# Seasonal changes in stable carbon isotopic composition of n-alkanes in the marine aerosols from the western North Pacific: implications for the source and atmospheric transport.

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**Abstract**
In order to constrain seasonal changes in the long-range atmospheric transport of land derived lipid biomarker compounds (LDLBs) over the open ocean, marine aerosol samples were collected on a biweekly basis from 1990 to 1993 at a remote island, Chichi-Jima (27°04'N, 142°13'E) in the western North Pacific. We report results of the compound specific stable carbon isotope analysis of the *n*-alkanes isolated from the marine aerosols fraction using gas chromatography/isotope ratio/mass spectrometry (GC-ir-MS). The isotopic data reveal seasonal variations in vegetation source, atmospheric pathways and transport processes. Concentrations of *n*-alkanes ranged from 0.1 to 14.1 $ngm^{-3}$, with concentrations highest in the winter. Compound-specific isotope analysis revealed, in particular, strong seasonal changes in the $\delta^{13}C$ values of the $C_{29}$ and $C_{31}$ *n*-alkanes (biomarkers for higher plants). Lighter $\delta^{13}C$ values were observed in winter (typically -32 to -34‰) – with a transition to heavier values in summer (typically -28 to -31‰). Using a mixing equation and typical end members for $C_3$ and $C_4$ plants, we found that this is due to relative increases in the contributions from $C_4$ plants in the summer season. However, in a few samples significant biasing of the $\delta^{13}C$ signal by anthropogenic fossil fuel contributions was also inferred. Using backward air mass trajectory analyses, we found that the Asian continent was the major source region for $C_3$ plant material during winter/spring, whereas Indonesia/Australia and possibly the Americas are the source regions for $C_4$ material during the summer/autumn. We also found that enhanced atmospheric transport of *n*-alkanes from $C_4$ plants occurred in 1991 summer/autumn during a strong El Nino event, which was associated with forest and bush fires in Indonesia and Australia. In addition to providing information on contemporary processes, this study also provides a base for future paleoclimatological work in ocean sediments.

# 1 INTRODUCTION

Aeolian dust fallout to the open ocean was first described in the Atlantic by Darwin (Darwin, 1846) and the importance of atmospheric transport of terrestrial materials to deep-sea sediments – including the Pacific basin - has long been a major issue of oceanography (Blank et al., 1985; Rex and Goldberg, 1958; Schramm and Leinen, 1987). The study of the fate of organic aerosols has been given further impetus recently, due to a significant role in climate forcing; directly through radiative forcing effects (e.g. Chung and Seinfeld, 2002; e.g. Jayaraman, 1999; Penner et al., 1992) and indirectly, by altering the microphysical properties of clouds (e.g. Novakov and Penner, 1993; Yu, 2000). A prominent contemporary example of the importance of organic aerosols is given by the phenomenon known as the Atmospheric Brown Cloud – a coalescent cloud of anthropogenic pollutants and natural wind-bourn dust - that is having a measurable impact on Asian climate (UNEP and $C^4$, 2002).

Analytical advances during the last few decades have made possible studies describing in detail the molecular and isotopic composition of the organic component of aeolian materials. A result of this work has been the demonstration that homologous series of high molecular weight (HMW) *n*-alkanes, *n*-alcohols, and fatty acids are typical land derived lipid biomarkers* (LDLBs) found in atmospheric dust (Conte and Weber, 2002; Fang et al., 2002; Gagosian et al., 1987; Kawamura, 1995; Kawamura et al., 2003; Matsumoto et al., 2001; Sicre and Peltzer, 2004), marine sediments (Gagosian and Peltzer, 1986; Huang et al., 2000; Ikehara et al., 2000; Ohkouchi et al., 1997a; Ohkouchi et al., 1997b; Ohkouchi et al., 2000; Schefuss et al., 2003a; Schefuss et al., 2003b; Westerhausen et al., 1993; Zhao et al., 2003) and lacustrine sediments (Huang et al., 2001; Huang et al., 1999; Street-Perrott et al., 1997; Zhang et al., 2004).

These lipids are synthesised by terrestrial higher plants as constituents of the epicuticular waxes (Eglinton et al., 1962; Eglinton and Hamilton, 1967). Particulate matter containing such compounds may be deflated to the troposphere either directly, by being sloughed off plant surfaces by winds (especially by sandblasting), or alternately they may be eroded from the soil reservoir during dust storms (Simoneit et al., 1977; Simoneit and Eglinton, 1977). Therefore, plant-wax lipids form a significant component of the "aerosol veil", even above remote ocean areas such as the North Pacific (Gagosian and Peltzer, 1986; Gagosian et al., 1981; Kawamura, 1995; Kawamura et al., 2003), South Pacific (Gagosian et al., 1987; Sicre and Peltzer, 2004) and Atlantic (e.g. Simoneit et al., 1991; Simoneit et al., 1977).

The refractory nature of some LDLB compounds (e.g. HMW *n*-alkanes, *n*-alcohols and *n*-monocarboxylic acids) (e.g. Cranwell, 1981; Kawamura, 1995) means that information contained within them (e.g. isotopic composition of the source material) may be preserved in modern and geological samples without suffering major modification. Studies of contemporary LDLB samples may elucidate a number of

* The term "biomarker" refers to an organic compound with an unambiguous link to a known natural product. In this case the biomarkers are compounds emanating from the epicuticular waxes of terrestrial higher plants.

modern earth processes, such as long-range atmospheric transport, circulation (Kawamura, 1995; Ohkouchi et al., 1997a) and deposition of organic carbon (OC) (Gagosian and Peltzer, 1986; Kawamura, 1995; Prahl and Muehlhausen, 1989) (including organic pollutants (Simoneit, 1997; Simoneit, 2002)) as well as the relative contributions to aerosol OC from $C_3$ and $C_4$ plant sources (Chesselet et al., 1981; Conte and Weber, 2002; Fang et al., 2002; Ohkouchi et al., 1997a; Prahl and Muehlhausen, 1989; Schefuss et al., 2003a). Moreover, in addition to providing information on processes of contemporary importance, such studies also provide a base for palaeoclimatological work (Bird et al., 1995; Ohkouchi et al., 1997b; Prahl and Muehlhausen, 1989; Schefuss et al., 2003b; Westerhausen et al., 1993).

*n*-Alkanes typically have carbon chain-lengths ranging from $C_{25}$ to $C_{35}$ and a strong predominance of odd-carbon-number over even-carbon-number homologues ($C_{27}$, $C_{29}$, $C_{33}$ are most common) (Eglinton et al., 1962; Eglinton and Hamilton, 1967; Kolattukudy, 1976). High molecular *n*-alkanes are more resistant to degradation than shorter-chain homologues and other n-alkyl components (order of stability: *n*-alkanes>n-alkanoic acids>n-alkanols) (Cranwell, 1981). The carbon isotopic analysis of tissues from plants using the different pathways of photosynthetic carbon fixation, $C_3$, $C_4$, and CAM, show different levels of $^{13}C$ depletion (Smith and Epstein, 1971). This carbon isotopic heterogeneity between plants types is also observed within specific compound classes, including *n*-alkanes (Chikaraishi and Naraoka, 2003; Collister et al., 1994). Thus compound-specific carbon isotope (CSIA) analyses of leaf-wax lipids can determine the plant types from which the lipids were derived. $C_3$ plants have *n*-alkane $\delta^{13}C$ values around –35‰ (-30 to –40‰ vs. Pee Dee belemnite), whereas $C_4$ plants have $\delta^{13}C$ values around –20‰ (-17 to –24‰) (Chikaraishi and Naraoka, 2003; Collister et al., 1994). The third type, CAM plants, can use both carbon fixation pathways, so their $\delta^{13}C$ values are intermediate (Chikaraishi and Naraoka, 2003; O'Leary, 1981; Rieley et al., 1993). Virtually all trees, most shrubs, and cool-season grasses and sedges use the $C_3$ pathway, whereas $C_4$ photosynthesis is found in warm-season grasses, sedges and maize. CAM plants form a much smaller proportion of the global biomass and include many succulents, such as cacti (Cerling and Quade, 1993; Spicer, 1989). The stable carbon isotope compositions of *n*-alkanes have been used to determine relative contributions by $C_3/C_4$ plant material in marine sediments (Bird et al., 1995; Huang et al., 2000; Kuypers et al., 1999; Schefuss et al., 2003b) and aerosol dust in the eastern Atlantic (Schefuss et al., 2003a). To date, no CSIA analysis of *n*-alkanes in dust in the western Pacific has been conducted.

Here we present data on the seasonal variations of stable carbon isotope compositions of *n*-alkanes from aerosols collected from Chichi-Jima in the western North Pacific (see Figure 1). We compare the variations in carbon isotope compositions of the *n*-alkanes collected at Chichi Jima with air mass back trajectory analysis and a new quantitative map of global $C_3/C_4$ abundance (Still et al., 2003) (Figure 1). The data provides insight into the higher plant and anthropogenic sources and transport pathways of terrigenous lipid materials in the continental dust carried out to the ocean. Moreover, in addition to providing information on contemporary processes, this study also provides a base for future paleoclimatological work in ocean sediments. CSIA of *n*-alkanes in sediments can help resolve palaeo-wind patterns and deposition of OC over the Pacific. Alternately, if wind patterns can be constrained (e.g. by modelling) sedimentary *n*-alkanes could reveal changes in the evolution of past $C_3/C_4$ plant abundances and hence the carbon isotopic discrimination of

terrestrial photosynthesis - an important parameter for accurate estimates of paleo-$\rho CO_2$ (Conte and Weber, 2002).

## 2 EXPERIMENTAL SECTION

### 2.1 Regional Setting and Sampling

Chichi-Jima is a small island (~8 × 5 km in size) located at 27°04'N; 142°13'E, approximately 2000 km west of the Asian continent and around 1000 km south of Tokyo (Figure 1). The region is strongly influenced by the Asian monsoon, with wind systems characterized by Westerlies during the winter to spring seasons, whereas Trade Winds dominate in summer to autumn. Figure 1 shows variations of wind direction at 700 hPa over Chichi-Jima Island over the 4-year sampling period basis from April 1990 to November 1993. During this period 69 aeolian dust samples were collected on a biweekly basis at the Satellite Tracking Center of National Aeronautic and Science Develop Agency (NASDA, elevation: 254 m). The climate of Chichi-Jima is sub-tropical with an annual rainfall of 850mm. The biomass is dominated by $C_3$ plants (>99% Hara, pers. comm.). There are a few species of $C_4$ plant, mostly sedges which are found in streams near the coast (Toyoda, 1981; Hara, pers. comm.) . It is highly unlikely that there would be a significant $C_4$ plant seasonal influence on the aerosols collected at the elevated sampling station.

