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Atmospheric Chemistry and Physics Discussions

# When does new particle formation not occur in the upper troposphere?

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## Abstract

Recent aircraft studies showed that new particle formation is very active in the free troposphere and lower stratosphere. And, these observations lead to a new question: when does new particle formation *not* occur? Here, we provide case studies to show how convection and surface area affect new particle formation in the upper troposphere, using the measured aerosol size distributions during the NSF/NCAR GV Progressive Science Missions in December 2005. There were ten research flights, including three days of nighttime experiments, at latitudes from 18 to 52° N and altitudes up to 14 km. About 78% of the total samples showed the new particle formation feature with number concentrations of particles with diameters from 4 to 9 nm, 670±1270 $cm^{-3}$, and the total particle number concentrations with diameters from 4 to 2000 nm, 920±1470 $cm^{-3}$. Our case studies show that new particle formation was closely associated with convection and low surface areas of preexisting aerosol particles (<4 $\mu m^2 cm^{-3}$). On the other hand, for the cases where no new particle formation events were observed, air masses usually did not experience a vertical motion and air often originated from either the upper troposphere or lower stratosphere where precursor concentrations are relatively low; in addition, it was also a general trend that non-event cases also had higher surface areas (~16 $\mu m^2 cm^{-3}$). These observations are consistent with other observations during the Progressive Science Missions (Young et al., 2007). Because of the lower temperatures in this region (T<250 K), nucleation is thermodynamically favorable; but because of low aerosol precursor concentrations, nucleation is sensitive to aerosol precursor concentration and surface area. Under such conditions, convection (which brings higher concentrations of aerosol precursors and water vapor to higher altitudes) and low surface area play critical roles on whether new particle formation takes place or not. Latitude dependence of new particles also shows higher particle concentrations in the midlatitude tropopause region than in the subtropics, consistent with Hermann et al. (2003).

## 1 Introduction

Recent aircraft studies showed new particle formation in the free troposphere and lower stratosphere (de Reus et al., 1998, 1999; Nyeki et al., 1999; Twohy et al., 2002; Lee et al., 2003; Young et al., 2007) with high frequencies (up to 86–100%) (Young et al., 2007) and strong magnitudes (up to 45 000 $cm^{-3}$) (Twohy et al., 2002). Hermann et al. (2003) have provided so far the most comprehensive statistical analysis of new particle formation in the northern hemisphere tropopause region from three-year aircraft measurements; elevated particle number concentrations of 1500–8000 $km^{-3}$ were frequently observed in a wide range of latitudes (5° N–50° N). Twohy et al. (2002) showed especially high number concentrations of new particles up to 45 000 $cm^{-3}$ in the mid-latitudes, associated with deep convection. Kulmala et al. (2006)'s predictions also suggested that deep convection can bring insoluble organic trace gases to higher altitudes to produce new particles. Minikin et al. (2003)'s aircraft studies also showed relatively high concentrations of Aitken mode particles (up to 1000 $cm^{-3}$) even in the southern hemisphere, where the anthropogenic emission of $SO_2$ is much lower than in the northern hemisphere; their comparison of particle number concentrations in the northern and southern hemisphere indicated that new particles were directly related to aerosol precursor sources. New particle formation events take place near or in orographic clouds during the nighttime (Wiedensohler el al., 1997; Mertes et al., 2005) and even in cirrus clouds (Lee et al., 2003). As new particle formation was observed in a wide range of the free troposphere and lower stratosphere (Ström et al., 1999; Twohy et al., 2002; Hermann et al., 2003; Minikin et al., 2003; Lee et al., 2003, 2004; Young et al., 2007), it is also important to understand when particle formation *does not* occur. This study attempts to address this important atmospheric nucleation question.

We present results from new particle formation studies during the National Science Foundation (NSF) and National Center for Atmospheric Research (NCAR) NSF/NCAR GV Progressive Science Missions. The GV is also known as HIAPER, the High-performance Instrumented Airborne Platform for Environmental Research. The Pro-



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gressive Science Mission was the first science mission with the GV; there were 10 research flights (Young et al., 2007). There is the Part I paper by Young et al. (2007) that used two days of measurements in the midlaltitude tropopause region (on 1 and 7 December 2005) from this mission to show how stratosphere and troposphere air mixing enhances aerosol new particle formation. The present study is the Part II paper, focusing on the rest of the measurements from the same mission. Here, we show the effects of vertical motion and surface area on new particle formation in the upper troposphere and try to understand when new particle formation does not occur. We also have a third manuscript (S.-H. Lee et al., 2007[1]) which included nighttime new particles formation observed from GV.

