

Surface ozone profiles at selected South Pacific sites

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Abstract

Surface ozone profiles were studied at Fiji (18.1°S, 178.2°E), Samoa (14.3°S, 170.6°W), Tahiti (18.0°S, 149.0°W), and San Cristobal, Galapagos (0.9°S, 89.6°W) from 1997-2003. Surface ozone levels at these Pacific sites showed a wintertime maximum and a summertime minimum with the lowest mixing ratios occurring at Tahiti and Galapagos, while the highest mixing ratios occurred at Fiji. The seasonal trends at each site were found to be dependent on the varying photochemical sink strength and the changing transport regimes in each season. It was established that the reduced photochemical sink strength in winter was one of the major reasons for the observed increase in surface ozone levels at each site in that season. The presence of the South Pacific Convergence Zone particularly for the Western Pacific (Fiji, Samoa and Tahiti) is the other factor contributing to the observed seasonality and variability in surface ozone levels. The changing frequency of transport from the southern mid-latitude (from high altitudes) is the only transport regime at the boundary layer having the most significant influence on the surface ozone levels at these Pacific sites.

Keywords: Ground-level ozone, South Pacific, Trends

1. Introduction

The atmospheric distribution of ozone (O₃) in the atmosphere is unique, with the largest concentrations occurring in the lower stratosphere, as the protective ozone layer. Ozone also occurs at the boundary layer and is an eye and bronchial irritant, which can damage living tissues of plants (Handley and Grulk 2008; Mittal *et al.* 2007; Paoletti, 2006; Sicard *et al.*, 2011, 2013) and animals, and is a major component of the summer-time smog which is prevalent in heavily industrialized cities such as Los Angeles. The Environmental Protection Agency (EPA) under the United States Clean Air Act of 1970 has promulgated an 80 ppbv ozone standard to protect the public of such cities in the United States (EPA 2004).

Ground-level ozone is an important atmospheric pollutant and climate forcer. Ozone, the third important greenhouse gas in terms of radiative forcing (Ramaswamy *et al.*, 2001), is an important air quality issue. The lifetime of tropospheric O₃ varies from one or a few days in the boundary layer to a few tens of days or even a few months in the free troposphere which enables transport from regional to hemispheric scale and hence proportionally has greater influence on climate than O₃ near the surface.

The annual cycle and trends of surface ozone in Europe have also been widely studied (Monks *et al.*, 2003; Vingarzan, 2004; Jonson *et al.*, 2006; Derwent *et al.*, 2007; Wilson *et al.*, 2012; Parrish *et al.*, 2012; Sicard *et al.*, 2009, 2013). In addition, the same authors have also studied the tropospheric ozone at northern and western parts of Europe.

While the South Pacific basin is often considered

pristine, being free of pollutants from heavily industrialized continents, indications are that pollution levels have an upward trend with large contributions from long-range transport (Schultz *et al.* 1999; Oltmans *et al.* 2001). The boundary layer ozone in particular has been seen to exhibit seasonal trends at the South Pacific locations such as Fiji, Samoa and Tahiti (Thompson *et al.* 2003; Oltmans *et al.* 2001). The major source of surface ozone is through photochemical oxidation of precursor molecules such as CH₄, CO and non-methane hydrocarbons (NMHC) in high nitrogen oxide (NO_x = NO + NO₂) environments (Liu *et al.* 1992). These precursor molecules enter the atmosphere from motor vehicle exhaust fumes, power plants, home furnaces, commercial fuel combustion, industrial facilities and biomass burning (Lelieveld and Crutzen 1994). Besides the chemical route, another important source of ozone is through the exchange of air between the stratosphere and troposphere (Sicard *et al.*, 2009). Ozone and its precursor molecules have been found to occur in the South Pacific region through long-range transport from heavily industrialized or biomass burning continents (Gregory *et al.* 1999; Oltmans *et al.* 2001; Thompson *et al.* 2003). The major sink for surface ozone is through photolytic destruction pathways, and catalytic destruction cycles involving oxidation of CH₄ or CO in low NO_x environments (Graedel and Crutzen 1993).