### 2.2 Extraction, derivertization and analysis of n-alkanes.

Aerosol samples were collected on a pre-combusted (500 °C, 3 h) Pallflex quartz fiber filter (20 × 25 cm) using a high volume air sampler (1 $m^3$ $min^{-1}$). Detailed procedures were described in Kawamura *et al.* (2003). The sampling time was approximately 100 h to collect enough material for chemical analysis. After the sample collection, the quartz filter was placed in a pre-cleaned glass jar with a Teflon-lined screw cap and stored in a freezer at -20 °C until analysis. An aliquot of the filters (10 – 20%) was extracted under reflux for 2 h with 100 mL of 0.1 M KOH in methanol, containing ca. 5% distilled water. The analytical method was described in Kawamura *et al.* (2003). Briefly, the extracts were isolated by filtration with a pre-cleaned Whatmann GF/A filter. The residue was further extracted with methanol and then methylene chloride under ultrasonification for 3 min. The extracts were combined and concentrated under vacuum and then separated into neutral and acidic fractions using the methods of Kawamura (1995). The neutral fraction was further separated into four sub-fractions using a silica gel column chromatography, that is, aliphatic hydrocarbons/*n*-alkanes (N1), aromatic hydrocarbons (N2), aldehydes and ketones (N3) and alcohols (N4) by elution with *n*-hexane, *n*-hexane/methylene chloride (2:1) mixture, methylene chloride and methylene chloride/methanol (95:5) mixture, respectively. Sub-fractionation of the acid classes is detailed in Kawamura et al. (2003) and Fang et al. (2002).

The *n*-alkanes were analyzed on a Carlo Erba Mega 5160 gas chromatograph (GC) equipped with a cool on-column injector, fused silica capillary column (Chrompack, CP-Sil 60m × 0.32mm i.d. × 0.25 μm) with an FID detector. The oven program was 70°C to 120°C at 30°C/min and to 300°C (22 min) at 5°C/min. Structural identification was confirmed using a GC/MS (Thermoquest, Voyager) with similar GC column conditions. Authentic $n$-$C_{29}$ alkane was used for quantification, recoveries were better than 80%. Analytical errors of the procedures were within 15%.

Laboratory procedural blanks showed that blank levels were less than 1% of the samples.

### 2.3 Stable Carbon Isotope Analysis.

Isotopic ratios of *n*-alkanes were determined using an HP 6890 gas chromatograph coupled with a Finnigan MAT Delta Plus isotope ratio mass spectrometer by a Finnigan MAT combustion furnace containing Cu and Ni wires which were doped with oxygen and maintained at a temperature of 850 °C. The GC was equipped with a cool on-column injector and a HP-5MS fused silica capillary column (30m × 0.32mm i.d., 0.25µm film thickness). The GC oven was programmed from 50 to 120 °C at 30 °C/min, from 120 to 300 °C at 5 °C/min and maintained at 300 °C for 22 min. The flow rate for He carrier gas was set at 1.6 mL/min. Two microliters of sample solution spiked with $C_{20}$ methyleicosanoate ($C_{20}$ FAME) as an internal standard of known isotopic composition were injected through an on-column injector. Peaks were simultaneously detected in Faraday collectors at *m/z* 44 ($^{12}C^{16}O_2$), 45 ($^{13}C^{16}O_2$ and $^{12}C^{17}O^{16}O$), and 46 ($^{12}C^{16}O^{18}O$, $^{12}C^{17}O^{17}O$, and $^{13}C^{17}O^{16}O$), amplified, corrected for the presence of $^{17}O$ at mass 45 using the Craig correction (Craig, 1957). The isotopic composition of *n*-alkanes is reported in the delta notation relative to the Pee Dee Belemnite (PDB) standard as follows:

$$\delta^{13}C\ (‰) = [(^{13}C/\ ^{12}C)_{sample}/\ (^{13}C/^{12}C)_{PDB} - 1] \times 10^3 \quad \textbf{(1)}$$

Isotope values reported were determined by averaging duplicate analyses whenever concentrations were sufficient. A minority of the samples (30%) contained only enough material for a single isotopic analysis of the dominant $C_{29}$ and $C_{31}$ homologues. For the duration of the analysis the condition of the machine was checked by injection of an external standard of *n*-alkanes (mixture of various carbon numbers from $C_{15}$ to $C_{33}$) of known isotopic composition. This gave a reproducibility of ~0.5‰ for all compounds (e.g. $\delta^{13}C$ 0.45‰, 0.5‰ and 0.57‰ for $C_{27}$, $C_{29}$ and $C_{31}$ respectively). The isotopic composition of sample *n*-alkanes was calculated by co-injection with a non-coeluting internal standard ($C_{20}$ FAME) of known isotopic composition ($\delta^{13}C$ -26.21‰).

### 2.4 Back-Trajectory Analysis.

Back-trajectory analyses were conducted using the Fifth-Generation NCAR/Penn State mesoscale model (MM5).The trajectory calculation was based on backward tracking of a selected air parcel released at a target location, assuming the parcel was moving along the ambient airflow. The flow pattern was updated every 6 h.

## 3 RESULTS AND DISCUSSION

### 3.1 Distribution of n-alkanes in aerosols

The concentrations and molecular distributions of the *n*-alkanes (0.17–14 $ngm^{-3}$, average 1.7 $ngm^{-3}$) collected at Chichi-Jima have been reported previously, with other lipid classes (Kawamura et al., 2003). The seasonal data is summarized in Table 1. The *n*-alkanes were characterized by a strong odd-carbon number predominance (CPI ratios, average 4.5) with a maximum at $C_{29}$ or $C_{31}$, indicating that terrestrial higher plant waxes were the primary source. Figure 3 illustrates a typical *n*-alkane class distribution, as seen in most of the samples. Absolute concentrations generally increased in winter/spring and decreased toward summer/autumn season, except for

the El Niño year of 1991 when the concentrations were relatively high in the summer/autumn (Kawamura at al., 2003). Such a seasonal trend suggested that atmospheric transport of terrestrial organic materials over the western North Pacific are enhanced during winter/spring season probably due to the strong westerly winds (see Figure 2). CPI values were generally high suggesting a minimal contribution from petroleum residues/pollution (Kawamura et al., 2003). Unresolved complex mixtures of hydrocarbons (UCM) were detected in some samples. UCM hydrocarbons are emitted to the atmosphere by the incomplete combustion of fossil fuels (e.g Boyer and Laitinen, 1975; Simoneit and Mazurek, 1981) and are typically abundant in aerosol and rainwater samples from the urban atmosphere (e.g. Kawamura and Kaplan, 1991; Kawamura et al., 1995; Simoneit, 1984; Simoneit et al., 1988). The presence of UCMs may demonstrate the influence of polluted air from Asian countries at Chichi-Jima. However, the concentrations were 2 or 3 orders of magnitude lower than for urban samples, thus confirming (as with the high CPI values) the relatively low levels of pollution at the remote Chichi-Jima site. There were no clear seasonal trends of the CPI ratios or UCM concentrations (Kawamura et al., 2003).

### 3.2 Stable isotopic compositions of *n*-alkanes.

Results of the stable isotopic measurements for the major *n*-alkanes are given in Table 2. The stable isotopic compositions of the dominant HMW *n*-alkanes ($C_{29}$, $C_{31}$) fell in the range –26.6 to –34.4‰ indicating that all the samples consisted primarily of varying relative amounts of $C_3$ and $C_4$ plant-derived leaf-wax lipids (Table 2). In general, for the dominant $C_{29}$ and $C_{31}$ *n*-alkanes, lighter $\delta^{13}C$ values were observed in winter (typically -31 to -34‰) with a transition to heavier values in summer (typically -28 to -33‰) (Figure 4 a,b). Within individual samples the odd numbered *n*-alkanes are relatively more depleted in $^{13}C$ with increasing chain length, a typical isotopic distribution is illustrated in Figure 3. In general the $C_{29}$ and $C_{31}$ alkanes were the most $^{13}C$ depleted compounds in each sample. The longer-chain odd-numbered compounds, $C_{33}$ and $C_{35}$, if measurable, were isotopically enriched relative to $C_{29}$ and $C_{31}$.

Fang *et al* (2002) previously reported the compound specific $\delta^{13}C$ values measured in the fatty acid compound class extracted from the same Chichi-Jima sample set. The isotopic values measured by Fang *et al* (2002) for the HMW fatty acids were also consistent with a higher plant source of varying relative amounts of $C_3$ and $C_4$ lipids (-30‰ to -28.2‰ with a mean of -29.1‰ ± 0.7‰). They suggested that heavier $^{13}C$ values (-26.9‰ to -24.8‰, with a mean of -25.8‰) measured in the LMW fatty acids ($C_{12}$ -$C_{19}$) may have indicated a contribution from marine biota (Fang et al., 2002), as marine organic carbon in the 40-50°N latitude range has a $\delta^{13}C$ of around -21‰ (Chesselet et al., 1981; Fontugne and Duplessy, 1978). However, the *n*-alkane compound class data does not suggest a significant marine algal input. *n*-Alkanes synthesised by aquatic algae typically have $C_{15}$ or $C_{17}$ as the dominant homologue (Blumer et al., 1971; Gelpi et al., 1970), but these compounds were not present as aerosol particles in concentrations measurable by GC-FID or GC-ir-MS.

As well as marine derived *n*-alkanes a significant contribution of fossil fuel *n*-alkanes to the aerosols collected at Chichi-Jima is also a possibility and may bias the $\delta^{13}C$ signal derived from higher-plant waxes. There is no consensus on the most

appropriate method to assess and/or correct for non-higher plant contributions. To estimate whether a potential fossil fuel/marine contribution of *n*-alkanes influenced our samples we applied a mass balance approach adapted from the method of Ishiwatari *et al.,* (1994):

$$\delta^{13}C_{nw} = \frac{(\delta^{13}C_{nx} \times A_x) - \delta^{13}C_n}{A_x - 1} \quad \textbf{(2)}$$

where $\delta^{13}C_{nw}$ is the higher plant wax $\delta^{13}C$ fraction and $\delta^{13}C_{nx}$ is the fossil fuel/marine $\delta^{13}C$ fraction of an *n*-alkane $\delta^{13}C_n$ with carbon number *n* ($C_n$) and $A_x$ is the fraction of the fossil fuel/marine *n*-alkanes contributing to the absolute abundance of $C_n$. For values of $\delta^{13}C_{nx,}$ we use the $\delta^{13}C$ value of the $C_{24}$ *n*-alkane in each sample, assuming that it is equal to the $\delta^{13}C$ value of the fossil fuel contribution. The $C_{24}$ *n*-alkane is often absent in pure plant wax samples (Chikaraishi and Naraoka, 2003; Collister et al., 1994; Eglinton et al., 1962; Eglinton and Hamilton, 1967), but occurs (in small quantities) in many of the samples from Chichi-Jima. Therefore, we assume that the $C_{24}$ *n*-alkane orginates from fossil fuel/marine sources. To calculate $A_x$ we used the ratio $C_{24}/C_{29}$ *n*-alkane as a measure of the fraction contributed by fossil fuel/marine *n*-alkanes. Some previous authors have used CPI ratios to calculate $A_x$ (Ishiwatari et al., 1994; Schefuss et al., 2003a). However, we are not satisfied that an uncontaminated/higher plant wax end member can be assigned to the mixing equation used for this method – given that natural CPI values vary so greatly (e.g. 1.4 to 40.3 in Chikaraishi and Naraoka, 2003; Collister et al., 1994).