## 2 NSF/NCAR GV progressive science missions

The NSF/NCAR GV Progressive Science Missions took place from 21 November to 19 December in 2005 in Broomfield, Colorado. The flights covered the western half of the United States, and parts of Canada and Mexico in latitude from 18 to 62° N and in longitude from 92 to 130° W. There were three days of nighttime experiments (2, 12 and 19 December 2005) in order to investigate the effects of sun exposure and the latitude and altitude dependence of new particles. Nighttime studies in this region are rare. The GV flew along the same flight track before and after sunrise (or sunset) over an 8-h period. There were also another seven days of science flights which were mostly made in the mid-latitude tropopause region.

Aerosol sizes and concentrations were measured with the University of Denver nuclei mode aerosol size spectrometer (NMASS) and focused cavity aerosol spectrometer (FCAS). These instruments are described in detail elsewhere (Jonsson et al., 1995; Brock et al., 2000; Lee et al., 2003, 2004; Young et al., 2007) and have been used for

[1]Lee, S.-H., Young, L.-H., Benson, D. R., et al.: Observations of nighttime new particles formation in the troposphere, J. Geophys. Res., submitted, 2007.

new particle formation studies in the upper troposphere and lower stratosphere previously (Lee et al., 2003, 2004; Young et al., 2007). Briefly, NMASS has five condensation nucleus counters that measure cumulative number concentrations of aerosols larger than 4, 8, 15, 30 and 60 nm, respectively. FCAS is a light scattering instrument and sizes aerosols from 90 to 2000 nm. Using an inversion algorithm, size distributions from 4 to 2000 nm are obtained. The inversion also includes sampling efficiency, anisokinetic inlet effects, and diffusion loss etc. The criteria for new particle formation are (i) concentrations of particles from 4 to 9 nm ($N_{4-9}$)>1 $cm^{-3}$, (ii) more than 15% of total particles with the diameter 4–2000 nm ($N_{4-2000}$) are $N_{4-9}$, and (iii) particles from 4 to 6 nm ($N_{4-6}$) are higher than those from 6 to 9 nm ($N_{6-9}$) (Young et al., 2007). Each new particle formation event is further classified as either a strong or weak event in the present study to better understand the condition in which new particle formation is not active. For a strong event the conditions are (1) $N_{4-9}$ and $N_{4-2000}$>500 $cm^{-3}$ and (2) a size distribution in which there are three modes present with peaks at <10, ~20 and 60–200 nm (similar to Young et al., 2007) and in which the smallest mode has a number concentration at least one order of magnitude higher than the other two peaks. For a weak event the requirements are (1) $N_{4-9}$ and $N_{4-2000}$<500 $cm^{-3}$ and (2) a size distribution in which there are three modes similar to what was mentioned above, the main difference being that all three peaks are on the same order of magnitude.

## 3 Results and discussion

Table 1 summarizes the measured particle concentrations and meteorological conditions during this mission, including the measured aerosol number concentrations from 4 to 9 nm ($N_{4-9}$), the total measured aerosol number concentration ($N_{4-2000}$), surface area density of preexisting aerosols, temperature, relative humidity over ice (RHI), the potential temperature ($\theta$), water mixing ratio, and altitude, along with the fraction of samples that satisfy the new particle formation criteria. A large fraction of the total particles were in the size range from 4 to 9 nm (71% on average), indicating that new



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particle formation is active in this region. However, for those non-event samples (22% of all samples), surface area densities were much higher (∼16 $\mu$m$^2$cm$^{-3}$, much higher than the average surface area in this region, which was only ∼4.7 $\mu$m$^2$ cm$^{-3}$, Table 1), a clear indication that low surface area is necessary for new particle formation, consistent with Lovejoy (2004)'s ion-induced nucleation predictions in this region.

Our results from the NSF/NCAR GV Progressive Science Missions show new particle formation was often closely associated with low surface area of preexisting aerosol particles and convection. In the present study, convection is referred as to the cases when the air mass is lifted over 3 km from a lower source altitude, usually <7 km within 72 h, based on the HYSPLIT trajectory outputs for altitude dependence with time. On the other hand, however, if it took longer than 72 hours for such lift, we considered such a case as a non-convection event. And, our results show that when the air did not experience strong vertical motion even if there were low surface areas (e.g., <1 $\mu$m$^2$ cm$^{-3}$, Fig. 8a), there were no events. To understand how these factors affect new particle formation, we analyzed several strong new particle formation events and non-events from three days of measurements (2, 12, and 19 December 2005), by combining with the backward trajectory calculations from the NOAA HYSPLIT models (Draxler and Rolph, 2003). Because the progressive science mission was the first science mission on the NSF/NCAR GV, there were a limited number of tracers measured onboard (ozone, CO and water vapor, etc.) without chemical analysis for aerosol particles. Although our qualitative analysis does not provide in-situ information on vertical motion, they still provide important insights into better understanding the conditions where new particle formation does not take place. Some of these strong events also took place during the nighttime (Lee et al., 2007[1]).

