to the Gleissberg minimum early in this century. However, the observed increase of about 20% is four times that expected based on the sunspot numbers.

The observed high ^{44}Ti and the inferred high GCR flux during the Gleissberg minimum suggest a much-weakened modulation of GCR during prolonged solar quiet periods. The heliosphere, during the 11-year solar cycles, is divided into two hemispheres of opposite polarity by a wavy heliospheric neutral sheet (HNS) whose inclination to the sun's rotational equator increases from a few degrees at solar minimum to >70° at solar maximum and whose undulations also increase with solar activity (*15*). The HMF is relatively smooth during the solar minima. The enhanced GCR fluxes required for the higher production of ^{44}Ti can be achieved if (i) the HNS becomes smooth and remains close to the helioequator; (ii) the HMF becomes weak, more regular, and orderly; or (iii) the size of the heliosphere shrinks during a prolonged period of low sunspot numbers, so that a larger flux of cosmic rays can enter into the inner heliosphere. The GCR flux, J_G(>1 GeV), near 1 AU during maximum and minimum of the 11-year solar cycle (*14, 16*) in the past four Schwabe cycles, varied between 1.4 and 2.2 to 2.5 protons/cm^2·s·4π sr, with an average value of 1.7 protons/cm^2·s·4π sr. The observed activity can be explained if J_G varied between 2.2 and 3.6 protons/cm^2·s·4π sr at 1 to 3 AU during solar cycles 12 to 15 covering the Gleissberg minimum at the turn of the century. Such a regularity of HMF may also justify the even higher GCR fluxes estimated for the Maunder minimum, when sunspot numbers were low for about 70 years (1645 to 1715), and can explain the high activities of ^{14}C and ^{10}Be observed on Earth (*17*).

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17 July 1995; accepted 3 October 1995

Particle Formation in the Upper Tropical Troposphere: A Source of Nuclei for the Stratospheric Aerosol

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Atmospheric measurements and numerical calculations described here indicate that binary homogeneous nucleation of H_2SO_4-H_2O particles occurs in the upper tropical troposphere. Particle concentrations decrease with increasing altitude above the tropical tropopause as a result of coagulation during the upward air transport produced by stratospheric circulations. During the extended periods of time that volcanic eruptions do not strongly influence stratospheric particle number concentrations, particles formed in the upper tropical troposphere provide nuclei upon which oxidized sulfur gases condense in the stratosphere. This particle source, coupled with aerosol microphysical properties and atmospheric transport, governs the number concentration of particles in the lower tropical and mid-latitude stratosphere.

The stratospheric aerosol layer, or Junge layer, is a fine mist of highly concentrated liquid sulfuric acid (H_2SO_4) particles that envelops the Earth at altitudes from a few kilometers above the tropopause (~9 to 17 km) to ~28 km. The particles are typically 0.1 to 0.3 μm in diameter and are present at number mixing ratios (NMR, number of particles per milligram of air) of ~60 mg^{-1} (*1–3*). These stratospheric H_2SO_4-H_2O particles provide sites for heterogeneous reactions that repartition nitrogen species, ultimately leading to reduced ozone concentrations (*4, 5*). In the polar regions, reactions on these particles may also activate chlorine, which catalytically destroys ozone (*6*). In both of these cases, the surface area provided by the particles for the heterogeneous reactions is an important parameter in the chemical process. In the polar vortices, H_2SO_4-H_2O particles are the nuclei on which polar stratospheric clouds (PSCs) form (*7*). Changes in the concentration of PSC nuclei may change the concentration of PSC particles, the surface area available for chlorine activation, and the process of particle sedimentation that leads to denitrification and polar ozone depletion (*8*).

The surface area available for heterogeneous reactions in the stratosphere depends largely on the number of particles upon which the total particle mass is distributed. Most of the mass of the stratospheric aerosol layer originates from reduced sulfur gases (OCS, CS_2, and SO_2) transported from the troposphere (*2*). These gases oxidize in the stratosphere to H_2SO_4, which then condenses onto preexisting particles (nuclei). In volcanically quiescent periods, OCS is believed to be the dominant source of stratospheric H_2SO_4 mass (*9*). The sources of the nuclei on which the H_2SO_4 condenses are poorly known. Many particles form in the stratosphere from homogeneous nucleation of H_2SO_4-H_2O after major volcanic injections of SO_2 (*10, 11*). However, within ~6 months after such eruptions, stratospheric NMRs decay to near prevolcanic values (*12*). When volcanic effects are small, the stratosphere's degree of supersaturation with H_2SO_4 vapor is insufficient for particle formation (*3, 10, 13*), with some minor exceptions (*14*). These findings im-

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ply that there are sources of particles outside of the stratosphere that provide the nuclei for maintenance of the Junge layer during volcanically quiescent periods, which often extend for several years (*3*).