The corrected $\delta^{13}C$ values for the $C_{29}$ and $C_{31}$ *n*-alkanes are given in Table 2 and illustrated in Figure 4c,d with the uncorrected values for comparison. Corrections could be made for only the samples where the $C_{24}$ *n*-alkane was abundant enough for a successful compound-specific $\delta^{13}C$ measurement. This was possible for 55% of the samples for which compound-specific $\delta^{13}C$ measurements had also been made for the $C_{29}$ and $C_{31}$ *n*-alkanes. For two thirds of these samples the differences in the uncorrected and corrected $\delta^{13}C$ values were insignificant (< 0.5‰), for one third of the samples the corrections were >0.5‰ and for five samples (QFF- 194, -214, -217, -223, -420) the corrections were >1‰. The potential source of the non-higher plant contributions to these samples is considered in more detail in section 3.5.

Figure 4 illustrates that the data-set which includes the corrected $\delta^{13}C$ values does not greatly affect the discernable seasonal trend in isotopic values (which become heavier in the summer/autumn seasons). This suggests that the seasonal trend must contain a highly significant natural signal that is a function of changes in the proportions of delivered $C_3$ and $C_4$ plant material. This is confirmed by the fact that measurements of UCMs and the CPI values at Chichi-Jima (which are sometimes used to infer the contribution from fossil fuel sources) did not have a similar, clear seasonal trend (Kawamura et al., 2003). For consistency within the dataset we use the uncorrected values in section 3.2.1 to assess $C_3$ and $C_4$ plant contributions and to compare with the modeled backward trajectories. This is because the corrections could not be applied to all of the samples (due to variable availability of $C_{24}$).

Another way to access both the potential biasing of the natural $C_3$ / $C_4$ plant signal by fossil fuel/marine inputs and whether the uncorrected or partially corrected *n*-alkane data set is preferable is to compare the *n*-alkane isotopic data with that

previously obtained from fatty acids. Fatty acids are less susceptible to non-higher plant contributions that *n*-alkanes as their concentrations are typically one order of magnitude higher in higher plant waxes. Figure 5 illustrates the linear correlation between the carbon isotopic compositions ($\delta^{13}C$) of the $C_{29}$ *n*-alkane (this paper) and the $C_{24}$ fatty acid (Fang *et al*, 2000). Linear correlations of higher significance existed between the $C_{24}$ fatty acid and the uncorrected $C_{29}$ ($R^2$ of 0.47) and $C_{31}$ ($R^2$ of 0.35) *n*-alkane isotopic data, than for the data-sets which included the corrected values ($R^2$ of 0.39 and 0.19 for the $C_{29}$ and $C_{31}$ respectively). The greater consistency of the uncorrected *n*-alkanes with the fatty acid supports the use of the uncorrected data-set.

### 3.3 Backward trajectories and sources of $C_4$ plant-derived wax lipids in aerosols

In general lighter $\delta^{13}C$ values were observed in winter (typically -31 to -34‰) with a transition to heavier values in summer (typically -28 to -33‰) (Figure 4). If the $\delta^{13}C$ values are averaged by season this trend is even clearer for the dominant $C_{29}$ and $C_{31}$ *n*-alkanes (Figure 6). The difference in the $\delta^{13}C$ of the same *n*-alkanes between the winter and summer seasons may reflect a seasonal variability in the source areas and/or and atmospheric transport pathways and hence differential source areas. The isotopic composition of plants can change in response to seasonal environmental stimuli. Lighter values of $\delta^{13}C$ for the $C_{29}$ and $C_{31}$ *n*-alkanes in several deciduous tree species have been observed in autumn leaves compared with leaves sampled at the start of the growing season ($\Delta\ \delta\ ^{13}C$ -0.6 to -5‰) (Chikaraishi et al., 2004; Lockheart et al., 1997). Spring lipids may be isotopically heavier due to the remobilization of stored carbohydrate at the beginning of the growing season (Lockheart et al., 1997). However, such phenomena can not explain the observed seasonal signal at Chichi-Jima, which is the opposite in sign (*n*-alkanes become isotopically heavier from the spring to autumn). We expect that, given the strong seasonality recorded in the prevailing winds at Chichi-Jima (Figure 2), changes in transport pathways dominate the signal at Chichi-Jima. This suggests that, every year, for a particular season the *n*-alkanes may be derived from relatively consistent sources and the aerosols are transported by similar atmospheric transport pathways and processes. In order to explore this in more detail, we estimated the source contributions of $C_3$ and $C_4$ plant material (derived from the *n*-alkane $\delta^{13}C$) and the transport pathways (by back-trajectory analysis).

Assuming that there is no significant isotopic fractionation during aerosol transport over the ocean the relative contributions to the *n*-alkanes from terrestrial $C_3$ and $C_4$ plants can be estimated by using a two-component mixing equation (Chesselet et al., 1981):

$$C_{alk} = C_3 + C_4 \quad \textbf{(3)}$$

$$C_{alk}\ \delta^{13}C_{alk} = \delta^{13}C_3 \times C_3 + \delta C_4 \times C_4 \quad \textbf{(4)}$$

Where $C_{alk}$ is the total concentration of an *n*-alkane, $C_3$ and $C_4$ are *n*-alkane concentrations from $C_3$ and $C_4$ plants, and $\delta^{13}C_3$ and $\delta^{13}C_4$ are the isotopic composition of the *n*-alkanes from $C_3$ and $C_4$ plant sources respectively. The end-member values for the mixing equation were averages taken from the literature and from measurements made at ILTS (see Table 3): for $C_{29,}$ $C_4$ plant *n*-alkanes = 20.2‰

and $C_3$ plant *n*-alkanes = -35.1‰; for $C_{31}$, $C_4$ plants *n*-alkanes = 20.4‰ and $C_3$ plant *n*-alkanes = -35.5‰ (Chikaraishi and Naraoka, 2003; Collister et al., 1994). This results in estimates of $C_4$ plant contribution varying from 6 to 37% for the individual winter samples and 7 to 59% for the individual summer samples (Table 2 and Figure 4, right axis).

Fang *et al.* (2002) estimated $C_4$ plant contributions to the Chichi-Jima aerosols from the isotopic composition of the $C_{28}$ fatty acid. They detected a similar seasonal trend of relatively higher inputs of $C_4$ material in the summer than the winter seasons (Table 4). However the absolute estimates of $C_4$ contributions are consistently lower when calculated from the $C_{28}$ fatty acid $\delta^{13}C$ (%$C_4$ 6 to 27) (Fang et al., 2002) than from the *n*-alkanes (%$C_4$ of 15 to 35%) (this paper). This discrepancy may result from differential seasonal metabolism of *n*-fatty-acids and *n*-alkanes. It may also be due the use of inaccurate end-member numbers for either or both the fatty acid and *n*-alkanes and perhaps highlights that caution is needed interpreting absolute source input contributions calculated from end-member models.

The relative contributions from the $C_3$ and $C_4$ plant sources are determined by the regional meteorological conditions and $C_3/C_4$ production and deflation to the atmosphere in the source areas. Comparison of composite back-trajectory analysis maps for each season (Figure 7) with a global map of the fraction of $C_4$ biomass (Still et al., 2003) (Figure 1) reveals the potential source areas of eroded plant material that are responsible for the seasonal changes in the $C_{29}$ and $C_{31}$ *n*-alkane $\delta^{13}C$ values observed at Chichi-Jima. In general, in winter/spring, more air masses originated in north Asia where for large regions $C_4$ plants comprise 0% of the mean annual biomass (Figure 1) (Still et al., 2003). In contrast in summer/fall, backward trajectories show a greater diversity of source areas: less air masses originated in north Asia, instead they were more often marine (easterly trades) Southeast Asian, Indonesian and sometimes (1991 and 1992) Australian in origin. In the latter regions $C_4$ plants form a more significant proportion of the annual mean biomass than in northern and western Asia, for example: up to 50% in eastern Chinese provinces such as Hebei and Shanxi, up to 70% in parts of Thailand and Cambodia and up to 100% in much of northern Australia (Figure 1) (Still et al., 2003).

### 3.4 $C_3$ and $C_4$ plant contribution and air mass source categories

To further confirm that changes in air-mass source areas control the seasonal $\delta^{13}C$ n-alkane signal at Chichi-Jima each sample was allocated an air mass source category, based on the dominant source area observed in the 10-day back trajectory. The categories were: Northeast Asia (NEA), Southeast Asia (SEA), Indonesia and Australia (IA) and Marine easterlies (ME) (Figure 1, Table 2). 96% percent of winter samples fall into the SEA or NEA source area categories, in contrast only 21% of the summer samples were of NEA or SEA origin, being of IA (30%) and ME (49%) origin respectively (Table 2). Box plots of the $C_{29}$ and $C_{31}$ $\delta^{13}C$ data for each of these major source areas are illustrated in Figure 8. The box plots show that samples in the NEA and SEA categories were significantly isotopically lighter (median values of around -32 to -33‰) than those of the ME and IA categories (median values of around -30 to -31‰). This confirms that the isotopically heavier values recorded at Chichi-Jima during the summer-autumn are a function of material carried by the IA

and ME air mass sources. The interquartile ranges (25 – 75% of the data) also demonstrate that - on the basis of CSIA measurements of the *n*-alkanes - it is largely possible to discriminate between samples with an Asian continental source (NEA or SEA) and the easterly (ME) or southerly (IA) sources. However, using this method further categorisation was not possible – there was a major overlap of the interquartile ranges of the NEA and SEA categories and of the IA and ME categories (Figure 8). The lack of significant difference between SEA and NEA categories and between IA and ME categories may be due to the entrainment and mixing by air masses of *n*-alkanes from wide geographic areas. It seems that the $\delta^{13}C$ of the aerosol *n*-alkanes at Chichi-Jima are only sensitive to the major seasonal changes in the prevailing wind systems. This suggests that paleoclimatic reconstructions using sedimentary material recovered from the Chichi-Jima sector of the western Pacific may record $C_3/C_4$ plant abundance changes over wide geographical areas.

In order to explore the potential for various combinations of higher plant biomarker parameters to further discriminate between the different source areas we looked at a matrix of scatterplots (Figure 9). A number of combinations of parameters showed a significant separation of the NEA and SEA categories from the IA and ME categories (e.g. $\delta^{13}C$ of $C_{24}$ fatty acids vs $\delta^{13}C$ of $C_{29}$ *n*-alkanes; $\delta^{13}C$ of $C_{31}$ *n*-alkanes vs UCM) but no combinations could separate out the NEA from SEA categories or the IA from ME categories.

Prior to this comparison of the global $C_4$ biomass with back-trajectory analysis Fang *et al* (2000) had suggested that the increase in $C_4$ inputs in the summer at Chichi-Jima were a result of material carried by the easterly trade winds from the North and Central Americas, where a diverse $C_4$ flora exists (Still et al., 2003; Teeri and Stowe, 1976) (Figure 1). Indeed 6 of the 8 isotopically heaviest *n*-alkane samples (>29.5‰ or around > 40% $C_4$) were of marine easterly (ME) origin. However, even 20 day back-trajectory analysis did not place the source air-mass parcels in the Americas (data not shown). Therefore, while there may be a high proportion of $C_4$ material of American provenance in these samples, we can not confirm the thesis of Fang *et al*., (2000) that it is transported directly to Chichi-Jima by specific metereological events. Our analysis further suggests that the increase in $C_4$ contribution observed in the summer seasons at Chichi-Jima was also the result of relatively less $C_4$ poor material being transported from north Asia and relatively more $C_4$ rich material being transported from Southeast Asia and Australia (Figure 7) as well as from the Americas.