Case study: sunset experiment (20051202)

Figure 1 shows the data taken during the sunset experiments on December 2, 2005, showing a strong new particle formation event which occurred at night. This figure shows (a) the measured $N_{4-9}$, $N_{4-2000}$, RHI, temperature and surface area as a function of time, (b) the measured average aerosol size distribution for the air mass corresponding to this event, and (c) the estimated solar flux, rainfall (which is the amount of precipitation occurring in a given time frame taken from the HYSPLIT trajectory calculations, expressed in the unit of mm $h^{-1}$) and relative humidity (RH) for the air mass from the previous five days derived from the HYSPLIT trajectory calculations, as a function of preceding hours of air mass. Figure 2 shows a similar set of data for a weaker new particle formation event which occurred during the daytime. There are substantial differences in the number concentrations and size distributions between the weak and strong event identified with the criteria listed above (Sect. 2). Interestingly, there was not much difference for the surface area or the RHI between the two events (Figs. 1a and 2a), and for both cases the values were lower than the average values for this region (Table 1). Also, while the solar flux was similar for both cases (Figs. 1c and 2c), the RH calculated from trajectory calculations for the strong event was consistently higher (>30% on average) from the previous five days compared to the weak event, in which RH is nearly 0% (Figs. 1c and 2c).

The distinctive difference between the two events, however, is the back trajectory from the previous five days (Fig. 3). These trajectories show two main differences. The first is the altitude that the air masses come from. For the strong new particle formation event, the air mass originated from a much lower altitude (6.5 km) three days prior to the event, whereas the air was in the upper troposphere (12 km) for the past 5 proceeding days for the weak new particle formation event, an indication of clear vertical motion. Such a difference suggests that the air mass from the strong new particle formation event underwent a significant extent of vertical motion, and thus brought higher concentrations of the aerosol precursors from lower altitudes to aid in new particle formation at higher altitudes with lower temperatures. This also implies that air mixing can take place during abrupt convection. It has been shown that the nucleation rate can be increased one order of magnitude with a temperature decrease of 2–3 K (Nilsson et al., 2000), and it is also possible that such air mixing also contributed to strong new particle formation events. However, this is not the case for the weak new particle



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formation event. Also, since the air masses came from two different origins (Fig. 3), it is possible that they had different aerosol precursor concentrations, although for the weak case the air mass was mostly in the upper troposphere, so the air mass origin can be less important in the case of non-convection.

## Case study: sunrise experiment (20051212)

Figures 4 and 5 show graphs for a strong and weak new particle formation event for the sunrise experiments taken on 12 December 2005. The strong event occurred before sunrise (nighttime) while the weak event occurred after sunrise during the day. Like the previous case study, most of the parameters (T, RHI and surface area density) were similar between the two events (Figs. 4a and 5a), but the major differences comes in the back trajectory data (Figs. 4c, 5c and 6) which suggests that air mass history is an important parameter that determines new particle formation event strength. The stronger event had clear evidence of vertical convection, which was absent from the weak event (Fig. 6). Also, similarly to the previous case study (2 December 2005), the origin for the air mass involved in the weak event came from a polluted continental environment, whereas the strong event was evolved from an air mass that originated from a clean marine atmosphere. For the strong event, RH (90%) and rainfall (1.5 mm $h^{-1}$ on average) were much higher for the previous five days than those for the weak event (0% and 0.1 mm $h^{-1}$ on average). Higher rainfall implies a greater scavenging of preexisting particles and thus less surface area for nuclei mode particles to condense on.

## Case study: sunrise experiment (20051219)

Figures 7 and 8 show another sunrise experiment conducted on 19 December 2005. For these two events, it actually seems that the weak event had better conditions for new particle formation with a lower temperature, higher RHI and similar surface area, compared to the graph for the strong event (Figs. 7a and 8a). Therefore it is not necessary that the conditions present at the time of new particle formation are responsible



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for such a strong event, other than the low surface area (<1 $\mu m^2\,cm^{-3}$), for this case. The differences come in the back trajectories and the air mass history seems particularly important. For the weak event there was some convection present, however it was much shallower and occurred over the whole five day span (Fig. 9), although both air masses originated from about the middle of the Pacific Ocean at an altitude of ∼3 km, unlike the two previous studies. Also, the rainfall was slightly higher for the strong event compared to the weak event (Figs. 7c and 8c). The solar flux was also slightly higher for the previous five days for the strong event (Figs. 7c and 8c), although it was still lower than the above-mentioned two case studies (Figs. 1c–2c, 4c–5c).

