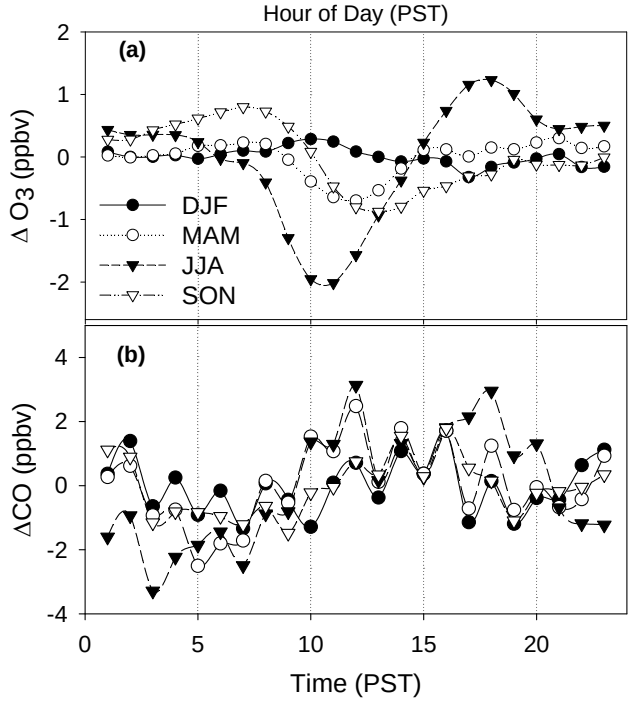

Fig. 3. Diurnal variation in **(a)** ozone and **(b)** CO for December–February, March–May, June–August, and September–November 2002–2006 at Whistler Peak. The plot shows the change in the mixing ration of ozone about a mean value throughout the day.

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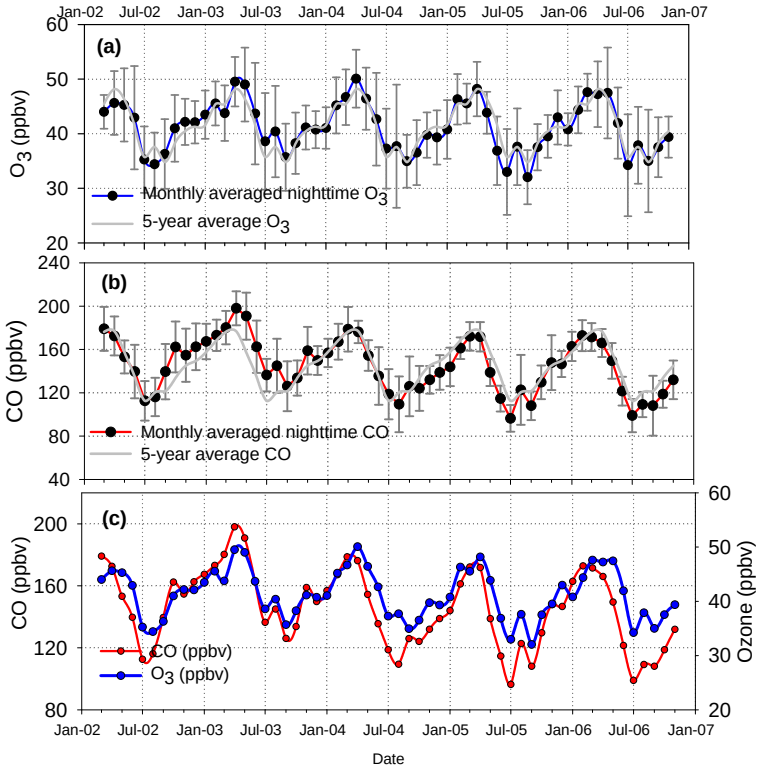

Fig. 4. Monthly averages of **(a)** O_3 and **(b)** CO nighttime data (18:00–08:00 PST) for March 2002 to December 2006 at Whistler Peak. Error bars are one standard deviation about the mean. The gray lines show the 5-year averages. **(c)** Monthly averages of both O_3 and CO.