### 3.5 Investigation of individual samples

In section 3.2 (Figure 4c,d) we identified five samples for which we estimated the $\delta^{13}C$ contribution from non-higher plant sources to be greater than 1‰. The back trajectory fields for the samples included the following densely populated regions:

- Sample QFF-194: Japan, Korea and Northeast China.
- Sample QFF-214: Eastern China and Northern Japan.
- Sample QFF-217: Taiwan, the Philippines and Southern Japan.
- Sample QFF-222: Southern Japan and Eastern China.
- Sample QFF-420: Japan, Korea and Northeast China.

Therefore, in all of the above cases it appears that the source of the non-higher plant *n*-alkanes must be anthropogenic materials exported to Chichi-Jima as part of the "Asian outflow" which is frequently polluted by fossil fuel/biofuel derivatives (e.g. Fuelberg et al., 2003; Liu et al., 2003; Woo et al., 2003). However, many of the other samples collected at Chichi-Jima also had back-trajectories which included these regions. Therefore a precise reason for the above samples having a higher anthropogenic signal at Chichi-Jima may be due to temporal changes in the production of or transport efficiency of anthropogenic material.

Previously Kawamura *et al*. [2003] noted that absolute concentrations ($ngm^{-3}$) of terrestrial biomarkers were generally lower in the summer to autumn seasons due to the predominance of maritime trade winds. However, higher concentrations were recorded during the summer and fall of 1991. During this period the highest proportion of $C_4$ material at Chichi-Jima was discovered for the $C_{29}$ *n*-alkane (Figure 4a). The year of 1991 was characterized by a strong El Niño event, expressed meteorologically by weaker subtropical high-pressure systems in the western Pacific and more frequent development of tropical low systems (typhoons). The back-trajectories demonstrated that more air masses associated with this year originated in Southeast Asia, including Indonesia, Kalimantan and Australia (see Figure 7 and Kawamura et al., 2003). The $C_4$ contributions to aerosols at Chichi-Jima were therefore enhanced by emissions from these areas. Samples QFF-216 and QFF-219 (Figure 4a) had high concentrations of $C_4$ plant material (45 and 55% respectively for the $C_{29}$ *n*-alkane), these samples also had back-trajectories which reached to northern Australia (see Figure 10), a region with a biomass of up to 100% $C_4$ plants over large areas (Still et al., 2003). Also during the 1991 El Niño event, the air over the western equatorial Pacific became dry and hot and many forest fires occurred in southeast Asia (Kita et al., 2000) and Australia (Verdon et al., 2004), as well as the following year. During the forest fires and biomass burning, lipid class compounds, originally present as leaf waxes, could have been significantly emitted as smoke particles (without severe oxidative degradation) via volatilization (Abas et al., 1995; Oros and Simoneit, 2001). Thereafter lipids associated with smoke produced by smoldering and flaming processes may have been uplifted by convection and transported long distances over the Pacific Ocean, hence, enhancing the input of $C_4$ plant material detected at Chichi-Jima.

## 4 CONCLUSIONS

The *n*-alkane class - in the remote marine aerosols collected from the western North Pacific - contains predominantly long-chain compounds with a strong dominance of odd carbon numbered compounds. These lipids are derived from epicuticular waxes of terrestrial plants and indicate the importance of vegetation sources for organic matter in aerosols. Compound Specific Isotope Analysis (CSIA) of the *n*-alkanes reveals relatively greater contribution of $C_3$ plant material in the winter/spring and $C_4$ material in the summer/fall at Chichi-Jima. Comparison of backward trajectory analysis and a global map of $C_3$ /$C_4$ biomass showed that higher plant *n*-alkanes transported from the east Asian continent were isotopically distinct from those of a marine easterly or Indonesian/Australian origin. But further discrimination between Northeast Asian and Southeast Asian sources or between marine easterly and Indonesian/Australian sources was not possible using $\delta^{13}C$ of the

HMW *n*-alkanes or by various combinations of higher plant biomarker parameters. In general in winter/spring, more air masses transported $C_3$ material from north Asia, whereas in summer/fall relatively more air masses transported material from south east Asia, Indonesia and Australia where $C_4$ plants are more abundant. Marine easterlies were also an important source of isotopically heavier *n*-alkanes in the summer, which may have been transported from the Americas. The highest mean $C_4$ contributions in the summer of 1991 may have been enhanced by strong El Niño conditions which influenced meteorology and biomass burning emissions.

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**Table 1.** Summarized data for air temperature (T, °C), total carbon (TC, ug/m$^3$), normalized concentration of *n*-alkanes, carbon preference index (CPI, dimensionless), unresolved carbon mixture (UCM) and ACL.

| Season[a] | Number of samples | Air *T*(°C) | TC (µg/m$^3$) | normalized concn[b] | | | CPI[c] | UCM | ACL[d] |
|---|---|---|---|---|---|---|---|---|---|
| | | | | $\Sigma C_{19-26}$ | $\Sigma C_{27-40}$ | Σn-alkanes | | | |
| Winter 90 | 2 | 19.9 | 1.45 ± 0.3 | 1.77± 0.9 | 6.07± 2.5 | 7.84± 3.3 | 8.4 | 3.8 | 29.55 |
| Summer 90 | 12 | 26.5 | 0.47 ± 0.9 | 5.53± 0.7 | 19.88± 2.2 | 25.38± 2.8 | 3.3 | 5.8 | 29.75 |
| Winter 91 | 5 | 21.5 | 0.94 ± 1.2 | 1.23± 0.2 | 7.92± 0.5 | 9.14± 0.6 | 4.8 | 4.9 | 29.50 |
| Summer 91 | 10 | 27.2 | 0.50 ± 1.6 | 5.86± 0.5 | 30.33± 3.2 | 36.20± 3.7 | 12.4 | 3.7 | 30.11 |
| Winter 92 | 10 | 19.9 | 0.78 ± 1.1 | 4.44± 0.5 | 29.78± 4.7 | 34.21± 5.2 | 3.8 | 3.8 | 29.80 |
| Summer 92 | 9 | 25.0 | 0.43 ± 1.7 | 2.84± 0.4 | 22.49± 3.2 | 25.33± 3.6 | 2.4 | 5.0 | 29.70 |
| Winter 93 | 10 | 19.0 | 0.71 ± 0.5 | 5.56± 0.4 | 29.56± 2.3 | 35.04± 2.6 | 3.8 | 4.2 | 29.75 |
| Summer 93 | 11 | 26.6 | 0.40 ± 1.0 | 3.06± 0.3 | 22.88± 1.9 | 25.93± 2.2 | 6.6 | 4.0 | 30.03 |

[a]Summer: May-October; winter: November-April. [b]Concentration of *n*-alkanes normalized to total carbon (TC), ng of *n*-alkanes /µg of C. [c] $CPI_{n\text{-alkanes}} = 2\ \Sigma\ \text{odd}\ (C_{25-35})/[\Sigma\text{even}\ (C_{24-34}) + \Sigma\text{even}\ (C_{26-36})]$. [d]Average chain length: the concentrated-weighted mean carbon chain length).

**Table 2.** Stable carbon isotopic analyses of *n*-alkanes and the calculated corrections for non-higher plant contributions.