## Latitude dependence of new particles

Figure 10 shows the latitude dependence of the measured particles for all 10 science flights measured in the upper troposphere near the tropopause region. These results show particle concentrations are higher in the subtropics and midlatitudes than in the tropics. Our results are consistent with the Hermann et al. (2003) trend. This is because both these studies were mostly conducted near the tropopause in the midlatitude region at similar latitude ranges. Air mixing induced by convection and the stratosphere and troposphere exchange is stronger in the midlatitudes than in the tropics. In fact, Young et al. (2007) have shown very high frequency (86–100%) and high magnitude (∼700–3960 $cm^{-3}$ $N_{4-9}$ and ∼1000–3990 $cm^{-3}$ $N_{4-2000}$) of new particle formation in the mid-latitude tropopause region. On the other hand, this trend is different from the previous report by Lee et al. (2003) which showed higher concentrations of ultrafine particles in the lower latitudes. This is most likely because the Lee et al. (2003) studies were made on a much larger scale of latitude and altitude (from 10° N to 90° N and 7–21 km; 56 flights) and a majority of the data were taken in the subtropics and polar regions.

## 4 Conclusions and discussions

New particle formation is active in the upper troposphere, because of low temperatures. Although the present study only provides qualitative analysis based on the measured aerosol size distributions and trajectory air mass history calculations, our case studies indicate that strong new particle formation is associated with convection and low surface areas; on the other hand, without convection which aids to increase aerosol precursor concentrations, even with low surface area conditions, new particle formation was not active. Our observations are consistent during other flights on the GV (Young et al., 2007) and consistent with the findings from previous aircraft observations (de Reus et al., 1998; Nyeki et al., 1999; Ström et al., 1999; Twohy et al., 2002; Lee et al., 2003; Minikin et al., 2003; Hermann et al., 2003). Convection brings higher concentrations of aerosol precursors [including $SO_2$ and water insoluble organics (Kulmala et al., 2006) and water vapor] to higher altitudes where temperature and surface areas are lower; abrupt air mixing can also take place during strong convection. These factors together can create an ideal condition for aerosol new particle formation: higher aerosol precursors, lower surface areas, low temperatures, and air mixing. Strong new particle formation events also were related to lower surface areas (Table 1), consistent with the aerosol nucleation predictions by Lovejoy et al. (2004), although we also showed here some cases where weak events also had relatively low surface areas. It is possible that one cannot expect direct anti-correlation of new particles and surface area, because nucleation is governed by both source and sink; as previously shown by Lee et al. (2003), new particles are a function of sulfuric acid production rate and surface area ratio rather than a function of surface area alone. Since this region has lower aerosol precursor concentrations, nucleation can be particularly sensitive to surface area and our results show that this is the case, in general (Table 1).

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involved in the NSF/NCAR GV Progressive Science Missions. We also thank C. H. Twohy for useful comments on our previous work, which inspired us to draft the current manuscript.

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**Table 1.** The average particle concentrations from 4–9 nm, $N_{4-9}$, average particle concentrations from 4–2000 nm, $N_{4-2000}$, and other key meteorological parameters measured during the NSF/NCAR GV Progressive Science Mission measurements. All 10 research flights are included here. One standard deviation values (1 $\sigma$) are also included. NPF indicates the new particle formation events.

| | All Days | NPF | Non-NPF |
|---|---|---|---|
| $N_{4-9}$(cm$^{-3}$) | 650±1250 | 670±1270 | 70±480 |
| $N_{4-2000}$(cm$^{-3}$) | 830±1420 | 920±1470 | 170±630 |
| Surface Area ($\mu$m$^2$cm$^{-3}$) | 4.7±39.1 | 3.6±18.0 | 16.1±132.7 |
| Temperature (K) | 233.5±19.8 | 228.7±13.5 | 248.4±28.0 |
| Relative Humidity Over Ice (%) | 22.6±31.1 | 17.6±21.2 | 40.1±49.5 |
| Potential Temperature (K) | 323.9±22.8 | 326.4±17.4 | 316.8±34.4 |
| $H_2O$ Mixing Ratio (ppmv) | 580±1120 | 290±540 | 1580±1850 |
| Altitude (km) | 8.75±3.63 | 9.52±2.49 | 6.54±5.28 |
| Fraction of samples (%) | 100 | 78 | 22 |



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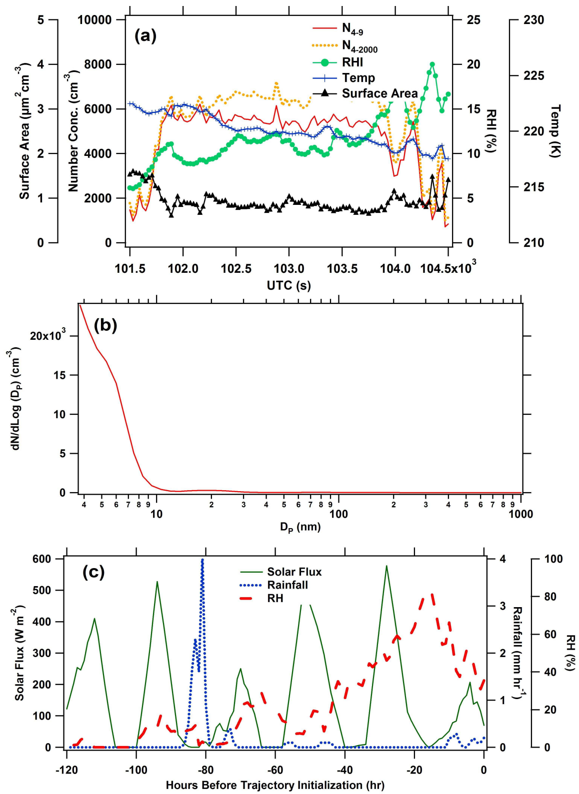