The main objective of this study was to present the annual profiles of surface ozone at the four Pacific sites and provide some possible explanations for these.

2. Instrumentation and Methodology

The data discussed for Fiji (18.1°S, 178.2°E), were from 1997 to 2003. Data from other sites, Samoa (14.3°E, 170.6°W), were from 1997 to 2003, Tahiti (18.0°S, 149.0°W) from 1997 to 1999 and Galapagos (0.9°S, 89.6°W) from 1998 to 2003, were provided by SHADOZ.

Vertical ozone profiles were taken on a weekly basis at Fiji from 1997 till the end of 2003 using an Electrochemical Concentration Cell (ECC) ozonesonde.

The ECC ozonesonde as described by Komhyr *et al.* (1995) uses platinum electrode as the ozone sensor. The ECC contains KI solutions of varying strength which undergo reaction 1.1 with ozone in ambient air. Air is pumped into the ECC by a battery operated non-reactive Teflon pump.



The output current is proportional to the rate at which ozone enters the sensor. Hence, if the flow rate and cell current (typically 0-6 microamperes) are known then the amount of ozone in partial pressures can be determined.

Using raw vertical ozone profile data from launches carried out in Fiji and from raw data available at the NASA/NOAA ftp site for the three other sites under study, surface ozone levels for all sites were calculated from 1997 to 2003. These were simply the ozone mixing ratios (in ppmv) at the surface before the launch is made. The type of equipment, procedures and sensing solutions, mostly for the Pacific sites were similar hence ozone data and trends were readily comparable.

3. Results and Discussion

Surface ozone levels showed a clear annual cycle with similar seasonal variations from 1997 to 2003 (Figure 1a to d).

The annual cycle for surface ozone shown for each site clearly portrays a summer time minimum and a winter time maximum. The summer minimum generally occurs from January through to March while the winter maximum generally appears from June to August. Apart from the seasonal variations, it is also evident from the plots that surface mixing ratios at each site vary from year to year for certain sites with slight variations between sites as well.

Surface ozone mixing ratios for Fiji (Fig. 1a) show a similar trend for each year from 1997 to 2003. The average maximum mixing ratios are also a little higher (20-32 ppbv) than other Pacific sites (Samoa 16-27 ppbv; Tahiti 18-25 ppbv; and Galapagos 17-23 ppbv) with mixing ratios surpassing 30 ppbv a number of times.

Maximum mixing ratios are seen to occur from June

to August which is the cooler period, while the minimum mixing ratios occur during the warmer months of December to March.

Maximum mixing ratios for Samoa (Fig. 1b) begin in June and last till September before decreasing in October. In fact after a high in June the average mixing ratios drop by about 10 ppbv during July and/or August, with mixing ratios reaching another maximum by September. The minimum mixing ratios occur from December to April. Relatively low mixing ratios are also seen throughout the year for 1999 and 2002. The mixing ratios for Samoa are below 25 ppbv most of the time.

The data for Tahiti (Fig. 1c) also follow a similar pattern for each of the three years with a relative minimum occurring between the maximum in June and/or August/September. This is also evident in the mixing ratios for some years in Fiji.

The average mixing ratios for Galapagos (Fig. 1d) show a clear minimum from January through to May and a clear maximum, which persist from July till September. An exception to this is the high average mixing ratios seen during January and February of 1998.

Tahiti and Galapagos have the lowest average mixing ratios for the four sites with mixing ratios remaining below 20 ppbv most of the time. Fiji shows higher averages for the maximum period while Tahiti and Galapagos have the lowest averages for the minimum period.

Similar trends have been previously reported by Ayers *et al.* (1992), Harris and Oltmans (1997), Olson *et al.* (1996) and Crawford *et al.* (2001). Studies by Ayers *et al.* (1992), and Harris and Oltmans (1997) have shown surface ozone seasonal trends in remote regions to be largely dependent on in-situ photochemical processes and transport processes.