Possible sources of stratospheric nuclei include condensed meteoritic vapor (*15*) and particles transported from the troposphere. Measurements of particle composition (*16*) and consideration of the upwelling air motions in the tropical stratosphere (*17*) provide evidence against a significant meteoritic component in particles below an altitude of 25 km. Although some studies have suggested that the troposphere could provide a source of nuclei for the stratospheric aerosol (*1*, *2*, *10*, *18*), there is only limited observational evidence for tropospheric particles entering the lower stratosphere (*1*, *19*).

The measurements analyzed here were obtained in the mid-latitudes, polar regions, and tropics in both hemispheres between 1987 and 1994 (*20*). We excluded data known to be affected by volcanic eruptions and by particles formed in the polar vortices (*14*). Measurements made in fall, winter, and spring dominate the extratropical data. Condensation nucleus counters (CNCs) measuring the NMR of particles in the diameter (D_p) range of 0.008 to ~3 μm (*21*) were flown on NASA U-2 and ER-2 aircraft to altitudes of ~21 km. Because the stratospheric aerosol population is usually dominated by particles in this size range, the total particle NMR was well represented by the measurements. In 1993 and 1994, a second CNC measured nonvolatile-particle NMR by heating the aerosol stream to 192°C before counting.

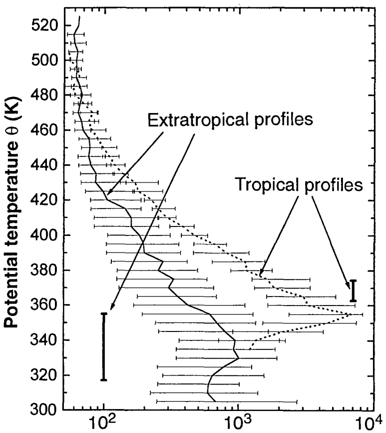

Fig. 1. Vertical profiles of particle NMR measured outside of the tropics (solid line) and within the tropics (dashed line), as defined by meteorological analysis and tracer relations. Data from each profile that penetrated the tropopause were binned into 5 K increments of θ, and the median values (lines) and interquartile ranges (containing the middle 50% of the data, shown as error bars) of these binned values were calculated. All profiles are thus weighted equally. The range of potential temperatures enclosing the tropopause heights for each data set are indicated by vertical bars.

Every profile that penetrated the tropopause was classified as tropical or extratropical on the basis of an analysis of chemical-tracer and meteorological data (*22*). The data were grouped into intervals of potential temperature θ, and the medians and interquartile ranges were calculated (Fig. 1). Potential temperature is the temperature that an air parcel would have were it compressed or expanded adiabatically from the local pressure to 10^5 Pa. Horizontal air motions in the stratosphere approximately follow constant-θ surfaces over time scales of a few weeks.

A maximum in particle NMR is present near the tropopause in both tropical and extratropical cases. This maximum is about seven times larger inside the tropics than outside. Particle NMRs decrease smoothly with θ above the tropical tropopause, reaching extratropical values near 470 K. The relative variability of the extratropical NMRs is considerably larger than that for tropical values between the tropopause and 440 K. High particle NMRs were found on two occasions in the upper troposphere outside of the tropics; thus, the processes that lead to the high particle NMRs are not unique to the tropics but are more common and widespread there.

Two pieces of evidence suggest that the particles observed near the tropical tropopause are composed of H_2SO_4-H_2O droplets. Laboratory studies show that the heated inlet of the CNC used in 1993 and 1994 is effective at volatilizing >90% of all H_2SO_4-H_2O particles with $D_p < 0.04$ μm. Near the maximum in particle mixing ratio just below the tropical tropopause, >95% of the particles were volatile at 192°C (Fig. 2A), consistent with the particles being composed of H_2SO_4-H_2O. Electron microscope analysis of individual particles collected by an impactor (*16*) just below the tropical tropopause at 20°N on 3 May 1993 showed the particles to be nearly monodisperse with diameters of ~0.015 μm and to contain sulfur as the only element significantly above background (*23*). Given these observations and the well-documented presence of sulfur gases and H_2SO_4 particles in the free troposphere (*16*, *24*), we conclude that a particle composition of H_2SO_4-H_2O is likely.