**Fig. 1.** Plots of various parameters for a strong new particle formation event occurring during the night on 2 December 2005. **(a)** The measured $N_{4-9}$, $N_{4-2000}$, RHI, temperature and the surface area of pre-existing particles as a function of time. **(b)** The measured, average particle size distribution for this event. **(c)** The calculated solar flux, rainfall and RH as a function of time before the event, from the NOAA HYSPLIT trajectories. This event occurred at 35° N, 109° W, and 10 000 km.

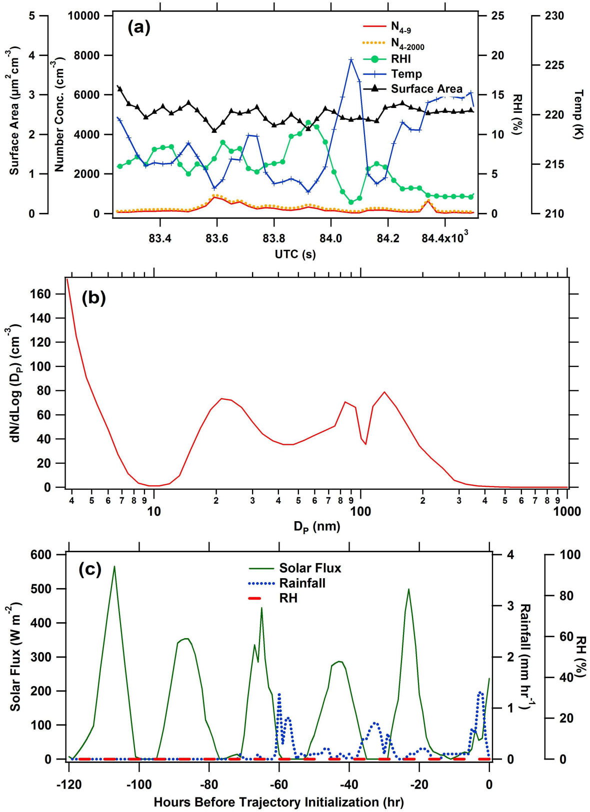


**Fig. 2.** The same as Fig. 1, except for a weak new particle formation event occurring during the day on the same day (2 December 2005). The second size mode at 20 nm **(a)** is representative of more aged particles that grew from newly formed fresh particles. The dip at ∼90 nm in the size distribution **(b)** comes from the inversion program when combining the NMASS and the FCAS data together and may not be representative of the actual aerosols sizes. The same is true for other figures. This event occurred at 35° N, 115° W, and 12 000 km.

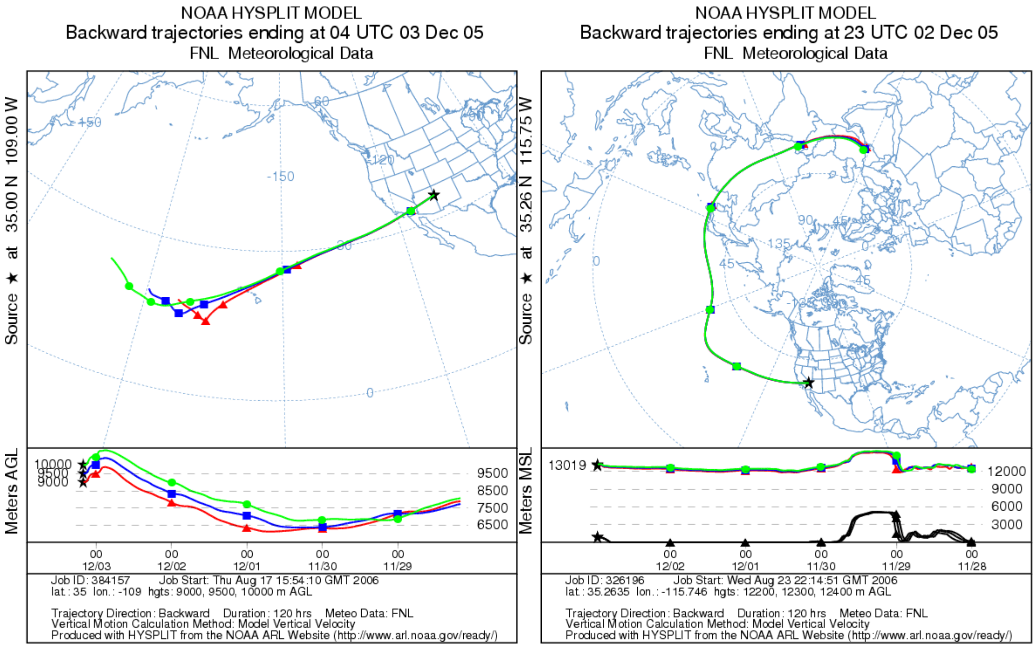


**Fig. 3.** Back trajectories for the strong (left panels) (corresponding to Fig. 1) and weak event (right panels) (corresponding to Fig. 2) on 2 December 2005. The star indicates where the event occurred. Altitude variations as a function of the number of days prior to the event are also shown.