3.1 Photochemical Considerations

Being pristine and remotely located, the South Pacific basin has a relatively low NO_x concentration. During the Pacific Exploratory Mission in the tropics (PEM Tropics A and B) conducted by NASA in 1997, it was found that NO_x concentrations were only 4-3 ppbv from 0° to 30° South near the surface (Schulz *et al.* 1999; Wei *et al.* 2003). Therefore, this low NO_x concentration in the South Pacific leads to the catalytic destruction of ozone near the surface (Graedel and Crutzen 1993). Since the NO_x concentrations are low in all seasons, the observed surface ozone trend does not appear to depend on this catalytic destruction mechanism.

The South Pacific basin with its high humidity and sunlight is also an effective sink for surface ozone through photolytic destruction mechanisms (Graedel and Crutzen 1993). A correlation study between

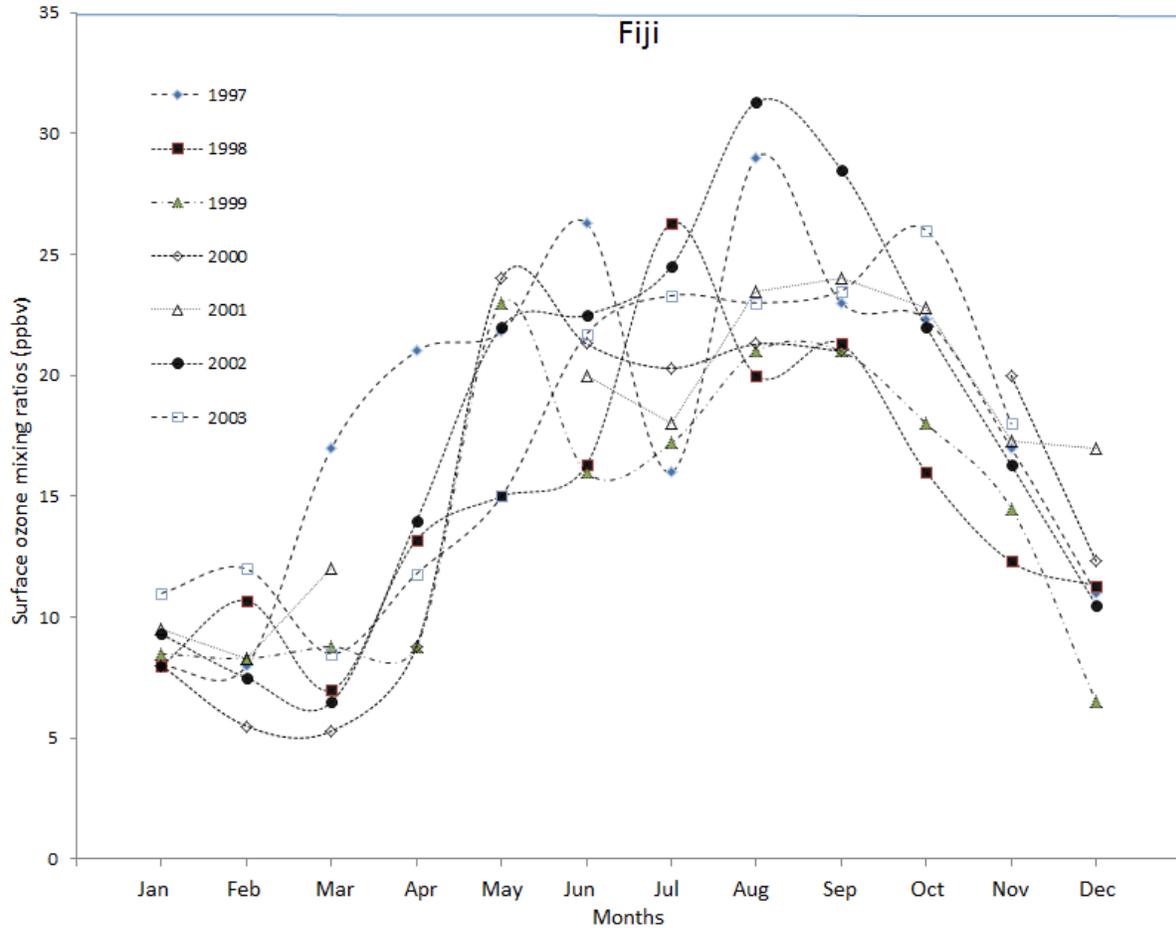


Figure 1(a). Monthly ozone volume mixing ratio from 1997 to 2003 for Fiji.

