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Six 'new' episodes of trans-Pacific transport of air pollutants

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Abstract

Chemical measurements of CO, O_3, non-methane hydrocarbons, aerosol chemistry, and aerosol scattering in air masses arriving at the west coast of North America demonstrate that a variety of chemical species can be transported across the Pacific from the Eurasian continent. In this paper, we analyze data from several ground sites in the Pacific Northwest and from aircraft observations in the region to identify six 'new' (i.e. previously unreported) episodes of trans-Pacific transport that occurred between 1993 and 2001. Of the six new episodes identified, one consisted of mineral dust combined with industrial emissions, while the other five appeared to consist of predominantly industrial emissions. In addition to gas and aerosol measurements, we calculated the Ångström exponent, an indicator of the aerosol size distribution, to help identify the aerosol characteristics in 4 of the cases considered.

Combining these data with previous reports, we find that there is a high degree of variability in these trans-Pacific episodes. In four episodes there were significant O_3 enhancement, with mixing ratios sometimes exceeding 80 ppbv. However, O_3 was only enhanced in episodes that were transported in the free troposphere and in the absence of mineral dust. In other words, transport in the boundary layer or transport of industrial emissions with mineral dust seems to preclude any significant O_3 enhancement. Clarifying such patterns increases our understanding of not only trans-Pacific transport of air pollutants, but also intercontinental transport in general.

Keywords: Long range transport; Ozone; Asia; Asian air pollution; Dust

1. Introduction

Gaseous and aerosol pollutants, including mineral dust, can be transported long distances, which under certain circumstances may have implications for local air quality (Prospero, 1999). In the Pacific region, both Duce et al. (1980) and Shaw (1980) have shown that Asian desert dust can be transported to the mid-Pacific. There are now numerous reports showing that not only dust, but also gases and aerosols from anthropogenic sources in Asia are transported to the Pacific (e.g. Merrill et al., 1989; Harris et al., 1992; Hoell et al., 1997; Jaffe et al., 1997; Perry et al., 1999; Bailey et al., 2000; Clarke et al., 2001).

Trans-Pacific transport of pollutants was at first suggested by Andreae et al. (1988) for non-seasalt sulfate. This was followed by a report by Kritz et al. (1990) who described an episode of rapid transport of Radon from Asia to California in the upper troposphere. Parrish et al. (1992) suggested that non-methane hydrocarbons observed on the California coast under westerly flow were of Asian origin. More recently, Jaffe et al. (1999) showed that a number of primary and secondary industrial pollutants (e.g. carbon monoxide

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(CO), peroxyacetyl nitrate (PAN), aerosols, non-methane hydrocarbons (NMHCs)) were transported across the Pacific to Washington State during an episode in March 1997. This was based on measurements at the Cheeka Peak Observatory (CPO) on the northwest tip of Washington State as part of the Photochemical Ozone Budget of the Eastern North Pacific Atmosphere (PHOBEA) experiment (Jaffe et al., 1999, 2001; Kotchenruther et al., 2001a, b) and using the Colorado State University—Regional Atmospheric Modeling System (CSU-RAMS, Pielke et al., 1992). The CSU-RAMS model was used to show that boundary layer pollutants were lifted in Asia during frontal passage and rapidly transported across the Pacific in the lower troposphere, consistent with the CPO observations (Jaffe et al., 1999).

Another documented event of trans-Pacific transport occurred in April 1998, when a major dust storm occurred in western China (Husar et al., 2001; McKendry et al., 2001). The dust was observed on satellite imagery as it crossed the Pacific in 5 days. Initially the dust was transported above 6 km, but was carried down to the surface over North America via large-scale subsidence and mountain waves (McKendry et al., 2001; Hacker et al., 2001). Despite the fact that the transport was indirect, surface concentrations of the dust were substantial. The dust was observed in North America by ground-based lidar, aircraft and surface aerosol monitoring networks (Husar et al., 2001 and references therein).

Satellite imagery can sometimes detect trans-Pacific transport episodes. For example, the well-documented April 1998 dust episode was detected by its spectral reflectance from sensors onboard the Sea-viewing Wide Field-of-view Sensor (SeaWiFS) satellite and by detection of the UV absorbing aerosols from the Total Ozone Mapping Spectrometer (TOMS) instrument on-board the Earth Probe satellite (Herman et al., 1997; Husar et al., 2001). However, to date, there is no evidence that these satellite sensors can give information on industrial pollutants in the Pacific. This is mainly due to the fact that the industrial pollution sources are not as concentrated as large dust or smoke events. Also, the light absorption by aerosols and the scattering of sub-micron aerosols is more efficient in the blue, where there is a larger background from Rayleigh scattering (R. Husar, personal communication, 2002). For example, in the trans-Pacific transport episode of industrial pollution from March 1997 (Jaffe et al., 1999), there is no evidence of this pollution in any satellite imagery examined. It is certainly possible that recently developed and future satellite sensors (e.g. Measurements of pollution in the troposphere (MOPITT), Global ozone monitoring experiment (GOME), etc.) will be able to routinely detect trans-Pacific pollution transport events. However, to date, only detection of Asian dust and smoke has been shown to be possible (Herman et al., 1997; Husar et al., 2001).

The problem of source attribution is complicated by several factors, including, the great distances involved, the chemical processing that occurs during transit, the low resolution of the global meteorological data, and the difficulty of finding chemical tracers that are unique to each potential source region. While most of the reports on trans-Pacific transport attribute enhanced pollutants to sources in East Asia, it is certainly possible that sources further west may also contribute (e.g. Bey et al., 2001). For longer-lived pollutants, such as CO and O_3, with lifetimes of weeks to months, global transit could be expected under some circumstances. Using a climatology of trajectories, Newell and Evans (2000) have suggested that European sources could also contribute to elevated pollutant concentrations in the Pacific region; however, to date, there is no observational evidence to verify this.

