JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 104, NO. D19, PAGES 23,935–23,940, OCTOBER 20, 1999

# Particle production in the lowermost stratosphere by convective lifting of the tropopause

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**Abstract.** Aircraft measurements of aerosol particles and trace gases were performed in the upper free troposphere and lower stratosphere during the Stratosphere and Troposphere Experiment by Aircraft Measurements (STREAM-96) campaign from Shannon airport, Ireland. During one measurement flight, ultrafine particle number densities up to $10^4$ cm$^{-3}$ (STP) were observed in the lowermost stratosphere. Concurrent with these very high number densities of ultrafine particles, high accumulation mode particle number densities were observed over the same geographical location in the free troposphere, which were attributed to convective transport in the troposphere. The observations suggest that adiabatic cooling of the stratospheric air, as a result of the convective transport in the troposphere that lifted the tropopause and the air in the lowermost stratosphere, was responsible for triggering the formation of new particles. However, also aircraft emissions could have contributed to the enhancement in ultrafine particles.

## 1. Introduction

Currently, much research is performed to better understand the abundance of particles and their formation in the atmosphere. A main reason for this is the potential impact of atmospheric aerosols on climate, that is, via scattering of sunlight or, indirectly, via the interaction with clouds [*Intergovernmental Panel on Climate Change (IPCC)*, 1996]. Moreover, aerosols provide a reactive surface for heterogeneous reactions, which can affect oxidation processes in the troposphere and stratosphere [*World Meterological Organization (WMO)*, 1994].

The processes controlling new particle formation are still not well understood. Homogeneous binary nucleation of sulfuric acid and water vapor is commonly assumed to be a main production mechanism for new particles. However, evidence is growing that other gases also play a role in the nucleation process, for example, ammonium chloride [*Korhonen et al.*, 1997] and ammonia [*Weber et al.*, 1996]. Dynamical processes are also important for new particle formation, as pointed out by *Bigg* [1977]. He proposed that mixing of air masses can enhance new particle formation, for example, through a local reduction of the saturation water vapor pressure. This is supported by model calculations by *Nilsson and Kulmala* [1998].

Several events of particle formation connected to convective clouds have been reported as well. *Perry and Hobbs* [1994], for example, found two regions with high ultrafine particle number concentrations in the vicinity of a marine cumulus cloud, one above the cloud top and one downwind of the cloud near the level of the anvil outflow. They explain particle formation in these regions by the high relative humidity and the low preexisting particle number concentration in the air detraining from the cloud. Both conditions are favorable for new particle formation. Similar observations were made by *Hegg et al.* [1990] near the tops of marine stratiform clouds. They, however, also showed one case where the enhanced Aitken particle layer was separated from the cloud, which they explained by advection of continental air. In this study we present observations of high number densities of ultrafine particles in the lowermost stratosphere, which are proposed to be associated with convective activity in the troposphere.

## 2. Experiment

During the Stratosphere and Troposphere Experiment by Aircraft Measurements (STREAM-96) campaign, six flights were conducted in the free troposphere and lower stratosphere from Shannon airport, Ireland. Instruments for in situ measurements of aerosols and trace gases were installed on a Cessna Citation II twinjet aircraft, operated by the Technical University of Delft, Netherlands. This study focuses on the particle measurements performed on May 22, 1996, over the Atlantic Ocean close to the west coast of Ireland, since this was the only flight where indications for new particle formation were observed in the lower stratosphere. A selection of the measured trace gases is used for the interpretation of the data, namely, ozone, carbon dioxide, and water vapor. On this day a vertical "stack flight," composed of five isobaric flight legs in the free troposphere and lower stratosphere, was carried out between 1400 and 1800 UTC. The flight pattern is shown in Figure 1. The flight section that we focus on covered a region of 6 km in altitude (between 6.5 and 12.5 km altitude), 2° in latitude (~220 km) and 1.5° in longitude (~135 km). Horizontal flight tracks were flown at 428, 315, 262, 216, and 178 hPa, respectively.