| Sample | Season | Source Allocation* | $\delta^{13}C$ values | | | $\delta^{13}C_{29}$ corr.[a] | $\delta^{13}C_{29}$ $\Delta^b$ | $\delta^{13}C_{31}$ corr.[a] | $\delta^{13}C_{31}$ $\Delta^b$ | %C4 ($C_{29}$) | %C4 ($C_{31}$) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | $C_{27}$ | $C_{29}$ | $C_{31}$ | | | | | | |
| QFF-181 | Winter 90 | NE A | -31.81 | -32.62 | -33.21 | -33.29 | -0.67 | -33.99 | -0.78 | 17 | 15 |
| QFF-182 | | SE A | | | | | | | | | |
| QFF-183 | Summer 90 | ME | -29.45 | -30.47 | -31.33 | -30.66 | -0.19 | -31.57 | -0.24 | 31 | 28 |
| QFF-184 | | ME | -31.21 | -32.41 | -33.39 | -33.01 | -0.60 | -34.11 | -0.73 | 18 | 14 |
| QFF-185 | | ME | -30.14 | -30.88 | -32.02 | -31.35 | -0.47 | -32.70 | -0.68 | 28 | 23 |
| QFF-187 | | ME | | -31.34 | -32.65 | -31.34 | | -32.65 | | 25 | 19 |
| QFF-188 | | ME | | | | | | | | | |
| QFF-189 | | ME | | -31.02 | -30.48 | -31.02 | | -30.48 | | 27 | 33 |
| QFF-190 | | ME | | -30.05 | -31.07 | -30.05 | | -31.07 | | 34 | 29 |
| QFF-191 | | ME | | | | | | | | | |
| QFF-192 | | ME | | | | | | | | | |
| QFF-193 | | ME | | -28.63 | -28.40 | -28.63 | | -28.40 | | 43 | 47 |
| QFF-194 | | ME | -29.16 | -30.48 | -32.75 | -32.29 | -1.81 | -35.88 | -3.13 | 31 | 18 |
| QFF-195 | | ME | | | | | | | | | |
| QFF-207 | Winter 91 | ME | -29.19 | -31.75 | -30.97 | -31.75 | | -30.97 | | 22 | 30 |
| QFF-208 | | NE A | -30.34 | -32.04 | -32.37 | -32.98 | -0.95 | -33.41 | -1.04 | 21 | 21 |
| QFF-209 | | NE A | -32.21 | -32.66 | -31.80 | -33.00 | -0.34 | -32.08 | -0.29 | 16 | 25 |
| QFF-210 | | SE A | -30.24 | -30.88 | -31.40 | -30.94 | -0.06 | -31.47 | -0.07 | 28 | 27 |
| QFF-211 | | SE A | -30.46 | -30.73 | -31.94 | -30.73 | | -31.94 | | 29 | 24 |
| QFF-212 | Summer 91 | ME | | | | | | | | | |
| QFF-214 | | SE A | -28.28 | -29.31 | -29.15 | -30.72 | -1.42 | -30.49 | -1.35 | 39 | 42 |
| QFF-216 | | IA | -28.31 | -28.69 | -30.00 | -29.17 | -0.49 | -30.74 | -0.74 | 43 | 36 |
| QFF-217 | | IA | -30.20 | -31.01 | -30.94 | -33.38 | -2.37 | -33.25 | -2.31 | 27 | 30 |
| QFF-218 | | IA | | | | | | | | | |
| QFF-219 | | IA | -28.08 | -27.23 | -27.56 | -27.23 | | -27.56 | | 53 | 53 |
| QFF-220 | | IA | -29.07 | -30.00 | -29.95 | -30.00 | | -29.95 | | 34 | 37 |
| QFF-221 | | IA | -33.12 | -33.38 | -34.44 | -33.50 | -0.12 | -34.61 | -0.17 | 12 | 7 |
| QFF-222 | | ME | -29.09 | -29.48 | -29.23 | -31.28 | -1.80 | -30.88 | -1.65 | 38 | 42 |
| QFF-223 | | NE A | -31.22 | -32.82 | | -32.82 | | | | 15 | |
| QFF-393 | Winter 92 | NE A | -30.92 | -32.55 | -31.92 | -33.36 | -0.81 | -32.58 | -0.67 | 17 | 24 |
| QFF-394 | | NE A | | | | | | | | | |
| QFF-395 | | NE A | -29.27 | -33.07 | -32.94 | -33.44 | -0.37 | -33.30 | -0.36 | 14 | 17 |
| QFF-396 | | NE A | -30.81 | -32.88 | -32.64 | -33.43 | -0.55 | -33.16 | -0.52 | 15 | 19 |
| QFF-397 | | NE A | -32.38 | -33.35 | -32.71 | -33.83 | -0.49 | -33.13 | -0.42 | 12 | 19 |
| QFF-399 | | NE A | -32.02 | -33.14 | -31.66 | -31.51 | -0.34 | -29.97 | -0.28 | 13 | 25 |
| QFF-400 | | SE A | -30.44 | -32.83 | -31.83 | -32.83 | | -31.83 | | 15 | 24 |
| QFF-401 | | SE A | -31.10 | -33.05 | -32.36 | -33.22 | -0.17 | -32.49 | -0.13 | 14 | 21 |
| QFF-402 | | NE A | -31.58 | -32.17 | -32.84 | -32.34 | -0.17 | -33.07 | -0.23 | 20 | 18 |
| QFF-403 | | SE A | -31.43 | -33.29 | -34.32 | -32.21 | | -32.89 | | 12 | 8 |
| QFF-404 | Summer 92 | NE A | -30.22 | -30.68 | -32.56 | -30.68 | | -32.56 | | 30 | 19 |
| QFF-405 | | SE A | -30.04 | -33.49 | -32.89 | -33.49 | | -32.89 | | 11 | 17 |
| QFF-406 | | ME | -29.52 | -29.96 | -29.32 | -30.01 | -0.05 | -29.36 | -0.04 | 34 | 41 |
| QFF-407 | | IA | -31.49 | -32.44 | -30.94 | -33.04 | -0.60 | -31.36 | -0.42 | 18 | 30 |
| QFF-408 | | ME | | -29.11 | -28.66 | -29.11 | | -28.66 | | 40 | 45 |
| QFF-410 | | IA | -29.95 | -30.83 | -30.51 | -30.83 | | -30.51 | | 29 | 33 |
| QFF-412 | | IA | -28.47 | -29.80 | -30.02 | -29.94 | -0.14 | -30.16 | -0.14 | 36 | 36 |
| QFF-413 | | ME | | -29.11 | -32.10 | -29.11 | | -32.10 | | 40 | 23 |
| QFF-414 | | NE A | -30.55 | -30.83 | -32.68 | -31.48 | -0.17 | -32.90 | -0.23 | 29 | 19 |
| QFF-415 | Winter 93 | NE A | -30.35 | -31.03 | -29.86 | -31.03 | | -29.86 | | 27 | 37 |
| QFF-416 | | NE A | | | | | | | | | |
| QFF-417 | | NE A | -32.32 | -33.71 | -33.21 | -33.96 | -0.25 | -33.42 | -0.21 | 9 | 15 |
| QFF-418 | | SE A | -29.54 | -31.71 | -31.52 | -32.18 | -0.47 | -31.96 | -0.45 | 23 | 26 |
| QFF-419 | | NE A | -33.53 | -34.26 | -33.79 | -34.45 | -0.19 | -33.95 | -0.17 | 6 | 11 |
| QFF-420 | | NE A | -29.87 | -31.42 | -31.65 | -33.28 | -1.86 | -33.59 | -1.94 | 25 | 25 |
| QFF-421 | | NE A | -31.47 | -33.42 | -33.15 | -33.42 | | -33.15 | | 11 | 16 |
| QFF-422 | | NE A | -30.87 | -32.45 | -31.76 | -32.71 | -0.26 | -31.97 | -0.22 | 18 | 25 |
| QFF-423 | | NE A | -30.73 | -32.12 | -32.07 | -32.12 | | -32.07 | | 20 | 23 |
| QFF-424 | | NE A | -31.69 | -32.87 | -33.65 | -33.15 | -0.29 | -34.05 | -0.40 | 15 | 12 |
| QFF-426 | Summer 93 | SE A | -29.91 | -33.49 | -32.23 | -33.49 | | -32.23 | | 11 | 22 |
| QFF-427 | | IA | -29.75 | -31.55 | -32.92 | -31.55 | | -32.92 | | 24 | 17 |
| QFF-428 | | IA | | | | | | | | | |
| QFF-429 | | NE A | | | | | | | | | |
| QFF-430 | | ME | -28.64 | -28.32 | -26.56 | -28.32 | | -26.56 | | 46 | 59 |
| QFF-431 | | IA | | -30.05 | -30.07 | -30.05 | | -30.07 | | 34 | 36 |
| QFF-432 | | ME | | | | | | | | | |
| QFF-433 | | IA | -29.40 | -28.83 | -28.77 | -28.83 | | -28.77 | | 42 | 45 |
| QFF-434 | | ME | | | | | | | | | |
| QFF-435 | | ME | | | | | | | | | |
| QFF-436 | | NE A | | | | | | | | | |
| QFF-437 | | NE A | -30.94 | -32.57 | -32.67 | -32.57 | | -32.67 | | 17 | 19 |

* Predominant source area in the 10 day back-trajectory. Acronyms are: NEA = Northeast Asia; SEA = Southeast Asia; Marine Easterlies; IA = Indonesia and Australia

n.d. = not determined, only isotopic data for *n*-alkanes with carbon numbers from $C_{24}$ to $C_{33}$ are shown, measurements for other carbon numbers ($C_{19} – C_{23}$, $C_{34} – C_{37}$) were sparse and have been omitted.

[a] $\delta^{13}C$ values of respective *n*-alkanes corrected for fossil fuel/ marine contribution.

[b] Difference in $\delta^{13}C$ between uncorrected and corrected values for respective *n*-alkanes.

**Table 3.** Stable carbon isotopic compositions (per mil) of *n*-alkanes from $C_3$ and $C_4$ plants.

| Plant Type | Source | $C_{29}$ $\delta^{13}C$ (s.d.) | $C_{31}$ $\delta^{13}C$ (s.d.) |
|---|---|---|---|
| **$C_3$ Plants** | Collister et al, 1997. 9 species | -35.4‰ (1.7) | -36.1‰ (1.3) |
| | Chikaraishi and Naraoka, 2003. 22 species | -35‰ (2.7) | -35.2‰ (2.9) |
| | ILTS this paper 1 species | -33.7‰ | |
| | Mean | -35.1‰ (1.7) | -35.5‰ (1.3) |
| **$C_4$ Plants** | Collister et al, 1997. 3 species | -21.5‰ (4.3) | -20.2‰ (2.3) |
| | Chikaraishi and Naraoka, 2003. 7 species | -19.2‰ (2.4) | -20.2‰ (2.3) |
| | ILTS this paper 1 species | -19.6‰ | -20.3‰ |
| | Mean | -20.2‰ (2.3) | -20.4‰ (2) |

* Measurements made at ILTS, plant leaves were extracted with DCM/MeOH (5:1) and then the extracts were saponified using 0.5 M KOH/MeOH. The *n*-alkanes were isolated and analysed by the methods described in the text.

**Table 4.** Calculated % $C_4$ source input of plants from end members* of $C_3$ and $C_4$ plants.

| Compound | W-90 | S-90 | W-91 | S-91 | W-92 | S-92 | W-93 | S-93 |
|---|---|---|---|---|---|---|---|---|
| $C_{29}$ *n*-alkane (this paper) | 17 | 30 | 23 | 33 | 15 | 30 | 17 | 29 |
| $C_{31}$ *n*-alkane (this paper) | 15 | 26 | 25 | 35 | 19 | 29 | 21 | 33 |
| $C_{28}$ fatty acid Fang *et al* (2000) | 6 | 20 | 22 | 27 | 9 | 14 | 8 | 18 |

* For the $C_{28}$ fatty acid Fang *et al* (2000) used end members of -30.9‰ for $C_3$ plants and -20.8‰ for $C_4$ plants.

**Figure 1.** Global map showing the sampling location (Chichi-Jima) in the western Pacific, the global C4 percentage of terrestrial biomass and the source areas of the four major air mass trajectories. Abbreviations: NE A = North east Asia; SE A = South east Asia; IA = Indonesia and Australia; ME = Marine easterlies. The C4 biomass is adapted from Still et al., 2003, the C4 biomass fraction was modelled by combining remote sensing products, physiological modelling, a spatial distribution of global crop fractions, and national harvest area data for major crop types.

**Figure 1:** Variations of wind direction at 700 hPa over Chichi-Jima in the western North Pacific (see a map in Figure 1).

**Figure 3:** Typical distribution of n-alkane concentrations and isotopic values from a North east Asian sample (QFF-417-N1) collected at Chichi-Jima, December 21-25, 1992.

**Figure 4:** Distribution of carbon isotopic composition (δ13C, left axis) of C29 and C31 *n*-alkanes and estimates of the percentage contribution of C4 plant material (%C4, right axis) in aerosol samples collected at Chichi-Jima Island between 1990 and 1993. a & b) uncorrected δ13C values of C29 and C31. c & d) δ13C values of C29 and C31 corrected for non-higher plant inputs (open circles) compared to uncorrected values (black circles), the samples with the largest corrections are highlighted by arrows. Grey shading delimits the summer (May-October) and white the winter seasons (November-April).

**Figure 5:** Linear correlation between carbon isotopic compositions (δ13C) of the C29 *n*-alkane (this paper) and the C24 Fatty Acid (Fang *et al*, 2000) in aerosol samples collected at Chichi-Jima Island between 1990 and 1993.

**Figure 6:** Seasonal mean distributions of carbon isotopic composition (δ13C) of C29 & C31 *n*-alkanes in aerosol samples collected at Chichi-Jima Island between 1990 and 1993.