The inter-continental impact of Asian emissions has also been considered in several modeling studies (Berntsen et al., 1999; Jacob et al., 1999; Yienger et al., 2000; Mauzerall et al., 2000; Bey et al., 2001; Wild and Akimoto, 2001). Berntsen et al. (1999) used a global 3-D chemical transport model (CTM) to quantify the influence of Asian emissions on CO, O_3 and PAN in the air arriving at the CPO site in Washington State. The CTM showed that the influence occurs in two ways: (1) "events," whereby air masses containing relatively high concentrations of pollutants from Asia are rapidly transported across the Pacific; and (2) background contribution. According to this study, Asian emissions are responsible for about 10% of the O_3 in the air arriving to North America. The work of Yienger et al. (2000) also shows that the impact of Asia on North America has both an episodic as well as a background contribution. During events, a substantial contribution from a single source region is super-imposed on the background contribution, which results from many different sources.

An important question to address is: How large an impact does inter-continental transport have on air quality in the downwind region? At this point the answer is not certain; however, in at least one case (Husar et al., 2001) PM10 and PM2.5 concentrations seen in North America from transported Asian dust were substantial and approached levels associated with health impacts. However, this particular event is one of the largest, if not the largest, of the cases we have examined.

The possibility of inter-continental transport of O_3 is particularly important. This is because O_3 mixing ratios observed during spring in the Northeast Pacific are already relatively high (Kotchenruther et al., 2001a), and not far from levels known to cause vegetation impacts (Hogsett and Lee, 1998). Jacob et al. (1999) used a global CTM to determine the impact of

intercontinental transport of air pollutants by estimating how future Asian emission increases will impact ground level O_3 in North America. Based on their modeling study, Jacob et al. concluded that increasing emissions from Asia in the next two decades will increase the monthly mean O_3 mixing ratio in the background air arriving to North America by 2–6 ppbv. Lin et al. (2000) conducted an analysis of the distribution of surface O_3 in the US over the past two decades. They found that while peak O_3 values declined during this time, the median value increased. They suggested that this was due to increasing background O_3 due to increasing emissions in Europe and Asia. However until now, there has been no evidence for O_3 enhancements in North America as a result of trans-Pacific transport and the global model results have not been verified with observations.

In this paper, we utilize a variety of chemical and meteorological data to examine six cases of trans-Pacific transport of pollutants across the Pacific to North America. For this analysis we draw on data from the PHOBEA projects, as well as data from the "Interagency Monitoring of Protected Visual Environments" (IMPROVE) network, which includes several sites in the western US. The focus of this analysis is to identify significant episodes of long-range transport across the Pacific, and to examine the variations in gas and aerosol chemical composition that result. In particular, we will focus on O_3, which has not previously been seen in trans-Pacific transport.

2. Data and methods

We analyzed a variety of data sources from the Pacific Northwest to identify a number of trans-Pacific transport episodes. The IMPROVE network, which consists of approximately 80 sites, primarily in US National Parks (NP) (Malm et al., 1994; Eldred and Cahill, 1994) was one data source for this work. At each site aerosols are collected in two size bins: less then 2.5 μm diameter (fine mode) and between 2.5 and 10 μm aerodynamic diameter (coarse mode). Quartz and Teflon filter samples are collected for 24 h (midnight to midnight, local standard time) for 2 days out of every week, Wednesday and Saturday. No measurements are made on the other days. The fine mass filter samples are analyzed for total mass and a wide array of chemical species, including major ions, metals, and organic and elemental carbon (the data can be accessed on-line at http://vista.cira.colostate.edu/improve/). For this analysis we have used the IMPROVE data from Crater Lake National Park, which is the cleanest west coast site in the IMPROVE network, but with some additional data from the Mt. Rainier and Lassen Volcanic sites as well. Ozone data are available for the Mt. Lassen site from the National Park Service air monitoring network (http://www2.nature.nps.gov/).

Additional data came from the PHOBEA campaigns, which took place in the springs of 1997–1999, and included extensive observations of chemical species related to O_3 production (e.g. NO_x, PAN, CO, NMHCs and O_3). Observations were made at the CPO and from a King Air aircraft off the coast of Washington state (Jaffe et al., 2001; Kotchenruther et al., 2001a, b). At CPO, observations were made during March–April 1997 and 1998. Airborne measurements were taken during 14 flights made between 26 March and 28 April 1999. Aerosol physical properties (scattering at three wavelengths, aerosol absorption and number density) were also observed, both on the ground and from the King Air.

PHOBEA measurements continued during the spring of 2001 in the same region using a Beechcraft Duchess aircraft. This airplane has a ceiling of $\sim$6000 m above sea level (asl). The smaller Duchess aircraft, compared to the King Air used in 1999 necessitated a smaller instrument package. Measurements on-board the Duchess in the spring of 2001 included O_3, aerosol scattering (three wavelengths), temperature, relative humidity, and canister samples for CO and NMHCs. Further details on the Duchess sampling can be found in Price et al. (2002).

For both the 1999 and 2001 aircraft sampling, flights were conducted only on days with generally westerly flow to minimize sampling of local (North American) sources. However, in a few cases, boundary layer winds and enhanced concentrations suggested that local pollution may have been sampled. For this analysis, these few points have been removed from the data set.

For aerosol data in the vertical profiles, the total scattering coefficient in the green (σ_{sp}, 550 nm), the Ångström exponent ($\mathring{a}$) and the particle absorption coefficient (σ_{ap}, only available in 1999) are used. The Ångström exponent is derived from the wavelength dependence of scattering (see Eq. (1)) and is sensitive to the aerosol size distribution.

$$\mathring{a}(\lambda_1/\lambda_2) \equiv -\log(\sigma_{sp,\lambda_1}/\sigma_{sp,\lambda_2})/\log(\lambda_1/\lambda_2). \quad (1)$$

Fig. 1 shows a map of the Pacific Northwest (PNW) marked with the ground sites and the aircraft study region. Table 1 shows the coordinates and elevations for these sites.