The flight was performed about 6 to 10 hours after the passage of a cold front, which was located 200 km south of Ireland at 1200 UTC. A low-pressure center associated with

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Paper number 1999JD900774.
0148-0227/99/1999JD900774$09.00

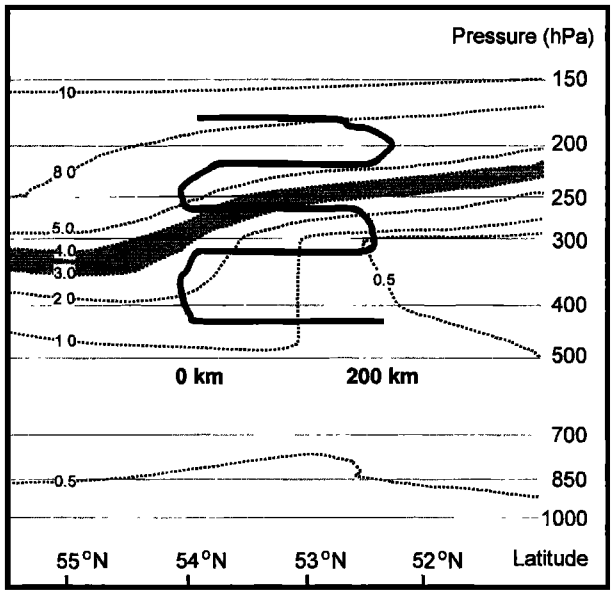


**Figure 1.** Flight track (thick solid line) of the measurement flight on May 22, 1996. The thin dotted lines represent the potential vorticity, and the gray area denotes the tropopause region, defined by a potential vorticity range of 3–4 potential vorticity units (PVU = K $kg^{-1}$ $m^2$ $s^{-1}$). The distance to the reference point used in Figures 2, 3, and 4 is also indicated.

this front, located 450 km northwest of Ireland, and a high-pressure ridge extending from the Azores in over France, generated a westerly flow at all levels over the measurement area. The northern parts of the tropospheric flight levels were conducted in a haze or thin cirrus, which appeared brownish in some viewing angles. Optically dense cirrus was only observed as patches in the beginning of the flight.

## 3. Instrumentation

The instrumentation used to measure the different species is briefly described below. The aerosol number density was measured by two TSI 3010 condensation particle counters (CPC), one standard and one modified version, in which the temperature difference between the saturator and condenser was increased by about 8°. Laboratory calibration using ammonium sulfate particles showed that the 50% counting efficiencies of the instruments were 0.015 and 0.006 $\mu$m diameter, respectively. The sample flow rates were controlled by critical orifices and were 1.0 L/min. The CPC data were logged five times per second. The number density of ultrafine particles (UCN) was obtained by subtracting the particle number density measured by the two CPC instruments, resulting in a size interval between 0.006 and 0.015 $\mu$m diameter.

Particles larger than 0.12 $\mu$m diameter were measured every second with an optical particle counter (OPC) particle measuring system (PMS) passive cavity aerosol spectrometer probe (PCASP). The aerosol was sampled through a ¼ inch stainless steel inlet pointing 180° opposite to the flight direction, only allowing particles smaller than about 1 $\mu$m to be sampled [*Schröder and Ström*, 1997]. Hence particles measured with this instrument, also referred to as accumulation mode particles or Nopc, have a diameter between 0.12 and 1 $\mu$m.

All particle number densities have been normalized to standard temperature and pressure (STP, 0°C, 1 atm). The uncertainty in the measured particle number density is dependent on counting statistics and equal to the square root of the number of particles counted in a unit time. Hence the uncertainty increases with decreasing particle number density.