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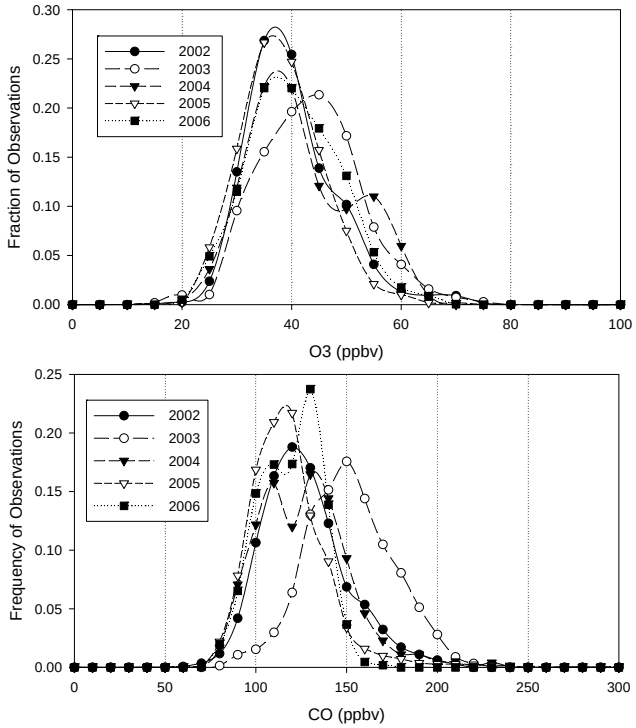

Fig. 5. **(a)** Frequency distributions of O_3 mixing ratios for June to August 2002–2006; **(b)** Frequency distribution of CO mixing ratios for June to August 2002–2006.

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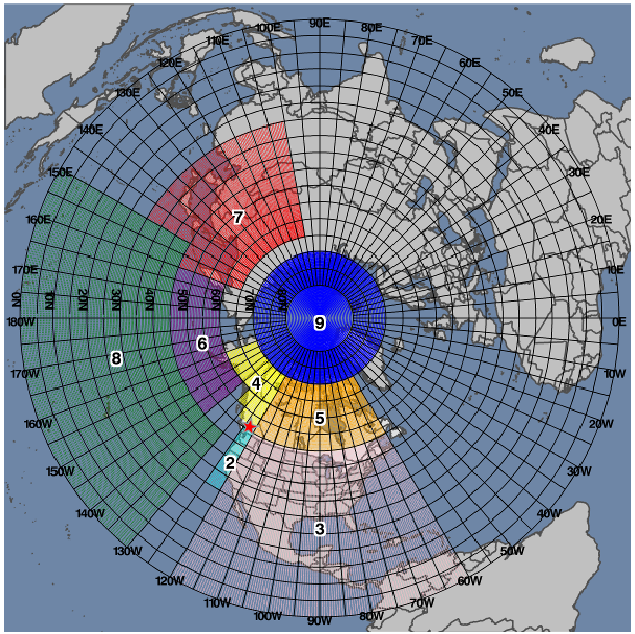

Fig. 6. Locations of boxes defined for trajectory analysis. Boxes 2–5 represent North American influence; Boxes 6–7 represent Asian sources and trans-Pacific transport; Box 8 is the southern part of the North Pacific.

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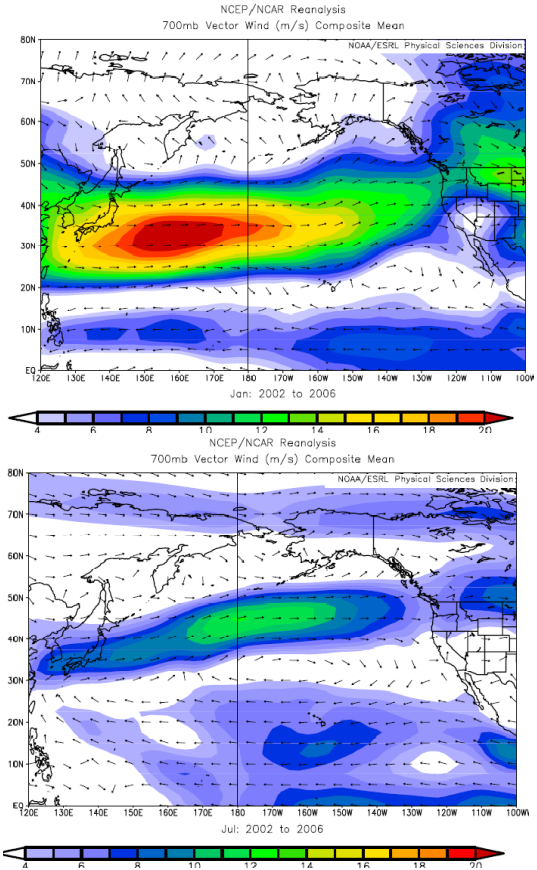

Fig. 7. Composite mean 700,mb vector winds from NCEP/NCAR (http://www.cdc.noaa.gov/) reanalysis for **(a)** January 2002–2006 and **(b)** July 2002–2006.