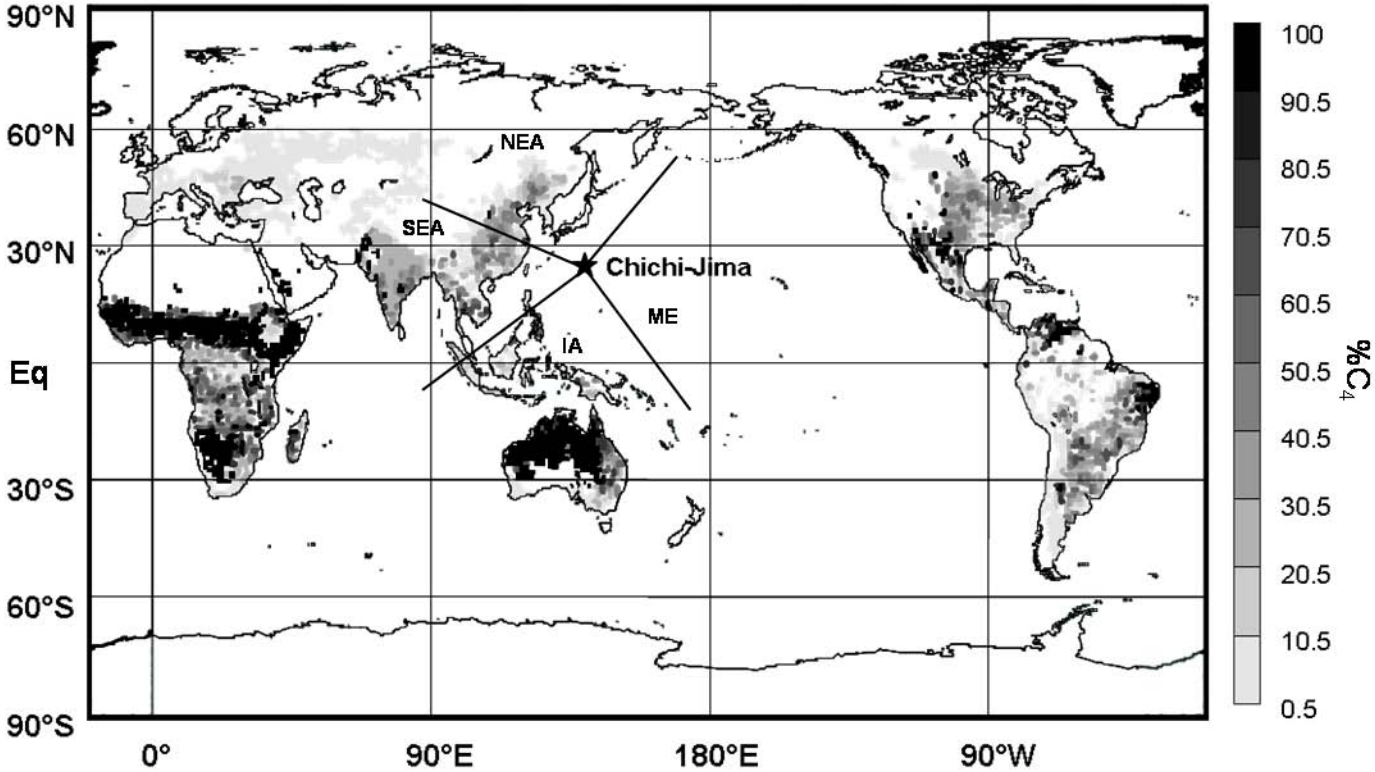
**Figure 7:** Composite seasonal 10 day Back-trajectory analyses for the sampling periods. Cnducted using the Fifth-Generation NCAR/Penn State mesocale model (MM5). The trajectory calculations were based on backward tracking of air parcels released at the target location (for the sampling periods) assuming the parcel was moving along the ambient airflow. The flow pattern was updated every 6 hours.

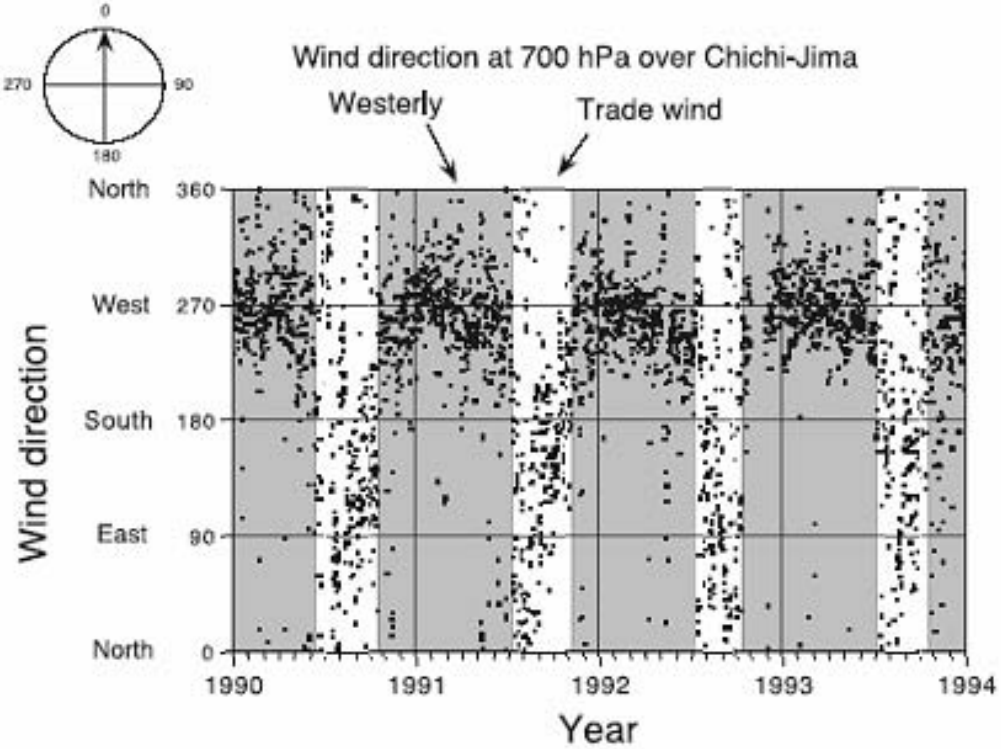
**Figure 8:** Boxplots of the carbon isotopic composition (δ13C) of C29 & C31 *n*-alkanes in aerosol samples collected at Chichi-Jima Island between 1990 and 1993, catagorised by back-trajectory source area. Each box shows the median (black line), the interquartile range (box), the min and max values that are *not* outliers (whiskers), mild outliers (circles) and extreme outliers (stars).

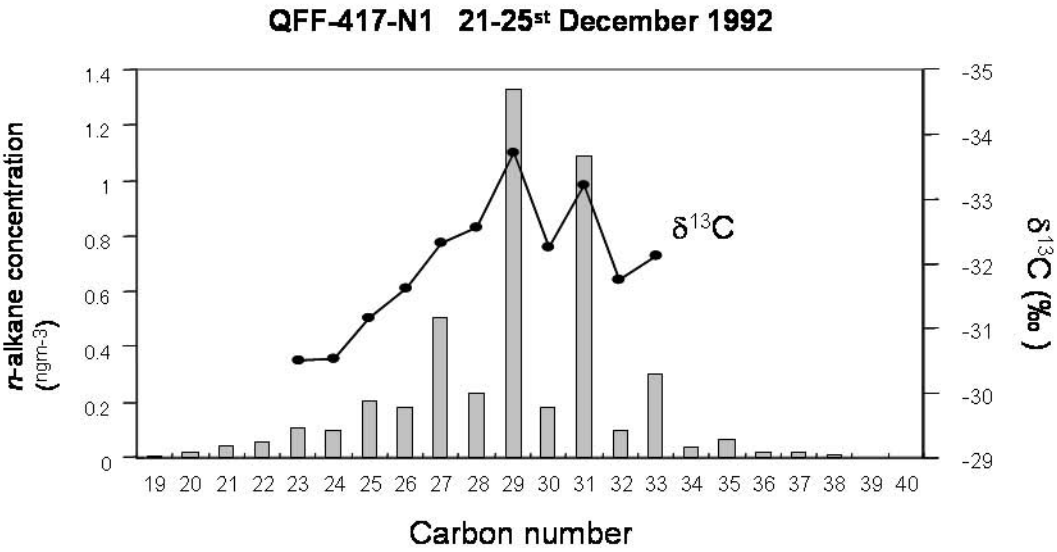
**Figure 9:** Data matrix showing plots of all possible combinations of the following variables measured in the aerosols collected at Chichi-Jima: C29 *n*-alkane δ13C; C31 *n*-alkane δ13C; C24 *FA* (fatty acid) δ13C; CPI; UCM –total n-alkanes; ACL (C27-C33). The coloured markers correspond to the source allocations.

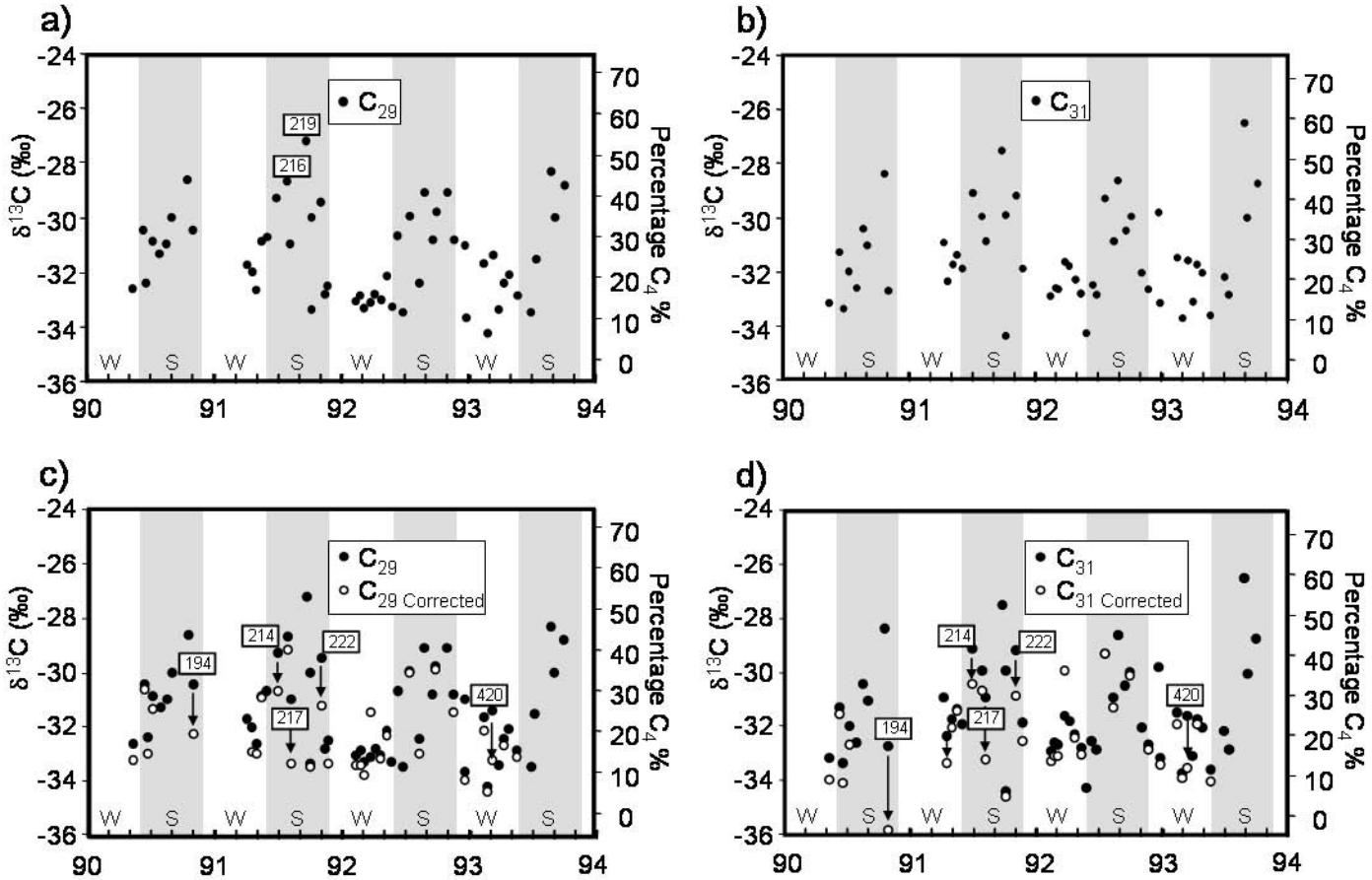
**Figure 10:** Composite seasonal 15 day Back-trajectory analyses for a) QFF-216 and b) QFF-219. Conducted using the Fifth-Generation NCAR/Penn State mesocale model (MM5). The trajectory calculations were based on backward tracking of air