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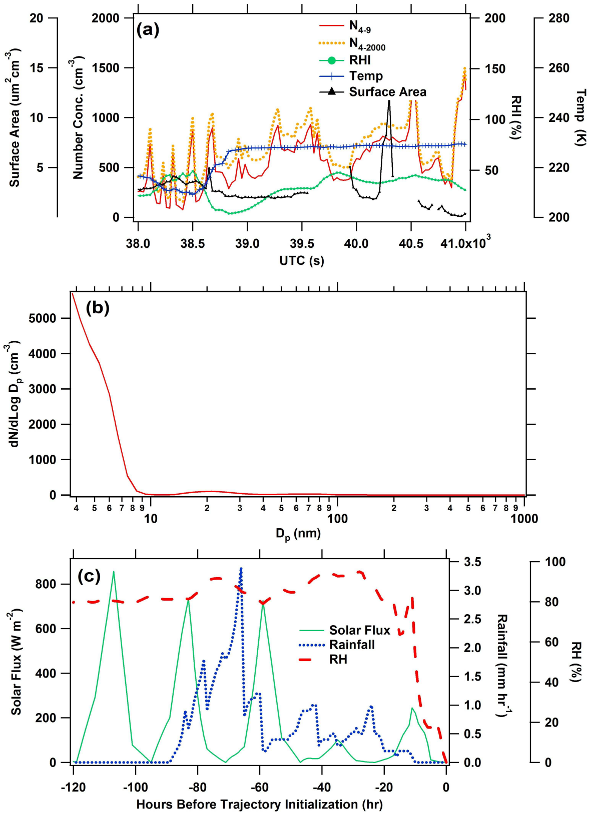


**Fig. 4.** The same as Fig. 1 except for a strong new particle formation event occurring before sunrise (nighttime) on 12 December 2005. This event occurred at 36° N, 115° W, and 10 000 km.



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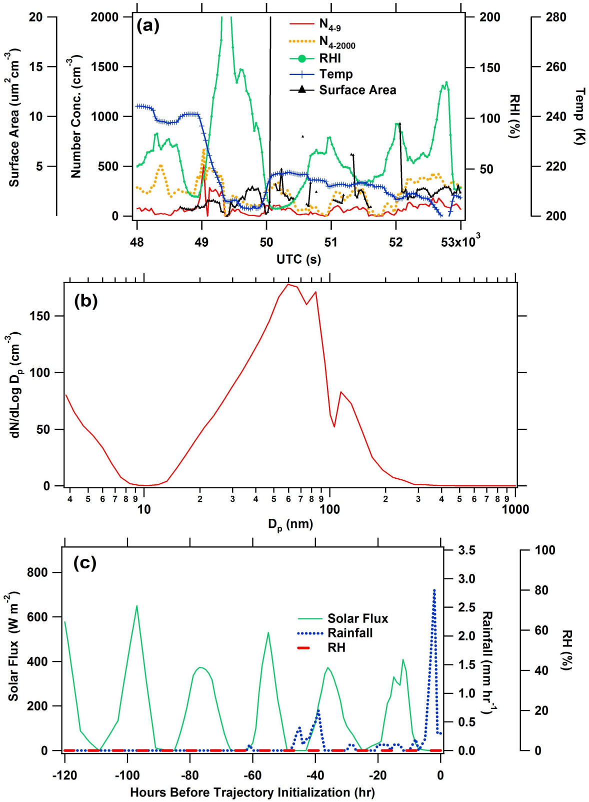


**Fig. 5.** The same as Fig. 1 except for a weak new particle formation event occurring during the day on 12 December 2005. This event occurred at 25° N, 122° W, and 14 000 km.