There is some concern that the counting efficiency of the condensation particle counters may change at reduced pressure. In a laboratory experiment, *Noone and Hansson* [1990] concluded that the counting efficiency of a TSI 3760 condensation particle counter was unaffected at pressures greater than about 200–250 hPa for particles with a diameter between 0.02 and 0.1 $\mu$m. Another study by *Zhang and Liu* [1990] showed that the counting efficiency for a TSI 3020, operating at standard flow rate, remains unaffected to a pressure of about 100 hPa. Since the top flight levels of the flight discussed in this paper are conducted at a pressure of about 200 hPa, we cannot exclude that the particle measurements are affected by the reduced pressure in the instruments. A change in the counting efficiency due to reduced pressure would cause an underestimation of the ultrafine particle number density at high altitudes.

Water vapor measurements were performed using a Lyman-$\alpha$ photofragment fluorescence technique [*Zöger et al.*, 1999], which has an accuracy typically better than 4% and a precision of 0.2 ppmv for water vapor mixing ratios of 3 ppmv. Ozone was measured by a modified pressure independent chemiluminescence monitor (Bendix 8002) [*Bregman et al.*, 1995], and $CO_2$ was measured with a nondispersive infrared gas analyser (LI-Cor 6262), with uncertainties of 7% and 0.4 ppmv respectively. All gas phase measurements were performed with a time resolution of 1 s.

## 4. Observations

To be able to directly compare the measurements at the different flight levels, the distance from a reference point has been calculated for each data point. In Figures 2a–2e, ultrafine and accumulation mode particle number densities (UCN, Nopc) are shown as a function of this distance for the different flight levels. Accordingly, Figures 3a–3e show the ozone, water vapor, and carbon dioxide mixing ratio, and Figures 4a–4e show the potential temperature and the relative humidity with respect to ice.

The ozone mixing ratio varies from 60 ppbv at the lowest levels at 400 ppbv at the top flight level. In a previous study using the STREAM-96 data the ozone mixing ratio was used to discriminate between tropospheric and stratospheric air [*de Reus et al.*, 1998]. Tropospheric air was characterized by ozone mixing ratios lower than 200 ppbv, and stratospheric air was characterized by mixing ratios exceeding 300 ppbv. The tropopause region, with an ozone mixing ratio between 200 and 300 ppbv, was regarded as a transition zone, experiencing substantial mixing of tropospheric and stratospheric air masses. Using the same criteria for this flight shows that the lower three flight levels were performed in the troposphere, whereas the two top levels where performed in the stratosphere. This is also supported by the very low water vapor mixing ratio observed on these upper two levels.

Accumulation mode particle number densities up to 3000 $cm^{-3}$ were found in the upper troposphere, while number densities of about 50 $cm^{-3}$ were observed in the lowermost stratosphere. Note that these values are normalized to standard temperature and pressure (STP). Ultrafine particles were observed episodically on all flight levels, except for the highest

level. Peak number densities of $10^4$ cm$^{-3}$ were observed on the lower stratospheric level (216 hPa), while the number densities were below 700 cm$^{-3}$ on the tropospheric flight levels. Note that the lowest flight level in this study is at 6.5 km altitude, so that the particle number densities presented here are only representative for the upper troposphere.

On the lowest flight level (428 hPa) a maximum accumulation mode particle number density was detected near 100 and 250 km from the reference point, respectively. This coincided with a maximum $H_2O$ and $CO_2$, and a minimum $O_3$ mixing ratio. A small enhancement of the ultrafine particle number density was also observed on this level at a distance of about 50 km, where the Nopc had its lowest value.

A large transition in Nopc and UCN was observed on the 315 and 262 hPa flight levels. On these levels the accumulation mode particle number density remained high from the reference point and dropped quickly to lower values after 100 to 125 km. The ultrafine particle number density showed an opposite trend, with high values beyond a distance of 125 km. Similar (anticorrelated) correlated trends were observed in the mixing ratios of the different trace gases (see Figure 3). From these observations we can distinguish two different regions on these flight levels. The first part, between 10 and 100 km, is characterized by low $O_3$ and UCN densities and high $CO_2$, $H_2O$, and Nopc densities. The second part (125–190 km) shows the opposite features. Even on a smaller scale, fluctuations in water vapor and carbon dioxide concentration correlated well with the accumulation mode particle number density, while ozone showed an anticorrelation. This indicates vertical redistribution of the air in the troposphere. The combination of high $H_2O$, $CO_2$, and Nopc, and low $O_3$ concentrations indicates that the air has been transported upward, whereas low $H_2O$, $CO_2$, and Nopc and high $O_3$ concentrations indicate downward transport relative to the flight level.