sunshine hours at Suva, Fiji and surface ozone showed an anti-correlation between the two (Chandra 2004). It was seen that the highest sunshine hours occurred during the summer months of December to February and lowest sunshine hours during the winter months of June to August. Thus, it appears that the winter maximum in surface ozone at Fiji is due to the decreased sink strength during the winter months while the summer minimum is due to the restoration of the sink strength during the summer months. Since other sites under study (Tahiti, Samoa and Galapagos) also occur in the Southern Tropics, it is expected that the sunshine hours at these sites will show maximum during summer and minimum during winter. Thus, the increased photochemical sink strength during summer and decreased photochemical sink strength during winter is a dominant (but not the only) factor determining the seasonality in surface ozone at each of the four tropical sites. Similar reasoning has also been given by Ayers *et al.* (1992) and Harris and Oltmans (1997) to explain the seasonal trend of surface ozone in remote locations. The amount of atmospheric water vapour is also a determining factor in the photolytic

destruction of ozone. Since the sites are tropically located, it is highly unlikely that water vapour could be present in limiting amounts in either season. Harris and Oltmans (1997) did not find any correlation between surface ozone and water vapour for Samoa while Olson *et al.* (1996) did find a good anti-correlation for Ascension Island. Hence, the variation in water vapour could affect this photochemically driven destructive cycle in certain seasons for some years. Since the amount of water vapour and singlet oxygen [$O(^1D)$] determine the local hydroxyl radical concentration, it is likely that this could have seasonal influence from the catalytic ozone destruction cycle where the first step (oxidation) involves hydroxyl radicals. The role of bromine in the destruction of surface ozone has also been studied by other researchers (Sturges *et al.* 1993).

Surface ozone precursors (NO_x , CO , CH_4 and NMHCs) from local sources are expected to be insignificant. Since all tropical sites under study are developing nations, industrial releases of these species are relatively small. In addition, since all are island nations, rapid mixing quickly reduces the concentration of precursors from any local sources.

While the changing sink strength is undoubtedly a dominant factor determining the surface ozone trends, it is however not sufficient to explain the increased variability in surface ozone amounts during the winter months. Moreover, the correlation between sunshine hours and surface ozone (though strong) was not entirely one to one. Hence, there appears to be another factor (probably transport processes), which could be having an added influence on the surface ozone trend at Fiji. The amount of cloud cover, solar zenith angle and total column ozone are also sources of variability in surface ozone amounts, but effects from these factors are expected to be small (Olson *et al.* 1996).

3.2 Transport Processes

The South Pacific is marked by two major low-level convergence zones, the Inter Tropical Convergence Zone (ITCZ) and the South Pacific Convergence Zone (SPCZ), which provide major barriers for atmospheric transport mostly below 5 km altitude (Hoell *et al.* 1999). Studies by Halter *et al.* (1988), Harris and Oltmans (1997) and research during the PEM-Tropics

A and B missions (Gregory *et al.* 1999) revealed that the sites within this region are affected by the flow from three distinct wind regimes. It was found that sites north of the ITCZ are affected by the flow from the Northern Hemisphere with air masses showing urban/industrialized chemical signatures. Sites south of the SPCZ were found to be affected by migrating cyclones bringing air from the mid-latitudes. This wind flow regime showed chemical signatures dominated by biomass burning emissions. A study by Ziemke *et al.* (2009) on biomass burning in the tropics showed a significant impact in increasing the tropospheric ozone levels by 10-25%. Gregory *et al.* (1999) noted that the resulting chemical gradients across each zone was more pronounced below 5 km which is consistent with the strong low-level convergence that is characteristic of each zone, becoming much less pronounced at higher altitudes.