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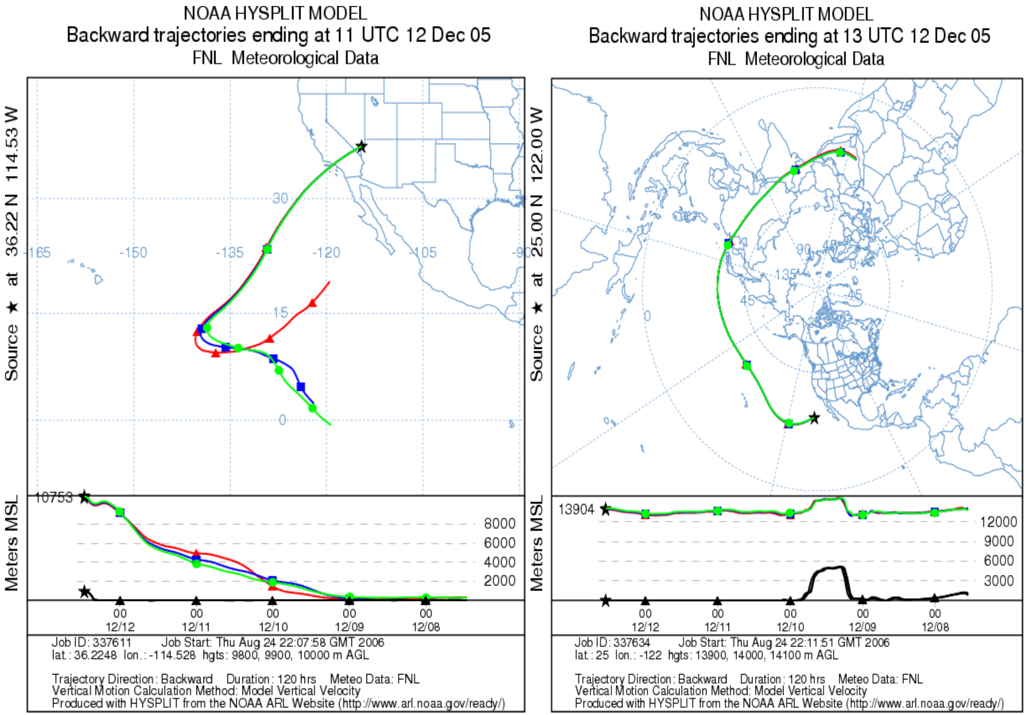


**Fig. 6.** The same as Fig. 3, except for the strong (left) (corresponding to Fig. 4) and weak (right) (corresponding to Fig. 5) new particle formation events occurring on 12 December 2005.



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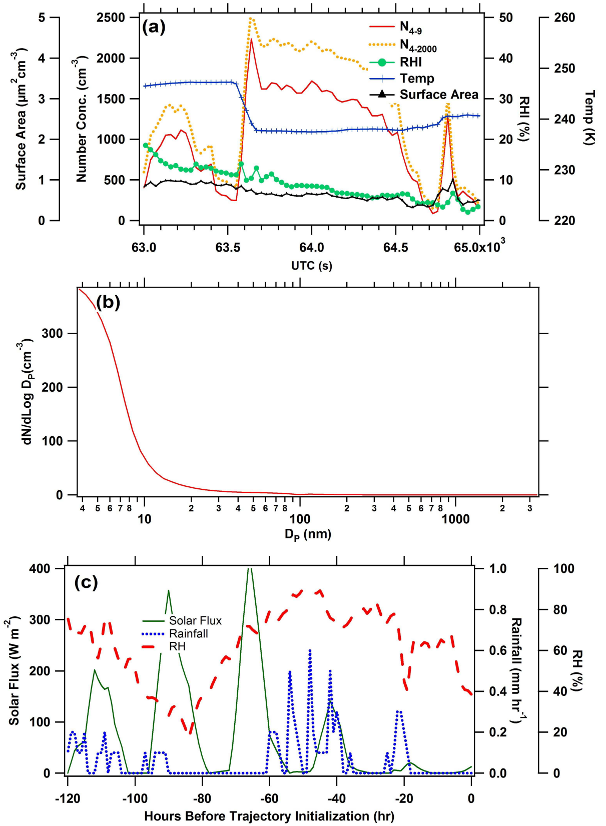


**Fig. 7.** The same as Fig. 1, except for a strong new particle formation event occurring during the day on 19 December 2005. This event occurred at 57° N, 116° W, and 8000 km.



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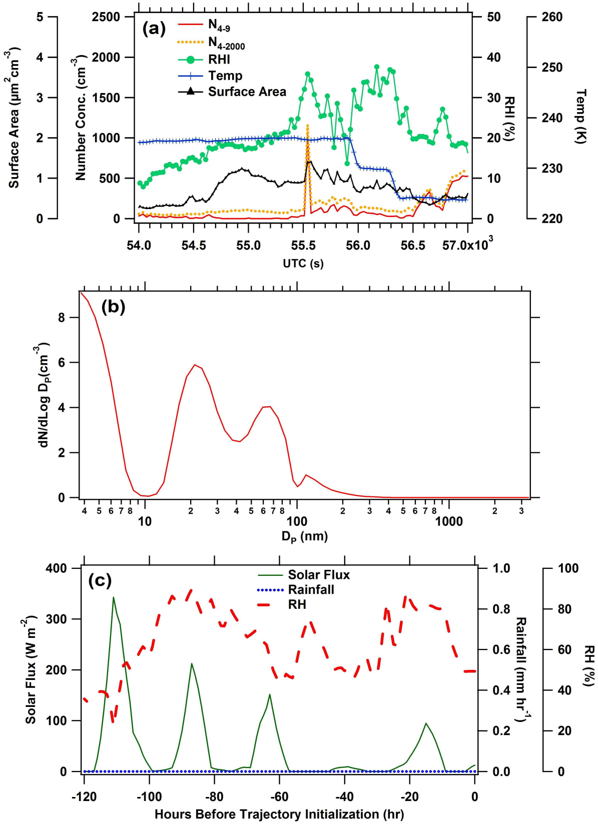