Back trajectories were calculated using the NOAA-Hysplit program (Draxler and Hess, 1997; see also http://www.arl.noaa.gov/ready.html) using the FNL data set, with the exception of the 1993 trajectories. For these trajectories the National Center for Environmental Prediction (NCEP) reanalysis data was used. These are based on a high resolution meteorological grid ($1^\circ \times 1^\circ$) and model vertical velocities. For the surface locations, we calculate trajectories at multiple arrival

heights, both within and just above the local boundary layer. It should be kept in mind that there are a number of possible sources of error associated with trajectories, including low resolution meteorological data and/or sub-grid scale vertical motions. In general, model-diagnosed vertical velocities from a dynamically consistent model give the best results (Fuelberg et al., 1996; Stohl, 1998).

For the Crater Lake site, local meteorological data were not available. For this site we used isentropic trajectories to segregate the data set. While most trajectories arriving to Crater Lake come from the Pacific marine environment, we assumed that the longer an air mass spent over North America before reaching Crater Lake, the greater the probability that it picked up North American emissions and lost its marine character. Consequently, each trajectory was classified as "local" or "marine." A trajectory was considered local if it spent more than 24 h over North America. The 24-h period was necessary since Crater Lake is about 180 km inland from the Pacific coast. As each filter sample spans a 24-h period, we included in our analysis only those samples where both trajectories in the 24-h period were classified as marine. For springtime, about 50% of the filter samples were classified as marine, i.e. both the 0 and 12 GM time trajectories came from the marine sector.

Trans-Pacific transport events were identified based on the following criteria:

(1) Two or more species must be significantly elevated above the background values;
(2) Local winds must be consistently from the Pacific and away from any regional pollution sources for the duration of the event;
(3) Trajectories must be consistent with the local winds and indicate trans-Pacific transport.

To quantify the influence of episodic transport of gases and aerosols across the Pacific we first define the background concentration for that location and season. Then the enhancement during selected periods can be compared with this value. This is similar to the approach we have used previously to examine the long-range transport of CO in the Pacific (Jaffe et al., 1997). However, because the observations are often log-normally distributed, it is more appropriate to compare median values, rather then means.

Fig. 1. Map showing the US west coast and sampling sites.

3. Trans-Pacific episodes

3.1. Episode 1: 28 April 1993

The aerosol index (AI) data from the TOMS instrument onboard the Nimbus 7 satellite showed large values over China for much of the month of April 1993. The AI values were especially high in the later part of the month over both western and eastern portions of China. The AI images also suggest that some of this aerosol was

Table 1
Sampling locations used in this work

Site	Coordinates	Measurement elevation (m)
CPO	48°20′N, 124°36′W	480
Crater Lake NP	42°53′N, 122°08′W	1982
Mt. Rainier NP	46°30′N, 122°07′W	436
Lassen Volcanic NP	40°32′N, 121°34′W	1799
PHOBEA aircraft profiles	Region defined by the box with corners near: 48°N 125′W and 40°N 128′W	200–7400

transported into the mid-Pacific. Given that the TOMS instrument can only detect large aerosol concentrations and cannot detect aerosols below layer cloud layers, it is difficult to ascertain from the satellite data alone whether this aerosol was transported to North America. Furthermore, as the TOMS instrument is only sensitive to UV absorbing aerosols, it is not possible to determine the degree to which industrial aerosols were also present. To see if these aerosols were transported to North America, we used the IMPROVE aerosol measurements from three sites in the western US.

Table 2 shows the IMPROVE data for Crater Lake, Oregon, Mt. Rainier, Washington, and Mt. Lassen in Northern California for 28 April 1993. As with all of the IMPROVE samples, the filter samples were collected from 0–24 local standard time (PST in this case). For the background values we used the springtime marine means from Crater Lake for the period 1988–2000, as described above.

From the measured data, several composite variables were calculated (UCD, 1995), including ammonium sulfate, total organic mass, total elemental carbon and total soil mass. The data for 28 April 1993 show that the total fine particle mass was divided roughly equally between ammonium sulfate, organic and elemental carbon, and soil.

All three sites show significant enhancements in fine mass, sulfur and organics, but very different values for the coarse mass and soil component. The Mt. Lassen data showed a slightly higher soil component compared to Crater Lake, whereas the Mt. Rainier data showed an almost insignificant soil component. The much lower coarse mass and soil component for the Mt. Rainier data on 28 April suggests that the air mass which arrived at this site was from a different source region or that these particles had been significantly depleted of mineral dust prior to reaching the Mt. Rainier site. Some clues as to the cause of this difference can be found from the back trajectories. While back trajectories for all three sites on 28 April 1993 originate from Asia, there are differences in the starting heights and regions within Asia. For Crater Lake, the back trajectories (shown in Fig. 2a) return to near Japan, the Korean Peninsula, and northeastern China at a height of approximately 2–3 km in 8 days, and reach Mongolia in 10 days. For Mt. Lassen, the trajectories (not shown) also return to the Japan–Korea–NE China region in about 8 days, but at a height of 4–6 km over this industrial source region. At 10 days back, the Lassen trajectories arrive to Mongolia, 2–3 km above the surface. The back trajectories for Mt. Rainier (not shown) return to the boundary layer near the Korean Peninsula, at a height of 0.5–1.5 km above the surface. The Mt. Rainier trajectories do not reach Mongolia within 10 days. These trajectories suggest that air parcels reaching the Mt. Rainier site had greater influence from the industrial regions of East Asia (northeast China, Korea and Japan) compared to Mt. Lassen, which had larger influence from mineral dust sources.