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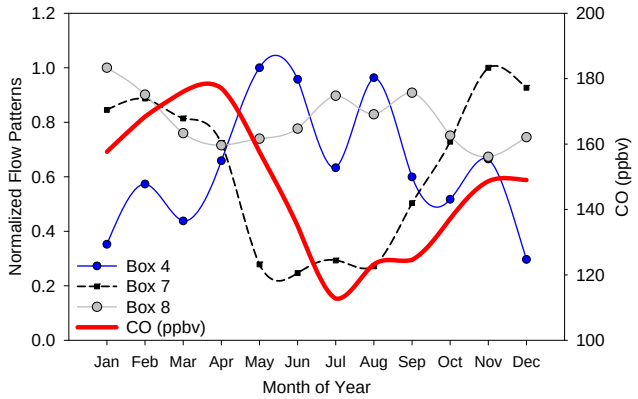

Fig. 8. Normalized frequency of transport through boxes defined in Fig. 12 as a function of month of year.

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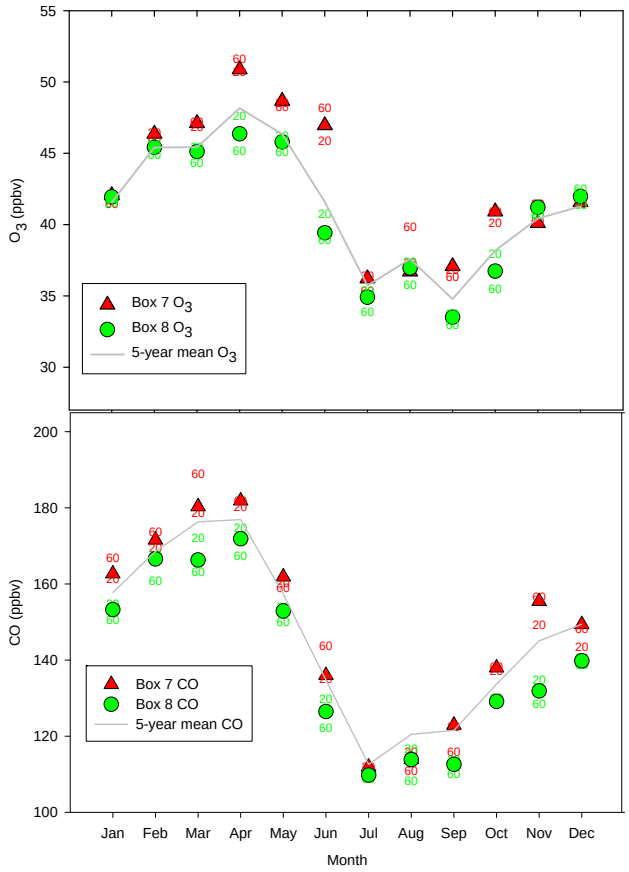

Fig. 9. (a) Mean values of O_3 associated with transport through Boxes 7 and 8 as a function of month of year. Boxes are assigned based on 40 % of the seasonally varying maximum time in a box. For comparison, mean values for 20 % and 60 % of time in a box are also shown. **(b)** As in **(a)** but for CO.

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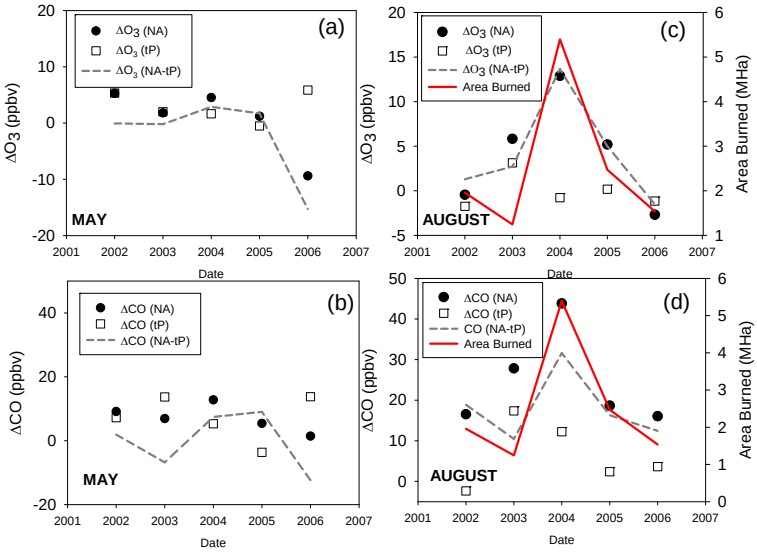

Fig. 10. (a) Mean O_3 mixing ratios in North American and in trans-Pacific boxes less the clean Pacific background in May 2002–2006. The dashed line shows the difference between NA and t-P O_3; **(b)** as in Fig11a but for CO; **(c)** as in Fig. 11a but shows ΔO_3 for August; **(d)** as in 11c but shows ΔCO for August.