Near-tropopause measurements of particle NMRs taken at extratropical locations contained a much smaller proportion of volatile particles (Fig. 2B). The likely cause of the reduced particle volatility is incorporation over time of small amounts of ammonia into the particles to form partially neutralized sulfate particles, which the heated inlet on the CNC is unable to volatilize. Thus, low concentrations of nonvolatile particles near the mid-latitude tropopause probably indicate that the particles were not recently formed by homogeneous nucleation of H_2SO_4 and H_2O.

The above measurements suggest that particles composed of H_2SO_4 and H_2O nucleate near the tropical tropopause. We have evaluated the formation rate of H_2SO_4-H_2O particles using a numerical model of homogeneous bimolecular nucleation (*25*). This model simulates nucleation rates under conditions in which the pure substances (H_2O and H_2SO_4) are subsaturated. Because of well-known uncertainties in the classical nucleation theory used for this calculation (*26*), we use the results as a qualitative indication that nucleation is likely or unlikely to occur, rather than as a quantitative estimate of particle production rates.

Vertical profiles of temperature, pressure, and H_2O mixing ratio measured aboard the ER-2 (*27*) were used as input to the numerical model. The mixing ratio of H_2SO_4 was estimated from a one-dimensional numerical model of the stratospheric aerosol (*28*). The height of the local tropopause was determined from in situ measurements and from a remote-sensing microwave temperature profiler on board the ER-2 (*29*). Particle nucle-

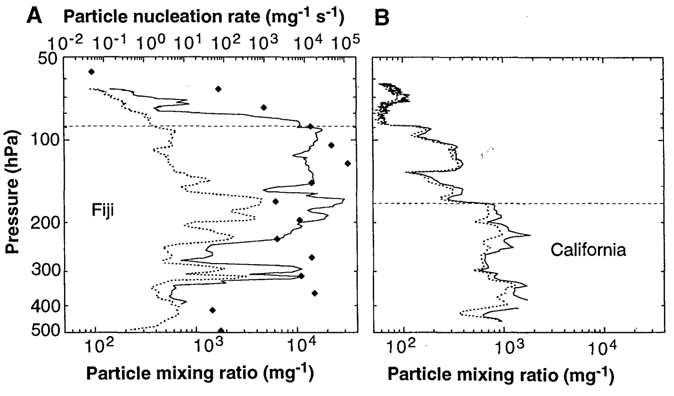

Fig. 2. Vertical profiles of total particle mixing ratio (solid line) and nonvolatile particle mixing ratio (dotted line). Volatile particles (the difference between the lines) are interpreted as being mostly H_2SO_4-H_2O. The approximate height of the tropopause is shown by the horizontal dashed lines. (**A**) Profile measured in tropical air during an ascent from Nadi, Fiji, on 29 March 1994. The formation rate of H_2SO_4-H_2O particles calculated from a binary homogeneous nucleation model with input data measured during this profile is shown by the symbols. (**B**) Profile measured in mid-latitude air during an ascent from northern California on 14 May 1993.

ation rates (in particles per milligram of air per second) as a function of altitude were calculated for the ascent from Fiji on 29 March 1994 (Fig. 2A). The model results indicate a relative tendency for nucleation in the upper troposphere, with a maximum nucleation rate at a pressure of 120 hPa, about 2 km below the tropopause. The shape of the nucleation rate profile is qualitatively similar to that of the measured particle NMR profile. The measurements show a rapid decrease in particle NMR just above the tropopause, suggesting that particle formation is not important in this region.

The results of the nucleation modeling support the observational evidence that new H_2SO_4-H_2O particles are formed by homogeneous binary gas-to-particle conversion in the upper tropical troposphere. This maximum in particle NMR lies in a region of net upward mass transport of air through the tropical tropopause and into the middle stratosphere (*17*). This net upwelling is a component of the residual circulation of the stratosphere. The mechanisms of cross-tropopause transport are poorly understood but probably involve penetration of the lower stratosphere by convective clouds in the tropics (*30*); small particles may survive such cloud processing (*31*). Regardless of the method of transport, our data suggest that particles of upper tropospheric origin are frequently found above the tropical tropopause.