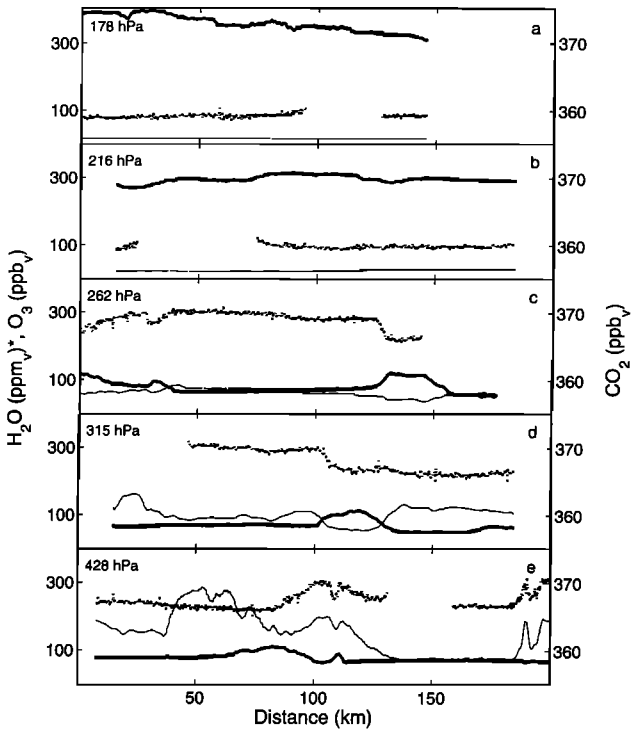


**Figure 3.** Ozone (thick solid line), water vapor (thin solid line), and $CO_2$ mixing ratios (dotted line) at the (a) 178, (b) 216, (c) 262, (d) 315, and (e) 428 hPa levels. Missing $CO_2$ data are associated with calibration of the instrument. For clarity reasons, the $H_2O$ mixing ratio is multiplied by a factor 5 in the upper two panels (178 and 216 hPa).

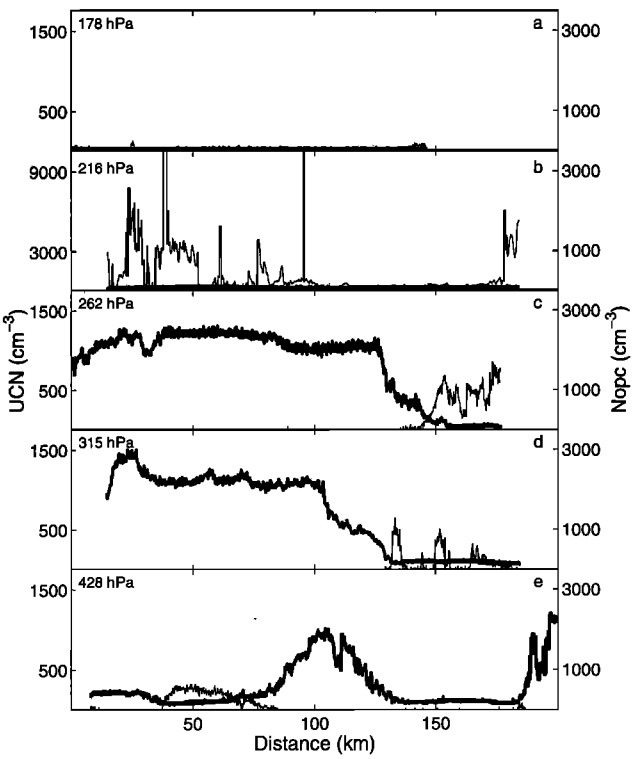


**Figure 2.** Ultrafine (thin solid line) and accumulation mode (thick solid line) particle number densities at the (a) 178, (b) 216, (c) 262, (d) 315, and (e) 428 hPa levels. Note the different scale in Figure 2b.