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Interannual variability of ozone and carbon monoxide

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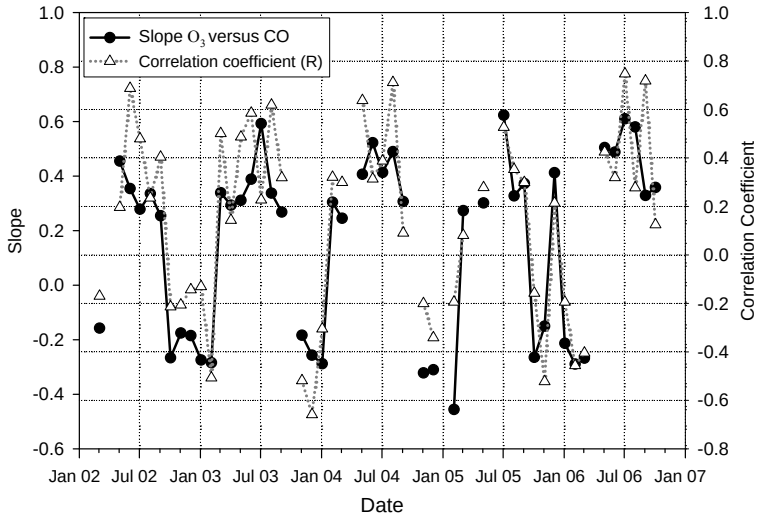

Fig. 11. Time series of slope and coefficient of determination for hourly O_3 versus CO relationships for each month based on the reduced major axis regression. The slopes are not shown when the *p*-value of the regression is less than 0.05.

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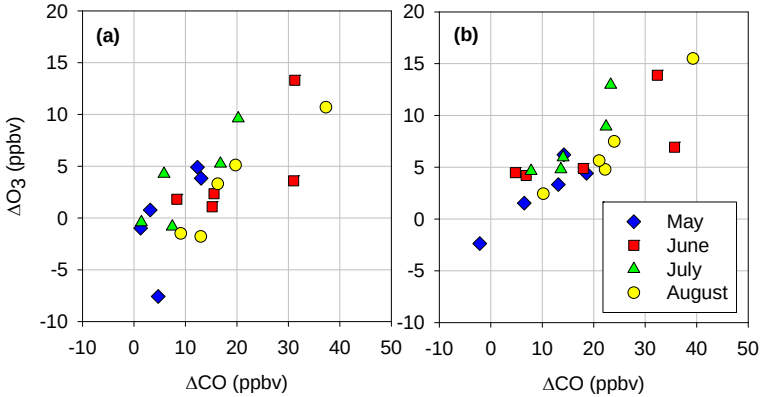

Fig. 12. Scatter plot of ΔO_3 vs. ΔCO on a monthly basis for 2002–2006 where ΔO_3 and ΔCO are the differences between the North American averages and the background values defined as the average from **(a)** Box 8 and **(b)** the 30th percentile for all data each month.

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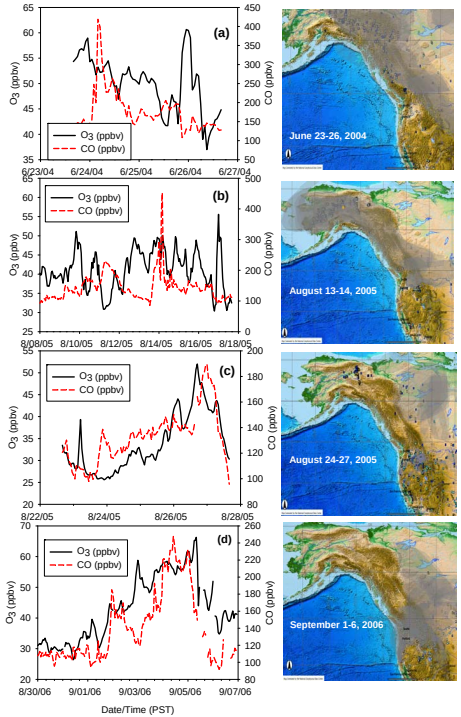

Fig. 13. Time series of O_3 and CO mixing ratios and analyzed smoke from the NOAA emission viewer for **(a)** 23–27 June 2004; **(b)** 8–18 August 2005; **(c)** 22–27 August 2005; **(d)** 31 August–6 September 2006.

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