In addition to the IMPROVE samples, there are ozone observations at the Mt. Lassen site. For 28 April 1993 the 24-h average ozone mixing ratio was 37 ppbv, compared to a long-term average for April of 44 ppbv. Thus, despite the large amounts of both industrial and mineral aerosol, ozone showed no enhancement, and

Table 2
IMPROVE aerosol data for 28 April 1993 and background values

Elevated species	Crater Lake (ng/m^3)	Mt. Rainier (ng/m^3)	Mt. Lassen (ng/m^3)	Background value (ng/m^3)	S.D. (ng/m^3)
Date	4/28/93	4/28/93	4/28/93	Spring-marine average (all yrs)	Spring-marine std. dev. (all yrs)
Fine mass	6419	5142	5605	2206	1610
PM 10 mass (fine + coarse)	13241	8730	11069	4655	2992
Non-seasalt sulfate	1712	1770	1019	304	254
Al	205	28	273	37	44
Ca	123	15	153	25	28
Fe	120	19	126	29	40
Si	438	62	496	80	91
NO_3^-	237	97	232	72.7	63.8
Pb	2.87	1.18	1.48	0.90	0.80
Total organic	1067	1350	1297	659	544
Total elemental carbon	174	238	168	135	84
Soil	2054	291	2411	397	430

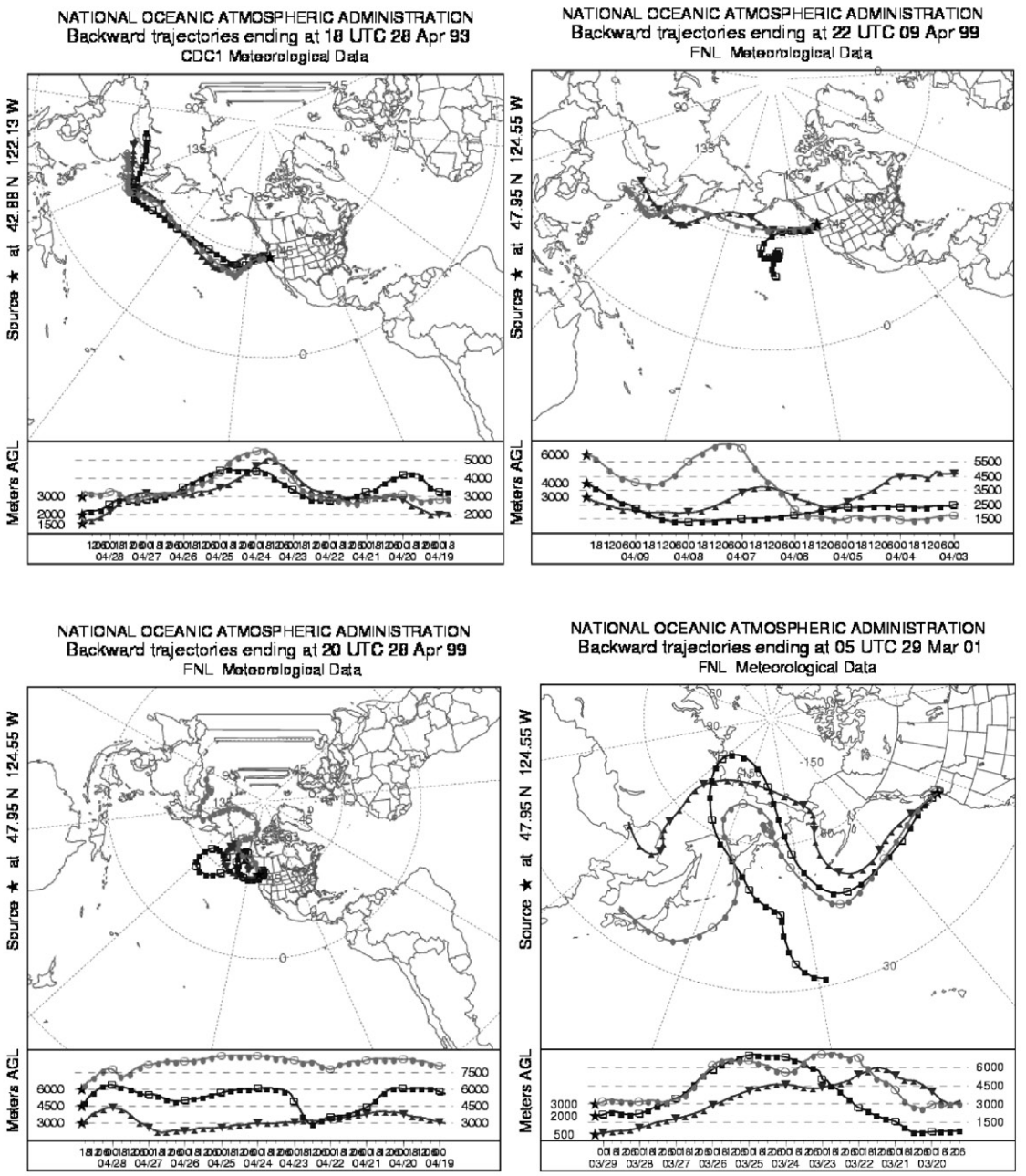

Fig. 2. Back trajectories for (a) 28 April 1993; (b) 9 April 1999; (c) 28 April 1999; (d) 29 March 2001.

was, in fact, somewhat depleted. This is similar to the results from the large dust event of April 1998, the trans-Pacific episode from March 1997 (Jaffe et al., 2001) and, as described below, similar to the large mineral aerosol event seen in April 2001. Apparently mineral aerosol either reduces ozone formation in the source region or else acts to destroy it during transport. This appears to be true even when the dust arrives together with large amounts of industrial compounds. A possible explanation for this is ozone–aerosol reactions that act to destroy ozone (Dentener et al., 1996; Phadnis and Carmichael, 2000).

It is useful to compare the April 1993 episode with the extensively studied April 1998 dust episode (Husar et al., 2001; McKendry et al., 2001). In the 1998 case a substantial dust cloud, which originated over western China was transported across the Pacific in 5–6 days in the mid-troposphere. Following its arrival to North America, the dust was brought to the surface by high-pressure subsidence and mountain wave activity (Hacker et al., 2001). Significant PM values were recorded at rural and urban sites throughout western North America (McKendry et al., 2001; Husar et al., 2001). Table 3 compares the IMPROVE particulate data from the April 1998 episode with the date from the April 1993 episode described above. The comparison shows that the April 1993 episode had a higher fraction of the total PM2.5 mass due to aerosol sulfate, nitrate, elemental carbon, and lead, compared with the April 1998 case, consistent with our attribution of industrial sources as the dominant contributor to this episode. Notwithstanding this interpretation, mineral dust still makes a 32% contribution to the PM2.5 mass for the April 1993 episode for Crater Lake.