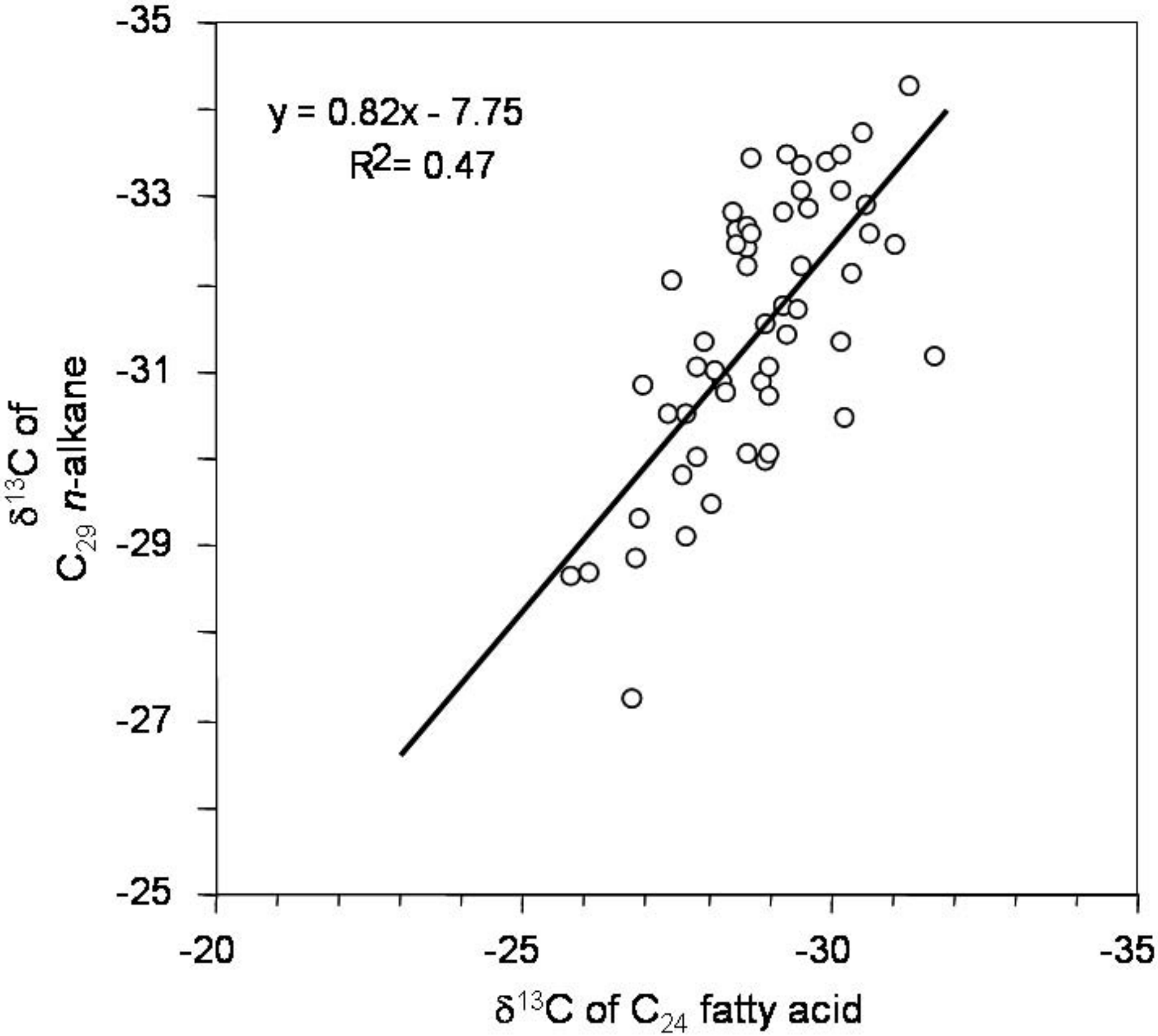
parcels released at the target location (for the sampling periods) assuming the parcel was moving along the ambient airflow. The flow pattern was updated every 6 hours.

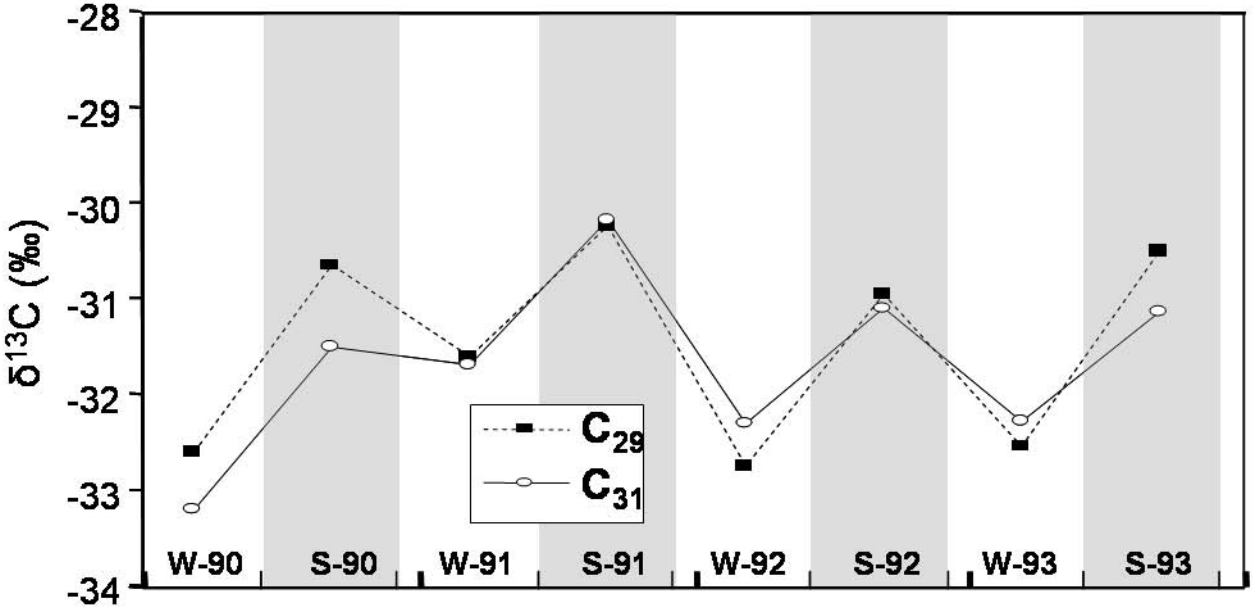

90°N
60°N
30°N
Eq
30°S
60°S
90°S
0°
90°E
180°E
90°W
NEA
SEA
Chichi-Jima
ME
IA
100
90.5
80.5
70.5
60.5
50.5
40.5
30.5
20.5
10.5
0.5
%C4


Wind direction at 700 hPa over Chichi-Jima
Westerly
Trade wind
0
270
90
180
North
West
South
East
North
360
270
180
90
0
Wind direction
1990
1991
1992
1993
1994
Year

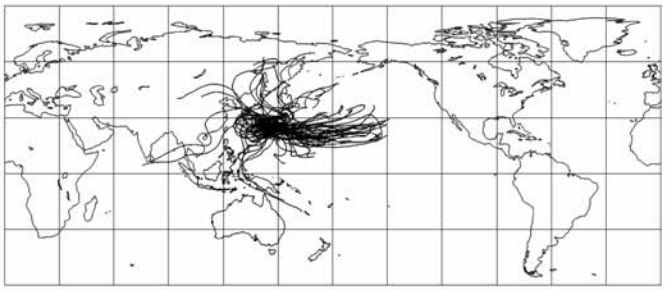

QFF-417-N1 21-25st December 1992
n-alkane concentration (ngm-3)
1.4
1.2
1
0.8
0.6
0.4
0.2
0
-35
-34
-33
-32
-31
-30
-29
δ13C (‰)
δ13C
19 20 21 22 23 24 25 26 27 28 29 30 31 32 33 34 35 36 37 38 39 40
Carbon number


a)
b)
c)
d)
δ13C (‰)
Percentage C4 %
C29
C31
C29 Corrected
C31 Corrected
219
216
194
214
222
217
420
W
S
-24
-26
-28
-30
-32
-34
-36
0
10
20
30
40
50
60
70
90
91
92
93
94

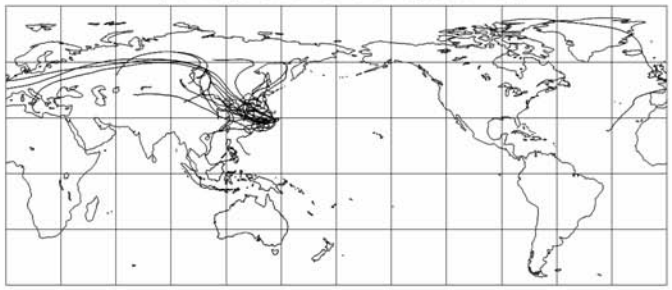

y = 0.82x - 7.75
R2= 0.47
-35
-33
-31
-29
-27
-25
-20
-25
-30
-35
δ13C of C29 n-alkane
δ13C of C24 fatty acid


-28
-29
-30
-31
-32
-33
-34
δ13C (‰)
C29
C31
W-90
S-90
W-91
S-91
W-92
S-92
W-93
S-93

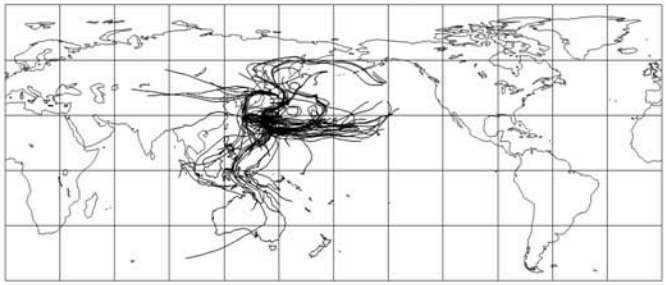
1990 Summer trajectories
(10 days backward trajectories)



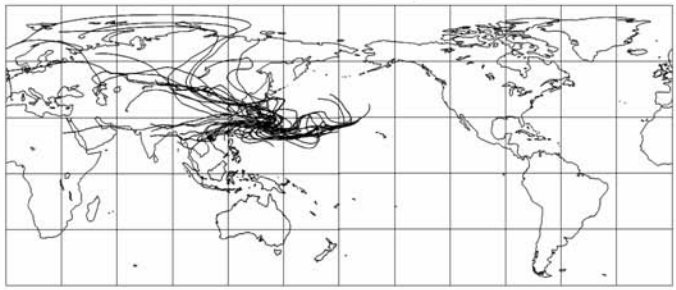
Winter 1990
(10 days backward trajectories)