A division of the flight leg into two parts is also discernible from the relative humidity (RH) and potential temperature (0) on the lower of the two stratospheric levels (216 hPa). However, the most noticeable difference appears in the ultrafine particle number density, which reaches values exceeding $10^4$ cm$^{-3}$ in the first part of the track, while being close to zero on the second part. Note that these densities are more than 1 order of magnitude higher than UCN densities observed on the tropospheric flight levels on the same day and more than 2 orders of magnitude higher than the particle number densities measured in the lower stratosphere during the rest of the campaign. The different trace gases showed little variation over the entire flight track.

At the 178 hPa level all measured variables show little variation. The mean values of all variables, as mentioned above, at both parts of the tracks are listed in Table 1. The first part of the flight track (10–100 km) is labeled 1, and the second part (125–190 km) is labeled 2.

## 5. Discussion

Vertical transport is the only plausible mechanism that can explain the presence of high number densities of accumulation mode particles in the free troposphere. The vertical coherence in the data further suggests that this vertical transport was caused by convection as opposed to large-scale lifting. This is supported by the observation of thin cirrus and "pancake-

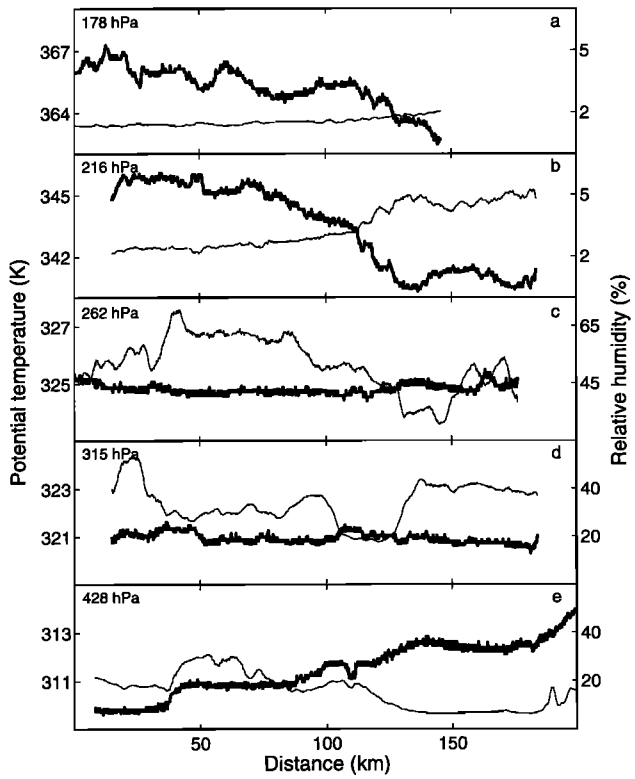


**Figure 4.** Potential temperature (thick solid line) and relative humidity with respect to ice (thin solid line) at (a) 178, (b) 216, (c) 262, (d) 315, and (e) 428 hPa levels.

shaped" low-level clouds during the flight, which possibly are remains of convective cumulus clouds. Moreover, satellite images show an area with convective activity behind the cold front which passed the measurement area 6–10 hours prior to the measurement flight.

The observation of high ultrafine particle number densities in the stratosphere, geographically located above the region with enhanced accumulation mode particles in the troposphere, leads to the following hypothesis regarding the formation of the UCN particles.