Table 3
Crater Lake IMPROVE data for two episodes of trans-Pacific transport

	28 April 1993	29 April 1998
Predominant source	Industrial + mineral dust	Mineral dust
PM2.5 mass (ng/m^3)	6419	21,490
PM 10 mass, fine + coarse (ng/m^3)	13,241	52,991
PM2.5/PM10%	48.5	40.6
NH_4SO_4 (% total 2.5 mass)	34.2	6.8
Total organic (% total 2.5 mass)	16.6	12.6
Total elemental carbon (% total 2.5 mass)	2.7	1.0
Total soil (% total 2.5 mass)	32.0	67.2
NO_3^- (% total 2.5 mass)	3.7	1.7
Pb (% total 2.5 mass)	0.045	0.018

Table 4
Comparison of PM2.5 elemental ratios

	Al/Fe	Ca/Fe	Si/Fe
Asian dust: Beijing observations*	1.04	0.80	3.5
Asian dust: April 1998 (Crater Lake)	1.86	0.94	3.36
Crater Lake: 28 April 1993	1.71	1.03	3.65
Mt. Lassen: 28 April 1993	2.16	1.21	3.92
Marine spring data (Crater Lake)	1.28	0.85	2.76
All spring data (Crater Lake)	1.44	0.90	3.07

*Mean ratio from aerosol samples ($2\ \mu m < d < 4\ \mu m$) from Beijing during a major dust event (Braaten and Cahill, 1986).

Additional confirmation of the presence of Asian dust on 28 April, is obtained by comparing the elemental ratios observed at Crater Lake for the 28 April episode with data from the April 1998 case (McKendry et al., 2001) and with data on elemental composition mineral dust collected in Beijing (Braaten and Cahill, 1986). Table 4 shows these data. The similarity of the elemental ratios for the Asian dust samples with the Crater Lake data from April 1993 provides further evidence for the presence of Asian dust. Comparing the ratios for 28 April, with the ratios in background marine air at Crater Lake, it is clear that the presence of Asian dust on 28 April, 1993 significantly modified these elemental ratios. Keep in mind that an exact match to the Asian dust ratios is not expected since the dust may have been significantly modified during transit and the PM2.5 aerosol on 28 April 1993 contains a significant amount of industrial pollutants, as described above, which undoubtedly has a different ratio of elements.

One final point should be made about possible effects on wet deposition from the April 1993 episode. Precipitation chemistry from the Hoh River National Atmospheric Deposition Program (NADP) site in Olympic National Park (available from http://nadp.sws.uiuc.edu) during this period suggests an influence from long-range transport. Nitrate and sulfate wet deposition values for April 1993 were 0.15 kgN/ha and 0.47 kgS/ha, respectively. These values are 234% and 162% of the long-term mean for all April data in the record (1981–2001). The largest N and S wet deposition values for the Hoh site during this time, occurred during the period from 20–27 April 1993, very close to the time of the IMPROVE filter sample taken on April 28. At the same time, precipitation for the month of April 1993 was 175% of the long-term April mean. Therefore, while it is difficult to separate out the influence of greater precipitation amounts on the total deposition, it seems likely that some of this enhanced signal was due to the enhancements in long-range transport during this time period.

3.2. Episodes 2 and 3: April 1999

During the spring of 1999 14 aircraft profiles off the coast of Washington state were made for the PHOBEA field campaign. A description of the instrumentation, flight patterns, overall results, correlations and general relationship to synoptic patterns is given in Kotchenruther et al. (2001a). Analysis of the in situ ozone chemistry and relationship to PAN is given in Kotchenruther et al. (2001b). In this work we utilize the 1999 data to examine several long-range transport events.

Table 5
Mixing ratios measured from selected flight segments in April 1999

Date	Altitude (km)	CO (ppbv)	O_3* (ppbv)	PAN (pptv)	Ethane (pptv)	Propane (pptv)	σ_{sp}* (Mm^{-1})	σ_{ap}* (Mm^{-1})	Ångström exponent
9 April 1999	2.8	171	67	622	1948	494	21.0	2.6	2.4
9 April 1999	6.3	n.d.	79	302	n.d.	n.d.	6.7	1.1	2.4
12 April 1999	4.9	n.d.	84	436	n.d.	n.d.	17.4	1.9	2.5
28 April 1999	3.0	154	69.5	274	1715	309	7.8	1.2	2.3
28 April 1999	6.0	n.d.	94	312	n.d.	n.d.	9.3	1.2	2.1
Sp. 1999:	0–2.5	146,	50,	54,	1744,	343,	2.5,	0.2,	2.0
Median, Mean $\pm$ 1 s.d.		142 $\pm$ 17	49 $\pm$ 10	88 $\pm$ 89	1644 $\pm$ 407	361 $\pm$ 230	2.7 $\pm$ 2.6	0.3 $\pm$ 0.4	2.0 $\pm$ 0.4
As above	2.5–4.5	142,	59,	90,	1677,	309,	2.6,	0.5,	2.1,
		137 $\pm$ 31	60 $\pm$ 11	180 $\pm$ 188	1516 $\pm$ 527	339 $\pm$ 204	4.7 $\pm$ 5.4	0.7 $\pm$ 0.7	2.1 $\pm$ 0.3
As above	4.5–8.0	136,	72,	224,	1633,	315,	3.6,	0.5,	2.2,
		133 $\pm$ 20	68 $\pm$ 14	208 $\pm$ 103	1556 $\pm$ 475	322 $\pm$ 193	5.4 $\pm$ 5.4	0.7 $\pm$ 0.6	2.3 $\pm$ 0.3

Notes: We have excluded one stratospheric intrusion episode on 3 April 1999. If this one event is included, the mean O_3 mixing ratio at 4.5–8 km increases to 77 ppbv, with a standard deviation of 29 ppbv. Aerosol scattering coefficient in the green ($\lambda = 550$ nm) and absorption coefficients are given relative to STP. Ångström exponents are calculated from the ratio of the green and red scattering coefficients ($\lambda = 550,\ 700$ nm). It is only calculated for layers with aerosol scattering coefficients that are well above their detection limit. N.d. refers to no data.