Sites that fall south of the ITCZ and north of the SPCZ, were found to be influenced by the south-easterly trades bringing in clean air of marine origin.

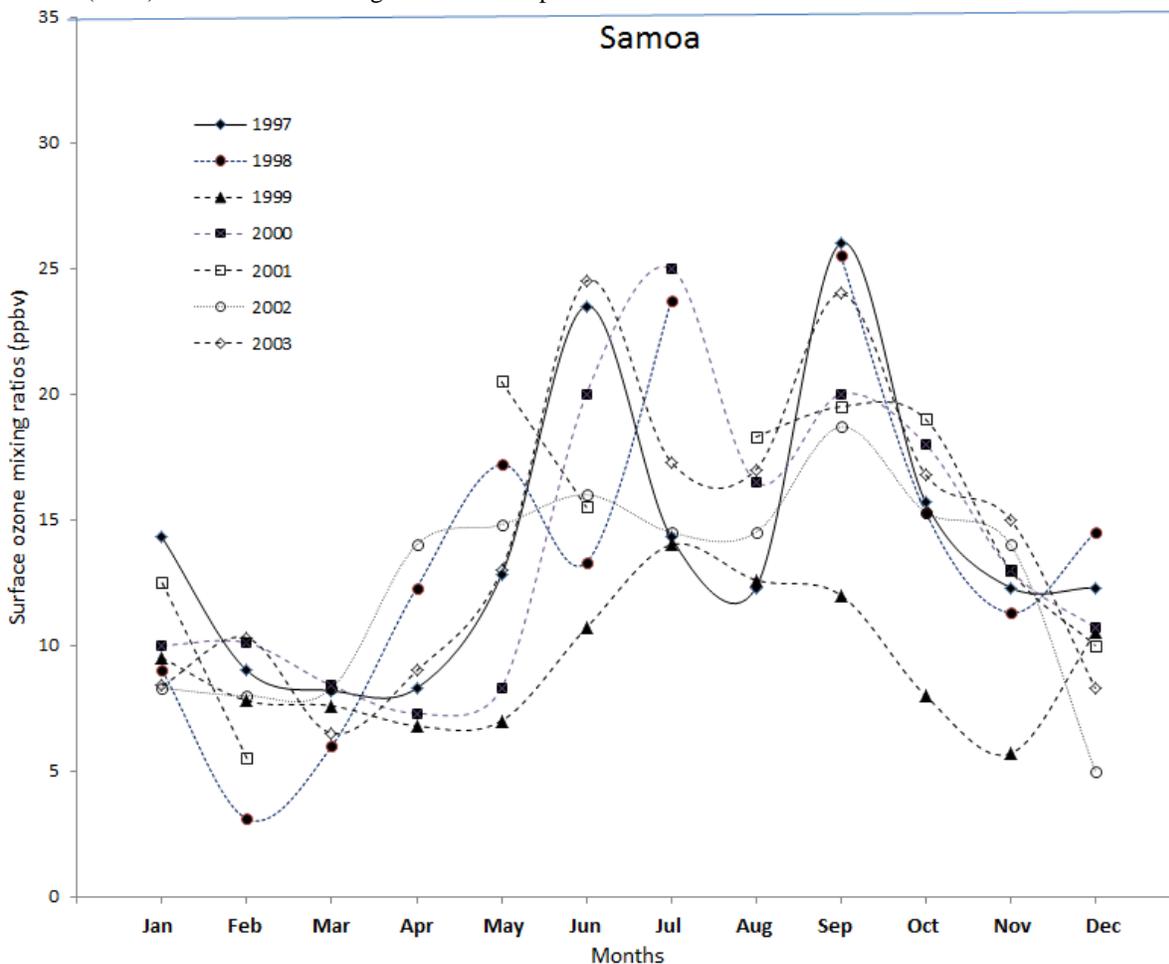


Figure 1(b). Monthly ozone volume mixing ratio from 1997 to 2003 for Samoa.