**Fig. 8.** The same as Fig. 1, except for a weak new particle formation event occurring during the day on 19 December 2005. This event occurred at 53° N, 114° W, and 8000 km.



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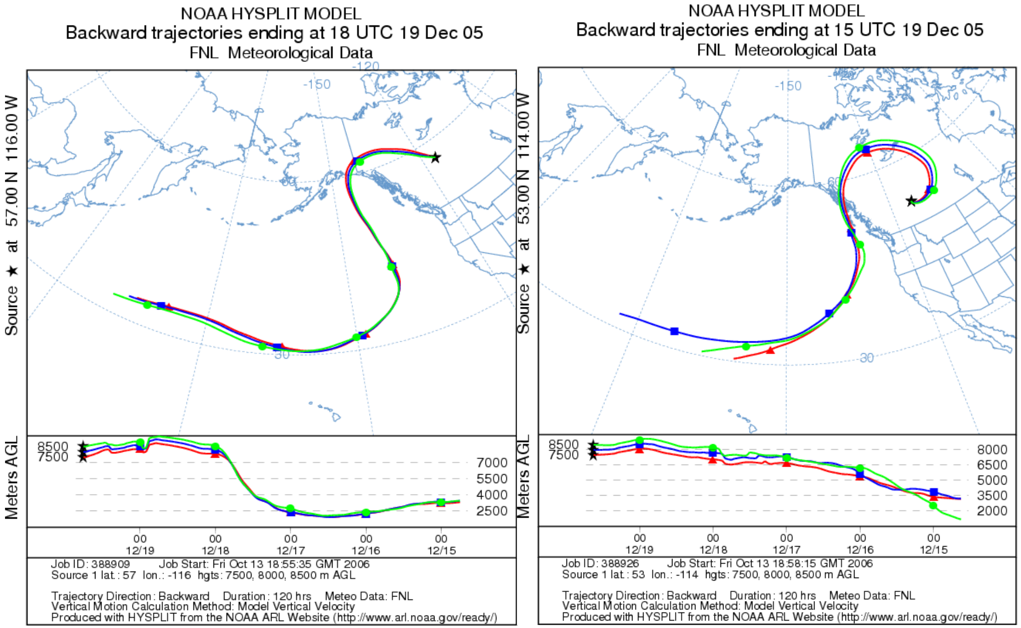


**Fig. 9.** The same as Fig. 3, except for the strong (left) (corresponding to Fig. 7) and weak (right) (corresponding to Fig. 8) new particle formation events occurring on 19 December 2005.

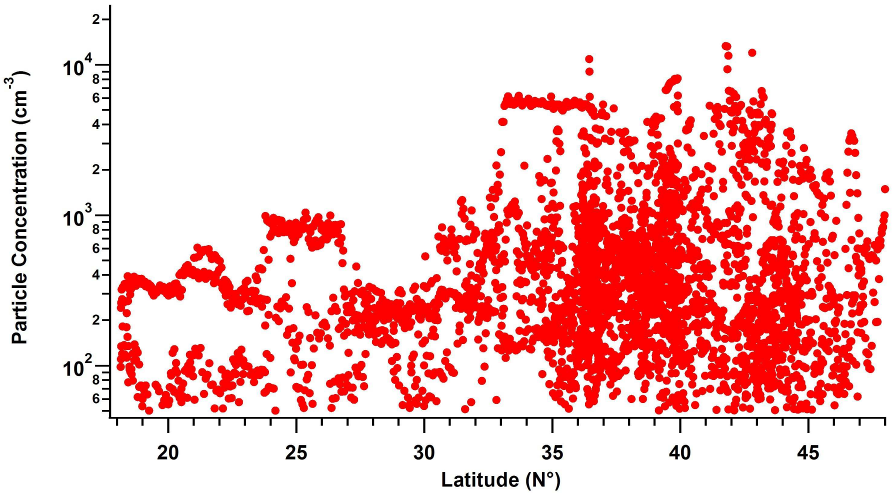


**Fig. 10.** Latitude dependence of particles from 4 to 2000 nm measured during the GV Progressive Science Missions (all 10 research flights are included here). Most of these measurements were made in the upper troposphere near the tropopause region in the midlatitude. Majority of these particles (>71%) are ultrafine particles (Table 1.)

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