Flights conducted on 4, 7, 9, 12, and 28 April all showed layers of enhanced gases and aerosols in air masses that appear to be of Eurasian origin. We focus on 9, 12, and 28 April which have the largest enhancements of the largest number of compounds. We assume that the data from 9 and 12 April are part of the same long-range transport event. Table 5 shows mixing ratios, aerosol scattering and absorption coefficients, as well as the Ångström exponent measured on these flights. The averages in Table 5 are for constant altitude flight legs of approximately 20 min duration. The Ångström exponent gives information on the mean aerosol size distribution of the sampled population, keeping in mind that the inlet used in 1999 most likely excluded particles greater than about 1 μm diameter.

For 9 April, the largest enhancements in PAN and aerosol scattering were seen at $\sim$2.8 km; however, significant enhancements of PAN and O_3 were also seen at $\sim$5 and 6 km elevation. At 4 km, most species were at background concentrations or below. As a gauge of the enhancement over background values, Table 5 also shows the median and mean concentrations for each species binned by altitude region, based on the data from all 14 flights. This yields 17 independent flight segments for the 3 km bin, and 11 flight segments for the 6 km bin (see Kotchenruther et al., (2001a) for more details on the flight segments and binning). Comparing the 9 April data with the background data, all values shown for the 3 and 6 km layers on 9 April are statistically different from the mean at a 95% confidence interval or better, except O_3 in the 6 km layer for 9 April, and ethane and propane in the 3 km layer. For ethane and propane, this partly reflects the relatively small number of canister samples that we collected for all flights in 1999.

Back trajectories for 9 April 1999 are shown in Fig. 2b and indicate that the air sampled at 3 and 6 km originated from close to the boundary layer near to the Korea–NE China region 6–8 days prior to our sampling date. The 4-km trajectory arrived from more southerly regions, near Hawaii, where it was probably caught up in slow moving cyclonic flow around the Pacific high. Thus, the NOAA-Hysplit back trajectory for 9 April based on the FNL data set, seems to give good agreement with the chemical observations for that day.

The question arises as to why the 3 km region on 9 April shows a greater enhancement then the 6 km region, even though the trajectories originated from approximately the same source region. For example, aerosol scattering in the 3 km layer is approximately 2 times the value at 6 km. Although slightly different transport paths, and therefore dilution, are certainly part of the answer, precipitation may also play a role. The trajectory arriving at 6 km, showed substantial lifting as it left the East Asian region near 1.5 km elevation. The Hysplit trajectory shows that for the air parcel arriving at 6 km the cumulative rainfall was 7.3 mm along its path, compared with 1.3 mm for the trajectory arriving at 3 km. As a result, the reduced aerosol scattering coefficients we observed at 6 km likely result from enhanced scavenging and washout of aerosols en-route. Note that as the values given in

Table 5 are relative to STP, it is appropriate to compare these directly.

On 12 April, elevated mixing ratios of nearly all species were seen between 2 and 6 km, with the highest values near 5 km (see Table 5). Very likely the episodes observed on 9 and 12 April 1999 are connected to the long-range transport episode observed during the PEM-Tropics mission. Clarke et al. (2001) describe a region of significantly enhanced aerosols during the final flight of the PEM-Tropics mission, which took place between Hawaii and California on 9 April 1999. Global model calculations for that date indicated that the air mass originated from Asia and that the largest enhancements were north of the PEM-Tropics flight track. Based on the back trajectories for 12 April (not shown) and the substantially elevated gases and aerosols we observed, it is likely that the air we sampled on 12 April came from the region of largest enhancements shown in Plate 6 of Clarke et al. (2001) near 40°N 140°W.

For 28 April 1999, CO, O_3, PAN and aerosols were all statistically enhanced to varying degrees in the air masses arriving at 3 and 6 km. Enhancements in many compounds were also seen at 4–5 km, as well, though not as large. The trajectories for 3.0, 4.5 and 6.0 km arrival heights, shown in Fig. 2c, do not suggest an obvious source region. It is important to note that while the trajectories do cross over western British Columbia prior to reaching our sampling location, the aircraft profiles in the lower troposphere and boundary layer show no evidence of local pollution. For example, short lived tracers, such as NO_x and ethene, are not enhanced in any part of the 28 April profile, thus making local influences unlikely.

Since the 28 April back trajectories does not point to Asian industrial emissions, two other sources can be considered: biomass burning emissions from Siberia and industrial emissions from Europe. Biomass burning in Siberia is common from late spring through fall period. Substantial burning in Siberia occurred between April and August 1998 (Kasischke et al., 1999; Tanimoto et al., 2000; Kajii et al., 2002), and this is believed to have impacted mixing ratios of a number of species in the northeastern Pacific (Jaffe et al., 2001). However, there is no evidence that biomass burning in this region was particularly strong during April 1999 (Duncan et al., 2002). Our examination of the measured hydrocarbons, the aerosol absorption coefficient and the Ångström exponent does not indicate an influence from biomass burning on this date. Thus, we are not able to unambiguously identify a source for the 28 April 1999 episode.

On 28 April, the O_3 mixing ratio at 6 km was 94 ppbv, well above background. Given that the back-trajectories arriving at 6 km came from higher in the troposphere (near 8 km), it is likely that the O_3 mixing ratio reflects some enhancement from the upper troposphere or lower stratosphere. But the presence of significant enhancements in PAN, aerosol scatter and absorption at 6 km indicates that some portion of this O_3 is due to photochemical production from industrial precursors.

3.3. Cases 4, 5 and 6: March–April 2001

Twelve aircraft profiles were also conducted from 29 March–6 May 2001 in the same region as the 1999 profiles. Price et al. (2002) give a description of the instrumentation, flight patterns and overall results from the 2001 campaign. The 2001 flights included in situ measurements of O_3, aerosol scatter at three wavelengths, and canister samples for analysis of CO and NMHCs. Significant gas and aerosol layers were observed during flights on 29 March, 14 April and 6 May. The largest aerosol scattering values were observed on 14 April, resulting from several major dust storms in Asia (Gong et al., 2002; Thulasiraman et al., 2002). Associated with this mineral dust were high mixing ratios of CO and NMHCs, indicating that this air mass also picked up industrial pollutants as it left Asia. Table 6 provides an overview of data from these three flights, along with background data from the spring 2001 observations.