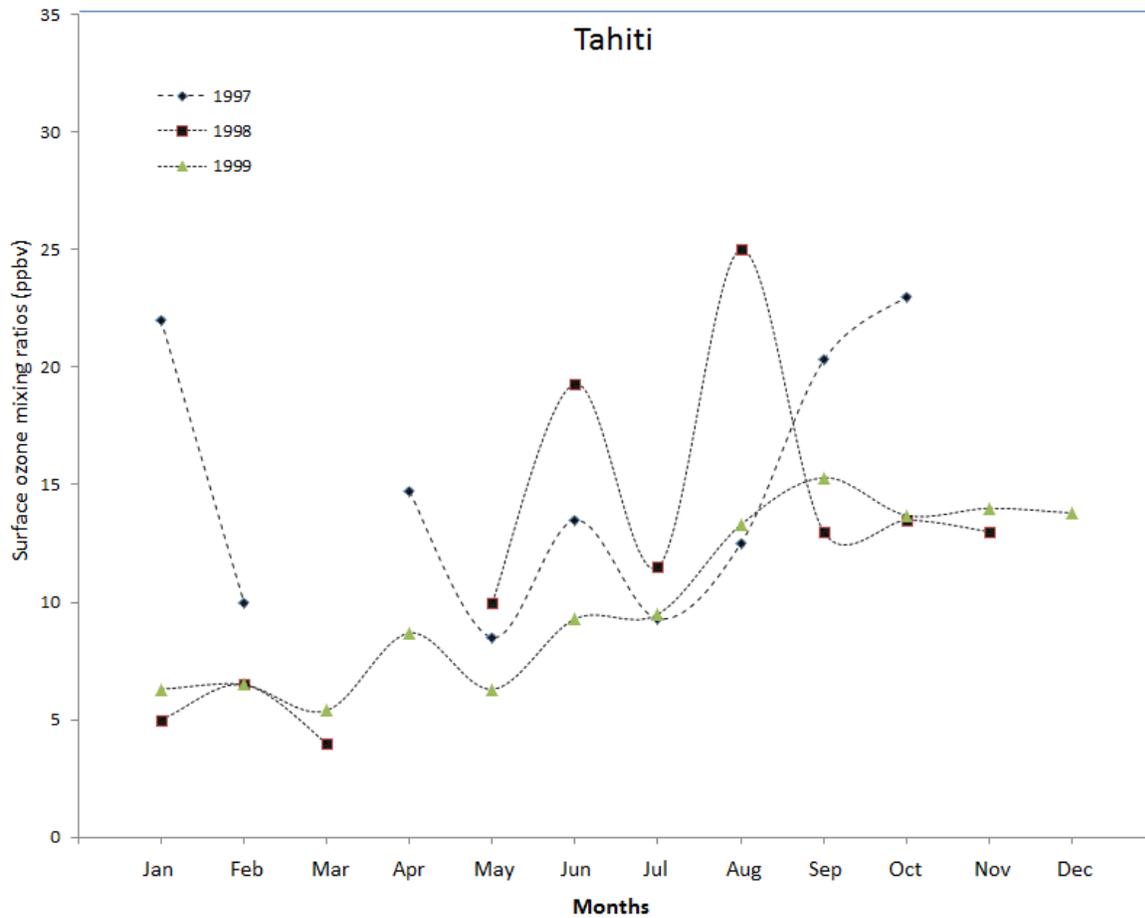


Figure 1(c). Monthly ozone volume mixing ratio from 1997 to 1999 for Tahiti.