A cold front passed the sampling area about 6–10 hours before the measurements, and in the unstable air behind this front convective clouds were formed. The convective energy of these clouds was sufficient to lift boundary layer air up to the tropopause, but not sufficient to penetrate it. However, the vertical motion caused the tropopause to bulge upward, which forced the air in the stratosphere to ascend and cool adiabatically. This rapid cooling caused aerosol precursor gases to become supersaturated and to nucleate new particles. Below we discuss the data in support of the hypothesized mechanism of new particle formation in the lowermost stratosphere.

Analogous to the formation of cloud droplets, new particle formation is a result of the unstable conditions that occur when condensable species exceed the saturation vapor pressure. This unstable state is favored by an increase in the amount of precursor gases or a change in the environmental conditions. In the free troposphere and lower stratosphere the major mechanism for new particle formation is thought to be binary homogeneous nucleation of sulfuric acid and water vapor. Sulfuric acid is mainly formed by a chemical reaction of $SO_2$ and OH. The major sink is condensation of sulfuric acid and water vapor on the surface of preexisting particles. Hence a low surface area (i.e., few and small particles), high humidity, and high precursor gas concentrations (OH and $SO_2$) are conditions that favor this type of particle formation. As a consequence of the nonlinear relation between the nucleation rate and temperature, relative humidity, and sulfuric acid concentration, only small fluctuations in these parameters are needed to cause large changes in particle production rates. *Easter and Peters* [1994] calculated that the nucleation rate increases by approximately 1 order of magnitude for each 2°–3°C decrease in temperature, 6–8% increase in relative humidity, or 20–30% increase in $H_2SO_4$ vapor concentration in the troposphere. Hence a 100 to 200 m vertical perturbation of the tropopause could result in a significant increase in the nucleation rate due to adiabatic cooling.

Since the potential temperature ($\theta$) normally increases with altitude, a concurrent decrease in $\theta$ and an increase in relative humidity may indicate ascending air. Using this relationship, the data in Figure 4b suggest a subsiding air motion at the position where the enhanced UCN number density was observed, which apparently contradicts the hypothesis. However, it is quite likely that the upward motion that is proposed to initiate the particle formation occurred prior to the measure-

**Table 1.** Mean Values of Particle and Trace Gas Concentrations and Meteorological Data for the Measurement Flight on May 22, 1996, From Shannon Airport, Ireland

| | 428 hPa | | 315 hPa | | 262 hPa | | 216 hPa | | 178 hPa | |
|---|---|---|---|---|---|---|---|---|---|---|
| | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 |
| UCN, STP $cm^{-3}$ | 86 | 3 | 0 | 90 | 0 | 293 | 1895 | 758 | 24 | 39 |
| Nopc, STP $cm^{-3}$ | 466 | 310 | 2289 | 209 | 2302 | 447 | 73 | 62 | 58 | 48 |
| $N_6$, STP $cm^{-3}$ | 1028 | 696 | 2096 | 975 | 2100 | 1464 | 2983 | 1255 | 109 | 153 |
| $O_3$, ppbv | 87 | 72 | 69 | 58 | 73 | 84 | 294 | 295 | 369 | 324 |
| $H_2O$, ppmv | 196 | 80 | 104 | 114 | 71 | 51 | 5 | 6 | 3 | 4 |
| $CO_2$ ppbv | 367 | 367 | 370 | 366 | 370 | 367 | 360 | 360 | 359 | 359 |
| Potential temperature, K | 310 | 312 | 321 | 321 | 324 | 325 | 345 | 341 | 366 | 363 |
| Temperature, °C | −30 | −28 | −43 | −43 | −52 | −52 | −50 | −53 | −50 | −51 |
| Wind direction, deg | 250 | 267 | 264 | 269 | 269 | 273 | 249 | 258 | 260 | 265 |
| Wind speed, m $s^{-1}$ | 20 | 29 | 37 | 41 | 39 | 42 | 25 | 28 | 18 | 22 |

The number 1 is referring to the first part of the flight track (10–100 km), and 2 is referring to the second part (125–190 km). UCN, Nopc, and $N_6$ are the particle number densities of particles with a diameter between 0.006 and 0.015 $\mu$m, 0.12 and 1 $\mu$m, and 0.006 and 1 $\mu$m, respectively.

ments and that, meanwhile, the air has descended again to its original position.