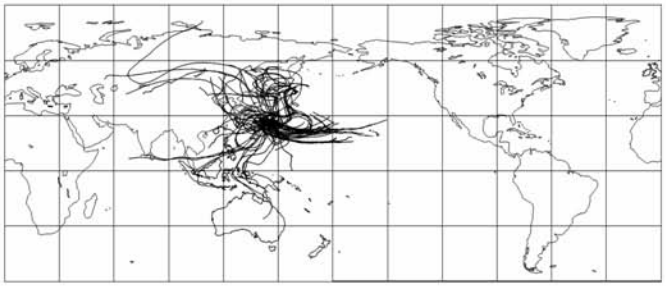
Summer 1991
(10 days backward trajectories)



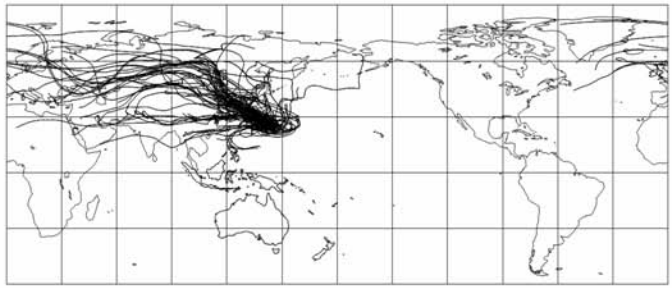
Winter 1991
(10 days backward trajectories)



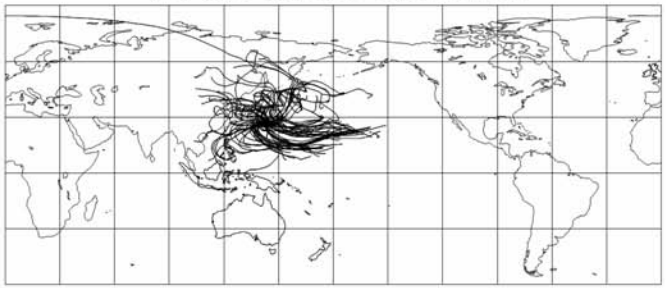
Summer 1992
(10 days backward trajectories)



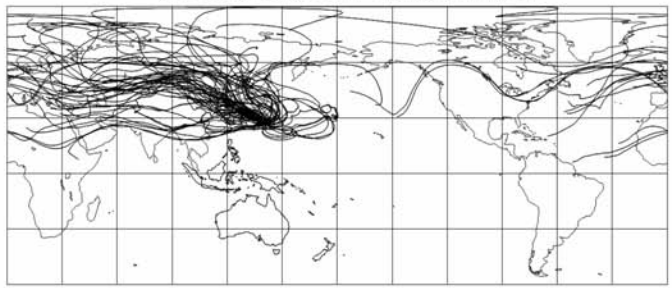
Winter 1992
(10 days backward trajectories)

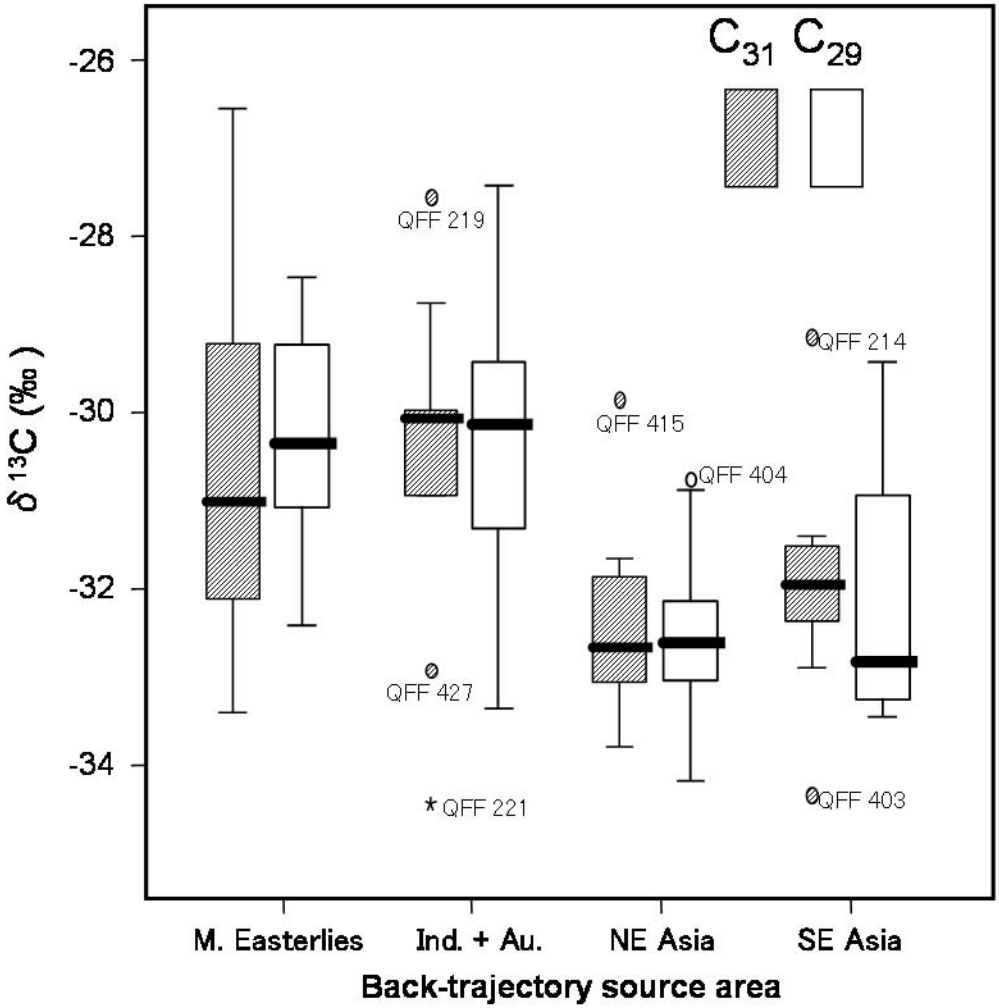


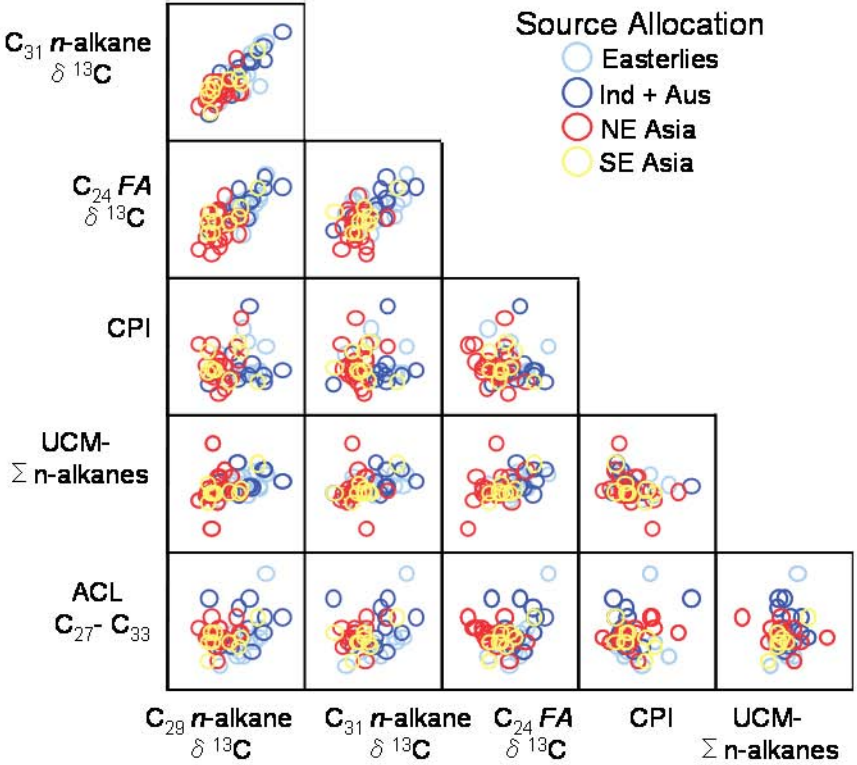
Summer 1993
(10 days backward trajectories)



Winter 1993
(10 days backward trajectories)

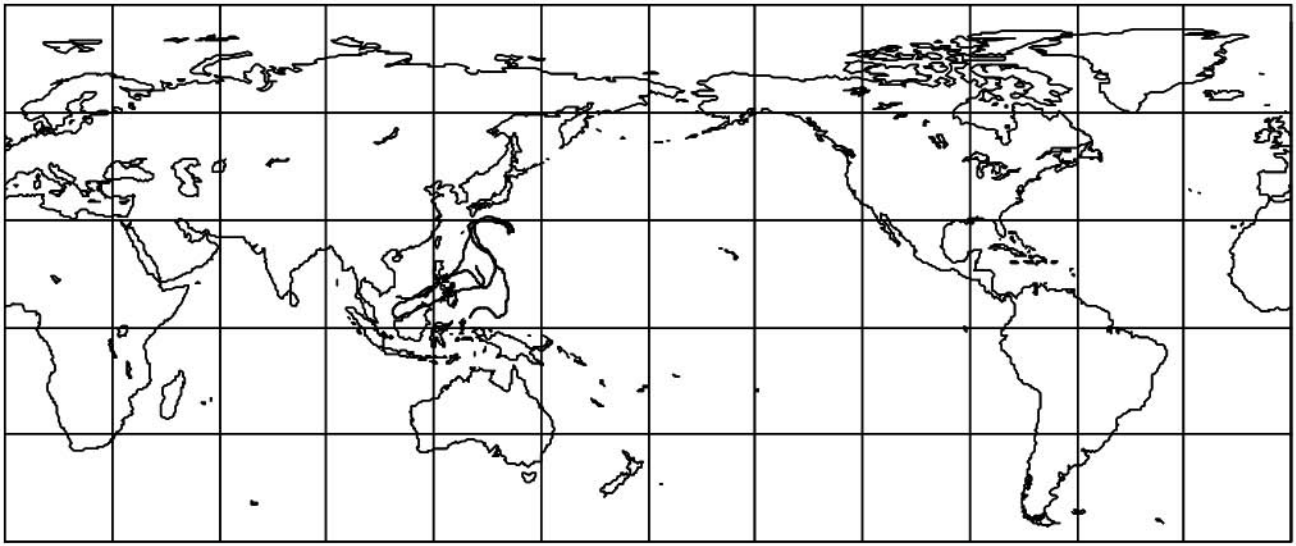



C31 C29
-26
-28
-30
-32
-34
δ 13C (‰)
QFF 219
QFF 415
QFF 404
QFF 214
QFF 427
QFF 221
QFF 403
M. Easterlies
Ind. + Au.
NE Asia
SE Asia
Back-trajectory source area


Source Allocation
Easterlies
Ind + Aus
NE Asia
SE Asia
C31 n-alkane δ 13C
C24 FA δ 13C
CPI
UCM- Σ n-alkanes
ACL C27- C33
C29 n-alkane δ 13C
C31 n-alkane δ 13C
C24 FA δ 13C
CPI
UCM- Σ n-alkanes

a)

QFF-216 June 1991 925-hPa
(15 days backward trajectories)



b)

QFF-219 August 1991 925-hPa
(15 days backward trajectories)