The episode of 29 March 2001, appears to be similar to the events identified in 1999. Aerosol scatter, CO, O_3 and NMHCs are all enhanced. Trajectories for this date at arrival heights of 0.5–3 km indicate industrial sources in East Asia (Fig. 2d). However, the fact that the trajectories arrive via higher latitudes, suggests that other sources may also contribute.

The 14 April episode originated as a major dust outbreak in China during the first week of April. By 5 April, the dust could be seen in the AI derived from measurements by the TOMS instrument onboard the Earth Probe satellite. On 8 April, an air mass with significantly enhanced AI was located in the northern Sea of Japan, as seen in the TOMS images (Fig. 3). Over the next 6 days, this air mass was transported across the Pacific and could be seen in the TOMS AI off the coast of the western US on 14 April. Vertical profile data from 14 April 2001 indicate the presence of an air mass with substantially enhanced mineral aerosol between 4 and 6 km. This is evident from the very large aerosol scattering values, the largest observed on any flights in 1999 or 2001, and the low value for the Ångström exponent. The later is consistent with an aerosol size distribution that has a maximum greater than 1 μm aerodynamic diameter (Price et al., 2002). Thus, despite the fact that the aerosol inlet for the aircraft likely excludes particles greater then 1 μm, the 14 April Ångström exponent indicates that the size distribution has its maximum particles with an diameter of more than 1 μm. Given the size cut of the aircraft inlet, it is likely that we measured only a small fraction of the total

Table 6
Mixing ratios measured from selected flight segments from March–April 2001

Date/time (gmt)	Altitude (km)	CO (ppbv)	O_3* (ppbv)	Ethane (pptv)	Propane (pptv)	σ_{sp}* (Mm^{-1})	Ångström exponent
29 March 2001	2.0	167	73	1478	419	20	n.d.
14 April 2001	6.0	173	48	1558	436	40	0.8
6 May 2001	3.0	153	63	1319	252	15	2.2
Sp. 2001 Median,	0–2.5	153,	42,	1478,	346,	2.2,	2.5,
Mean ± 1 s.d.		148 ± 17	44 ± 9	1479 ± 354	399 ± 184	3.6 ± 4.6	2.3 ± 0.4
As above	2.5–4.5	138,	47,	1317,	309,	2.4,	2.2,
		137 ± 19	44 ± 9	1296 ± 326	323 ± 164	3.7 ± 4.1	2.1 ± 0.4
As above	4.5–6.5	131,	45,	1196,	245,	2.4,	2.4,
		135 ± 22	45 ± 10	1200 ± 329	260 ± 132	5.4 ± 9.5	2.2 ± 0.7

Notes: Aerosol scattering coefficient in the green ($\lambda = 550$ nm) and absorption coefficients are given relative to STP. Ångström exponents are calculated from the ratio of the green and red scattering coefficients ($\lambda = 550,\ 700$ nm). It is only calculated for layers with aerosol scattering coefficients that are well above their detection limit. N.d. refers to no data.

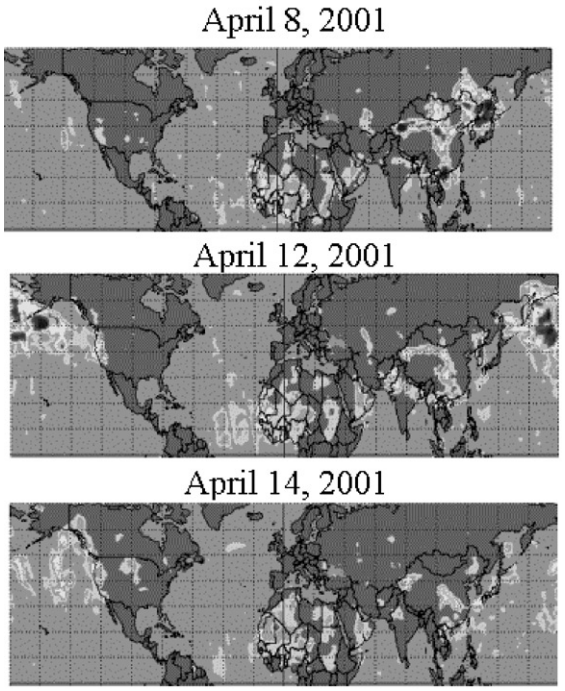

Fig. 3. TOMS Aerosol Index for 8, 12 and 14 April 2001.

aerosol scattering on this date. For most other flights, the Ångström exponents indicates a size distribution which has a maximum in the sub-micron size range, and thus our measured aerosol scattering probably includes a large fraction of the total aerosol scattering (Price et al., 2002).

On 14 April, CO and NMHCs were also significantly enhanced in the 4–6 km region. In fact CO on the 14 April flight was the highest value measured on all flights in 2001. Trajectories from this date (not shown), originated in East Asia, consistent with the TOMS AI mentioned above. The trajectory arriving at 6 km, passes over both the industrial region (Korea, north-eastern China) as well as Mongolia within 8 days. A global aerosol model of the April 2001 dust events suggests that the aerosol we measured on 14 April 2001 originated from the Gobi desert (Gong et al., 2002). We surmise that the gaseous pollutants were picked up by this dust-laden air mass as it crossed over the industrial areas in China and the Koreas. This air mass was then transported across the Pacific in the mid-troposphere.

Synoptic conditions over Asia during early April 2001 show a strong similarity to the pattern seen in the April 1998 dust event, the most significant feature of which is a deep low located at 115°–125°E and 50°N. The presence of a low pressure system near 50°N, 125°E on 8 April 2001, 0 GMT suggests that the passage of a cold front was able to lift material from the lower troposphere up into the middle troposphere, where it could be quickly transported across the Pacific. The position, intensity and frontal activity of such lows enable severe dust storms to develop over the interior Eurasian deserts (Husar et al., 2001). These events are also characterized by strong zonal flow at 500 hPa.