Once particles have reached the lowest regions of the stratosphere in the tropics, they are advected horizontally and vertically. Mean vertical air velocities in the stratosphere have been estimated recently from global meteorological and chemical data coupled with radiative transfer calculations (*17*) and can be derived from satellite measurements of water vapor (*32*). Vertical velocities within the lower tropical stratosphere are $\sim 2 \times 10^{-4}$ m s^{-1}, with higher velocities near the tropopause (*33*). Small particles in the lower tropical stratosphere have negligible gravitational sedimentation velocities and should also experience a net upward motion of this magnitude. Because the particles undergo significant diffusive movement, they may collide and coalesce during transport, which reduces NMR at a rate roughly proportional to the square of the particle concentration.

It has been asserted that steep gradients in the altitude profiles of particle NMRs in the tropics are inconsistent with a tropical tropospheric source for stratospheric nuclei (*34*). When the vertical axis is transposed from altitude to θ (Fig. 1), the observed average gradient in total particle NMR with increasing altitude is relatively gradual and monotonic above the tropical tropopause. We used a coagulation model to examine if this profile is consistent with coagulational depletion of particle number during ascent away from a near-tropopause source. An air parcel was initialized at 370 K (which is within the range of potential temperatures encompassing the observed tropopauses) and advected vertically from 370 to ~480 K with mean tropical velocities derived from calculations of the stratospheric residual circulation (*17*). Particle NMR was calculated as a function of θ by a detailed model of particle coagulation (*35*). Particle NMRs measured at 370 K and particle sizes estimated from the measurements were used as input, and mixing of aerosol with mid-latitude characteristics into the tropical parcel was simulated (*36*).

Nominal values of the input parameters result in particle NMRs that very closely match the median tropical profile (Fig. 3). The results are sensitive to the assumed particle diameter and to the vertical velocity profile and less sensitive to mixing of the tropical parcel with mid-latitude air.

There are several important implications of the coagulation model results. First, nucleation of particles above the troposphere is not required in the model to match the observations. The particle NMRs observed at $\theta > 475$ K (~20 km) can be explained purely by an upper tropospheric particle source and upward transport with horizontal mixing. Second, coagulation rates become very slow at the low particle NMRs found near the top of the profile: particle NMR half-lives for coagulation exceed 1 year. Thus, the particle NMR observed near 20 km in the tropics can account for the particle NMRs observed throughout the stratospheric sulfate layer, even over long transport times. Third, to accurately simulate the observed profile of particle mixing ratio, one must incorporate both horizontal mixing and the vertical variation in vertical velocity. A quantitative understanding of atmospheric motions, as well as of particle microphysics, is necessary in order to understand the spatial distribution of particle number and surface area in the stratosphere. Fourth, several statistically robust studies of stratospheric particle composition (*16*, *37*) have not found the diversity of particle compositions typical of the mid-latitude free troposphere (*16*). However, homogeneous nucleation of H_2SO_4-H_2O particles near the tropical tropopause, followed by vertical transport to the lower tropical stratosphere, would explain the relative homogeneity of stratospheric particle composition observed.

Finally, profiles of particle NMR in the mid-latitude lower stratosphere often show regions of enhanced values above the tropopause. These enhanced particle NMRs appear in the averaged mid-latitude profile in Fig. 1 as a large relative interquartile range and minor layering in the median values. Because vertical transport is extremely restricted in the lower stratosphere in mid-latitudes (*38*), the mid-latitude troposphere is unlikely to be the source of these layers. A profile measured during descent above northern California shows the magnitude of the layering (Fig. 4). The maxima in the particle NMR profile lie between the median extratropical and tropical profiles. Measure-