Seasonal shifts in these two convergence zones cause some sites to oscillate between the three transport regimes described above. Studies by Chandra (2004), revealed that unlike other South Pacific Islands such as Samoa and Tahiti, Fiji from 1997 to 2003 was mostly located south of the SPCZ, hence was most frequently exposed to wind flow from biomass burning regions. This is reflective of the relatively high surface ozone evident in Fiji. Furthermore, Harris and Oltmans (1997) found that sites within the South Pacific, experience south-easterly trades 80% of the time but in winter time, this flow occurs only 50% of the time. Since the south-easterly trades are relatively clean and are exposed to ozone sinks for a longer period of time, which is greater in summer, it contributes much less towards the surface ozone levels during summer. This further supports the observation of the low level of surface ozone seen during the summer months in Fiji. Variability during the summer months is much less and this could be attributed to the homogeneity of transport from the southeast Pacific. Similarly it was found by

Chandra (2004) that the SPCZ was more frequently located south of Samoa and Tahiti during this study period, hence reflective of the surface ozone levels seen at these sites.

Surface ozone at Galapagos has low variability in both seasons, since the flow to Galapagos is not affected by the SPCZ position. Thus, a tradewind-type flow could be the dominant transport regime affecting Galapagos in each season. Galapagos is also expected to be influenced continuously by continental outflows due to its proximity to the American continent. However, surface ozone levels seen at Galapagos do not indicate such prominent pollution source. Flow to Galapagos at 1 km (near to the boundary layer) is mostly from the south and surface ozone levels reflect the uniformity of this flow regime.

It is evident that the observed seasonal trends in surface ozone for the four tropical sites are due both to varying ozone sink strength and changing wind flow patterns in different seasons. There is also a possibility that the observed increase in ozone levels during the