A different mechanism to explain the presence of enhanced particle number densities in the vicinity of clouds was proposed by *Perry and Hobbs* [1994]. On the basis of measurements near marine cumulus clouds, they suggested that droplets detrained from the clouds can humidify the air at the edges, and due to the relatively low particle surface area $H_2SO_4$ and $H_2O$ can nucleate new particles. A signature consistent with this mechanism is seen on the 315 and 262 hPa flight levels (Figures 2c and 2d). Ultrafine particles were observed where the number of densities of accumulation mode particles were low, together with a small enhancement in relative humidity. At the 216 hPa level, however, the increase in UCN did not have a coherent drop in Nopc, which was consistently low along the entire flight track.

Moreover, the observations on the 216 hPa level show no evidence of recent intrusions of or mixing with tropospheric air, which could explain the formation of new particles on that level [*de Reus et al.*, 1998]. The relatively low $H_2O$ and $CO_2$ mixing ratios and high $O_3$ mixing ratio leave no doubt that this air is indeed stratospheric.

Calculated back trajectories show that the air mass originated from the northern part of Canada 5 days prior to arriving in the measurement area (P. Siegmund, Royal Netherlands Meteorological Institute (KNMI), personal communication, 1996). All trajectories calculated for different points along the 216 hPa flight track show the same origin. Hence the enhancement of ultrafine particles on the 216 hPa flight level cannot simply be explained by a difference in air mass origin.

The fact that the region west of Ireland is located near the North Atlantic flight corridor makes aircraft exhaust a potential source for small particles in the stratosphere. A good tracer for aircraft emissions is the $NO_y$ mixing ratio, which was also measured during the STREAM-96 experiment. Unfortunately, no $NO_y$ data are available between 30 and 90 km at the 216 hPa level, due to calibration of the instrument. At a distance of about 25 and 180 km, local maxima in the $NO_y$ mixing ratio at this level coincide with an enhancement in the ultrafine particle number density (H. Fisher, Max Planck Institute for Chemistry, Mainz, personal communication, 1996). This suggests that at least some of the enhanced UCN could be explained by aircraft emissions. For instance, the very narrow peak close to a distance of 100 km at the 216 hPa level coincided with the observation of an aircraft, which flight track was crossed about 3 min after the passage of the aircraft. However, it would require a large number of aircraft to explain enhancement in UCN. This is illustrated in the following section.

The total particle number density ($N_6$) at the 216 hPa level shows a similar pattern as the ultrafine particle number density shown in Figure 2b; however, it never drops below 300 $cm^{-3}$. A baseline can be drawn through the minimum observed $N_6$ particle number density, which can be considered as the background $N_6$ number density, in case no enhancement in particle number density would have occurred. This baseline density ranges from 305 $cm^{-3}$ at the beginning of the flight track to 365 $cm^{-3}$ at the end, which is in good agreement with the particle number densities observed in the lower stratosphere during the rest of the campaign [*de Reus et al.*, 1998]. Subtracting the baseline density from the observed particle number density gives the particle enhancement, which we try to relate to aircraft emissions below.

The fuel flow for a wide body aircraft is about 10 t $h^{-1}$, and the emission index is $10^{16}$ particles $kg^{-1}$ fuel [*Schlager et al.*, 1997]. The typical aircraft speed is about 900 km $h^{-1}$, and the plume dispersion rate is 250 $m^2$ $s^{-1}$ [*Gierens*, 1996]. This results in a plume strength of $1.1 \times 10^{14}$ particles $m^{-1}$.