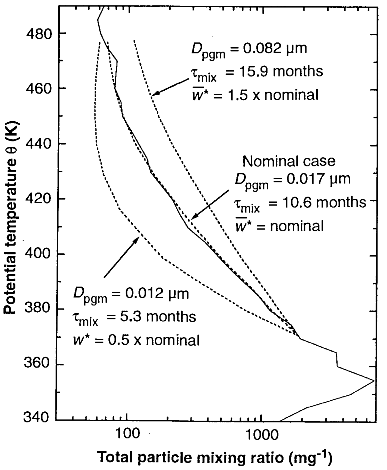

Fig. 3. Total particle mixing ratios calculated with a numerical model of particle coagulation and mixing in an air parcel ascending in the tropics. Dashed lines are model output for the nominal case and for the extreme cases from sensitivity tests. The model runs were initiated at θ = 370 K, within the range of tropical tropopause potential temperatures. The nominal residual mean vertical velocity $\bar{w}^*$ varied with altitude, falling from $\sim 8 \times 10^{-4}$ m s^{-1} at 370 K to $\sim 2 \times 10^{-4}$ m s^{-1} at 475 K (*33*). For sensitivity tests, extremes of possible geometric mass median diameter (D_{pgm}) were determined from the response characteristics of the CNC and optical particle-size spectrometers aboard the ER-2. The time scale for mixing of mid-latitude air into the tropical parcel, τ_{mix}, was estimated from tracer relationships (*41*). The profile of the tropical median total particle mixing ratio from Fig. 1 is given by the solid line.

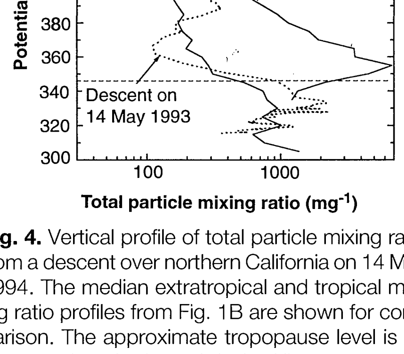

Fig. 4. Vertical profile of total particle mixing ratio from a descent over northern California on 14 May 1994. The median extratropical and tropical mixing ratio profiles from Fig. 1B are shown for comparison. The approximate tropopause level is indicated with a horizontal dashed line.

ments of gas-phase species used as tracers (*39*) indicate that the layers had been transported from the tropics. Such layered profiles are typical of the lower mid-latitude stratosphere in fall, winter, and spring (our summertime data are too few to allow general interpretation). Other analyses of tracer measurements have shown that quasi-horizontal transport from the near-tropopause region of the tropics occurs rapidly in fall, winter, and spring to the mid-latitudes between the tropopause and altitudes of at least 19 km (*38, 40*). Together, these observations indicate that particles produced in the upper tropical troposphere are frequently transported to the lower mid-latitude stratosphere, where they may significantly affect particle number and surface area mixing ratios.

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10 July 1995; accepted 17 October 1995

Transcription Against an Applied Force

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The force produced by a single molecule of *Escherichia coli* RNA polymerase during transcription was measured optically. Polymerase immobilized on a surface was used to transcribe a DNA template attached to a polystyrene bead 0.5 micrometer in diameter. The bead position was measured by interferometry while a force opposing translocation of the polymerase along the DNA was applied with an optical trap. At saturating nucleoside triphosphate concentrations, polymerase molecules stalled reversibly at a mean applied force estimated to be 14 piconewtons. This force is substantially larger than those measured for the cytoskeletal motors kinesin and myosin and exceeds mechanical loads that are estimated to oppose transcriptional elongation in vivo. The data are consistent with efficient conversion of the free energy liberated by RNA synthesis into mechanical work.

RNA polymerases play a critical role in gene expression by synthesizing RNA transcripts containing genetic information copied from DNA templates. The nascent RNA chain is elongated in a chemical reaction during which appropriate ribonucleoside triphosphates (NTPs) are condensed with the RNA 3′ end and pyrophosphate anions (PP_i) are released. RNA polymerases move along DNA while copying it, advancing on average a distance of 1 base pair (bp) (~0.34 nm along the DNA helix axis) for each nucleotide added to the transcript [(*1*) but see also (*2, 3*)]. In vivo, this movement performs mechanical work against hydrodynamic drag and other external forces (*4, 5*) and therefore requires a source of free energy. The energy is provided by the condensation reaction itself, which is energetically favorable at physiological NTP and PP_i concentrations (*1*). Thus, RNA polymerases may be viewed as molecular motors that catalyze a biosynthetic reaction while using a portion of the excess free energy from the reaction to perform mechanical work. Although translocation of RNA polymerase molecules along DNA has been detected by microscopy (*6–8*), little is known about the ability of these enzymes to move against opposing forces or about their energy conversion effi-

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