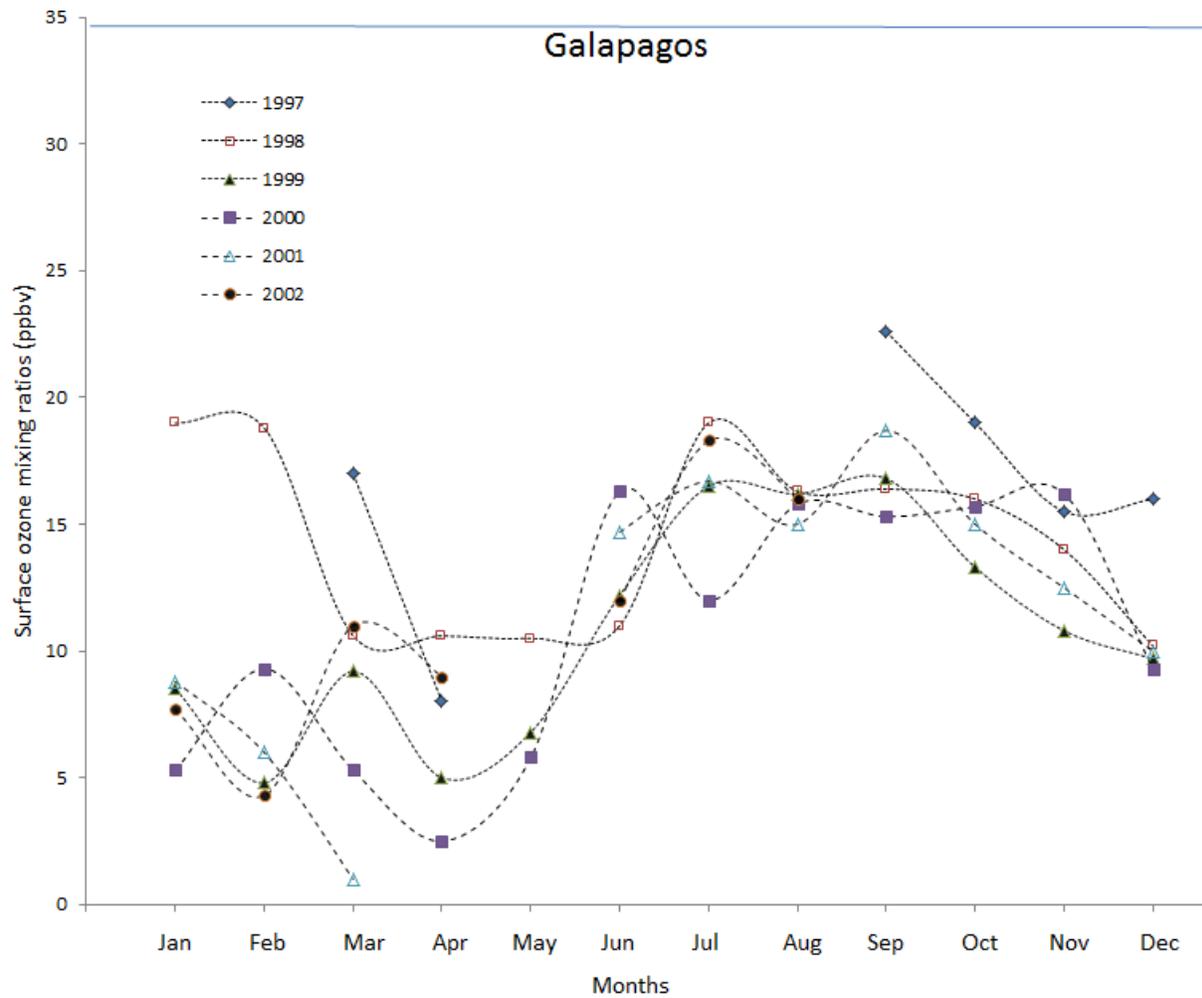


Figure 1(d). Monthly ozone volume mixing ratio from 1998 to 2003 for Galapagos.

winter/spring season (Figure 1) is due to the increased source strength in these seasons. The most significant source of ozone and ozone precursors in the South Pacific is from biomass burning (Oltmans *et al.* 2001). It was revealed during the PEM-Tropics mission that ozone production within the South Pacific's lower troposphere (below 4 km) is dominated by NO_x resulting largely from the decomposition of peroxyacetylnitrate (PAN) transported into the region with biomass burning pollution at high altitudes (Schultz *et al.* 1999). However the strength of this source cannot be determined with certainty to confidently confirm the observed trends in surface ozone. The surface ozone levels at all the study sites do not pose any serious health risk, as they are well below the EPA standard of 80 ppbv in 8 hours.

Some studies have indicated that intercontinental transport appears to be an important factor that may explain observed ozone trends (Derwent *et al.*, 2007;

Sicard, 2009). Climate change, local meteorological conditions and stratospheric-tropospheric ozone exchange are other additional factors that may be contributing to the varying surface ozone mixing ratios.

4. Conclusion

Surface ozone levels at the four Pacific sites have shown a similar seasonal cycle with winter time maximum and a summer time minimum. The levels at the study sites were seen to range from nearly 0 to 32 ppbv with an overall average for the study period (1997-2003) ranging from 12 ppbv to 27 ppbv. It was seen that Tahiti and Galapagos showed the lowest average mixing ratios of the four sites while Fiji had the highest.

The ozone levels seen at the study sites do not pose any serious health risk as they are below the EPA standard of 80 ppbv per 8 hours. The observed seasonality and variability in surface ozone levels were

generally found to be dependent on the varying photochemical sink strength and the changing transport regimes in different seasons.

An anti-correlation between surface ozone and total sunshine hours was found for Suva, Fiji. This indicated that the reduced sink strength during the winter period resulted in the observed ozone maximum during the period.

The presence of the SPCZ in the Western Pacific is seen to be a barrier in transport from the predominant Southeast Pacific (tradewinds) and transport from the southern mid-latitude to the Western Pacific sites (Fiji, Samoa and Tahiti). The tradewinds-type flow is considered to be relatively clean, originating from within the Pacific while flow from the southern mid-latitudes has an upper tropospheric and stratospheric source with possible contributions from biomass burning.

Fiji is more exposed (south of SPCZ) to flow from the southern mid-latitude while Samoa and Tahiti experience only occasional exposures. Therefore, on average Fiji has the highest surface ozone level while Galapagos appeared to be the least affected by any varying transport regimes or continental outflow from the American continent at the surface.

Climate change, local meteorological conditions and stratospheric-tropospheric ozone exchange are other additional factors that could be considered in future in terms of their effect on ozone levels at the surface and the troposphere.

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