Owing to aircraft separation, it is not likely that any of the plumes that were presumably crossed were younger than 2 min. This is also supported by the fact that no turbulence was reported by the observers on board of the measurement aircraft and only one aircraft was visually observed. After 2 min, the plume cross section has increased to about 30,000 $m^2$, which gives a particle enhancement due to one aircraft of 3700 $cm^{-3}$. It takes the measurment aircraft about 1 s to fly through this plume. Assuming that all air traffic took place close to the 216 hPa level and at the same time, the number of aircraft that is needed to generate the observed particle enhancement can be calculated by dividing the observed particle enhancement for each data second by 3700 $cm^{-3}$ and subsequently adding all the fractions together. Note that the particle number density has to be corrected for ambient temperature and pressure. This results in a total number of 130 aircraft, flying within 2 min prior to the measurement flight, which is necessary to explain the observed particle enhancement at the 216 hPa level.

After 6–7 min, the plume cross section is about 100,000 $m^2$, which takes the measurement aircraft 2 s to fly through. Each aircraft contributes then with 1100 $cm^{-3}$ to the ambient particle number density, and a total of 225 aircraft is necessary to explain the observed particle enhancement. Similar calculations show that for a plume of 10–15 min old, 300 aircraft are needed to explain the observed particle enhancement and for a plume of 15–30 min old, 350 aircraft are necessary. All calculations are performed assuming that the measurement flight was performed perpendicular to the flight track of the commercial aircraft. A sharper angle between the two flight tracks would decrease the estimated number of aircraft, since a longer time is spent in the plume. For a 30° angle the time in the plume will be approximately doubled, and hence the number of aircraft will be halved, which still results in a large number of aircraft.

Moreover, measurements performed in the North Atlantic flight corridor show much lower particle number densities as observed during this flight. *Schlager et al.* [1997] presented total particle number densities ($Dp >$ nm) up to 600 $cm^{-3}$. They attributed part of the particle peaks to 22 aircraft flying in the corridor. We observed total particle number densities ($Dp >$ 6 nm) up to 2500 $cm^{-3}$ (at ambient temperature and pressure) which is a factor 4 more than observed by *Schlager et al.* [1997]. Furthermore, no indications for aircraft exhaust were found at the 262 hPa level, which is also a common flight level for commercial aircraft. Hence we do not believe that the North Atlantic flight corridor influenced our measurements to a degree that aircraft emissions only could explain the enhanced UCN number densities in the stratosphere. Even though particles from aircraft cannot explain all UCN observed, the accumulation of precursor gases from aircraft exhaust can potentially play a role in new particle formation in the stratosphere when triggered by some other mechanism.

## 6. Summary and Conclusion

Observations in the lowermost stratosphere show an almost 100 km wide region of enhanced number densities of small aerosol particles; number densities typically exceeded $10^3$

$cm^{-3}$ (STP) for extended periods. Observations of accumulation mode particles and trace gases in the troposphere give evidence of recent vertical transport b convective clouds. The occurrence of the ultrafine particles in the stratosphere located over the same geographical position as the convective motion in the free troposphere suggests that adiabatic lifting of stratospheric air, induced by the convective clouds in the troposphere, triggered the formation of new particles in the lowermost stratosphere. Although we concluded that aircraft emissions only cannot explain the observed ultrafine particle number density, they could have contributed to the enhanced particle number density and the total load of precursor gases in the lower stratosphere.

Since there are only few reports in the literature about aerosol measurements in the lowermost stratosphere, the general nature of this phenomenon is difficult to assess. However, this rather spectacular observation adds to our knowledge about the aerosol life cycle and improves our understanding about new particle formation in the lowermost stratosphere.

**Acknowledgments.** Special thanks to the Citation Team and the crew from the Technical University of Delft for the excellent collaboration during the campaign. The STREAM project is supported by the European Union (DG XII). Support by the Swedish Natural Science Research Council (NFR) is also gratefully acknowledged. Thanks also to the anonymous reviewers who with their constructive criticism and suggestions helped in improving the manuscript.

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(Received December 31, 1998; revised July 16, 1999;
accepted July 21, 1999.)