Ozone during the 14 April period has rather different behavior then the other compounds. For most periods when sub-micron aerosol scattering and CO are elevated, O_3 is also elevated. However, for 14 April, this is not the case. While the presence of CO indicates a contribution from urban and industrial emissions, the lack of an O_3 enhancement indicates that either elevated O_3 was not present in the Asian boundary layer at that time or that the O_3 was destroyed enroute.

Surface data from CPO did not show a significant enhancement on 14 April 2001, consistent with the aircraft data from that day. However, on 16 April,

significant peaks in aerosol mass, both PM10 and PM2.5, were observed at CPO and in the Seattle urban air monitoring network. Simultaneously, data from several IMPROVE sites also showed significant aerosol enhancements on 16 April. This includes observations from the sites near the Mt. Rainier and North Cascades National Parks in Washington, the Crater Lake and Mt. Hood sites in Oregon, and the Mt. Lassen site in Northern California. PM10 values at the IMPROVE sites ranged from 17 $\mu g/m^3$, at the sites in Washington, to 35 $\mu g/m^3$ at Crater Lake. These samples contain significant amounts of mineral dust, as indicated by the high coarse mass fraction and the elevated Si/Fe ratio of 3.1–3.7, similar to the values reported in Table 4. The wide distribution of this dust, the chemical similarity to the previous observations of Asian dust and the TOMS AI images all point to an Asian source. However, because of greater complexities in boundary layer transport, the exact mechanism by which this Asian dust was transported to the surface is less certain and remains under investigation.

Data from 6 May 2001 are shown in Table 6. Observations at 3 km show a significant enhancement in O_3 and aerosol scattering and a slight enhancement in CO. Observations at 2 km show very similar enhancements in $O_3$and aerosol scattering; however, no CO data is available for this altitude. The trajectories from 2 and 3 km (not shown) suggest pollution from Asia, similar to other events that we have identified.

4. Discussion and summary

Table 7 summarizes the previously reported data on trans-Pacific transport, along with the six new cases identified in this paper. The 1998 and one of the 2001 events contained large amounts of mineral dust, whereas several of the other events were dominated by industrial pollutants or contained mixed signals. The common features of these trans-Pacific transport events seems to be frontal lifting of boundary layer emissions in the source region, followed by fairly rapid transport (Jaffe et al., 1999; Husar et al., 2001; Thulasiraman et al., 2002). None of the previously reported episodes identified long-range transport of O_3, although several of the new episodes contain substantial O_3 enhancements.

The question arises as to why the 29 March 1997, 28 April 1993 and 14 April 2001 episodes did not have enhanced O_3, even though each had significant amounts of other industrial pollutants, including O_3 precursors.

Table 7
Summary of documented trans-Pacific episodes

Dates	Receptor site	Enhanced compounds	References
Summer 1983, 1984	Free troposphere	Rn	Kritz et al., 1990
24 April–5 May 1985	Pt. Arena, Calif.	NMHCs, PAN	Parrish et al., 1992
3–12 May 1985	Free troposphere	Rn, SO_2, SO_4^{2-}	Andreae et al., 1988
28 April 1993	Crater Lake, Mt. Rainier, Mt. Lassen	S, mineral dust, metals, organics, etc.	This work
29 March 1997	CPO	CO, O_3, NMHCs, PAN, Rn, aerosols	Jaffe et al., 1999
Late April–early May 1998	CPO, Crater Lake, and many other locations in the US	Mineral dust, S, etc.	Husar et al., 2001 McKendry et al., 2001
9,12 April 1999	Free troposphere	CO, O_3, NMHCs, PAN, Rn, aerosols	This work
28 April 1999	Free troposphere	CO, O_3, NMHCs, PAN, Rn, aerosols	This work
29 March 2001	Free troposphere	CO, O_3, NMHCs, aerosols	This work
14 April 2001	Free troposphere	CO, NMHCs, aerosols (dominated by super-micron aerosols)	Thulasiraman et al., 2002; and this work
6 May 2001	Free troposphere	CO, O_3, NMHCs, aerosols	This work

Two common elements of trans-Pacific episodes with an ozone enhancement are transport in the free troposphere and absence of mineral aerosol in the transported air masses. The presence of large amounts of mineral dust may reduce O_3 in one of two ways. First, it could prevent O_3 from forming in the source region due to reductions in UV reaching the surface (Dickerson et al., 1997). Second, mineral aerosol is known to facilitate several aerosol–ozone reactions that destroy O_3, effectively reducing the O_3 lifetime (Dentener et al., 1996; Phadnis and Carmichael, 2000). Based on our data, it is not possible to ascertain if elevated O_3 was present in the original air mass or whether it was lost during transport. Either way, it is apparent that the absence of mineral dust seems to be necessary to get significant O_3 enhancements during long-range transport.

In addition, the limited observations described herein suggest that marine boundary layer (MBL) transport also does not give rise to O_3 enhancement. This is based on the observations from 29 March 1997 (Jaffe et al., 1999) and those from the 28 April 1993 episode. Generally, MBL transport takes longer than transport in the free troposphere. This suggests that the O_3 lifetime in the MBL is shorter then the transport time and/or that something in the MBL is destroying O_3 en-route. Both OH and halogen radicals could play a role in this chemistry (Dickerson et al., 1999; Platt, 2000; Toyota et al., 2001).

In summary, recent observations from the west coast of North America suggest that episodic trans-Pacific transport of gases and aerosols is relatively common, particularly during spring. In this paper, we described a number of "newly discovered" episodes of this phenomenon. Of particular note is the diversity of episode types in terms of the combinations of pollutants that arrive. Industrial sources, mineral dust and possibly biomass burning emissions can all contribute in varying amounts to the concentration of gases and aerosols arriving into western North America in any particular episode. Furthermore, it is apparent that varying meteorological and emission conditions in the source regions, the myriad transport pathways across the Pacific and weather patterns in the receptor regions all contribute to the diversity of episodes we have identified.

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