Influence of Solar Activity Cycles on Earth's Climate

Task 5 – Hypothetical Physical Mechanisms

WP503 – Role of Ionisation

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CONTENTS

1. Introduction

Cosmic rays are the main source of ionization in the troposphere (Bazilevskaya 2000), and there is increasing evidence that cosmic rays, which are modulated by the solar wind, can noticeably affect Earth's climate, via an influence on tropospheric cloud properties (Carslaw et al. 2002; Marsh and Svensmark 2000a, b, 2003b; Svensmark and FriisChristensen 1997).

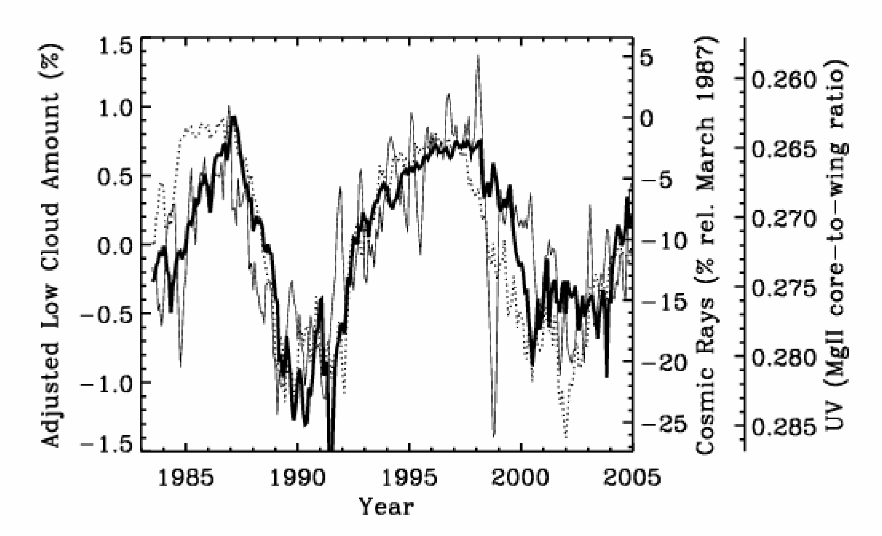

Fig. 1: Monthly averages of ISCCP-D2 IR global Low Cloud Amount derived from a combination of polar orbiting and geostationary satellites (thin), cosmic rays (thick), and solar UV (dotted). The Low Cloud Amount has been adjusted to take account of a possible inter-calibration problem after 1994 (see Marsh and Svensmark 2003b).

The latest update of low cloud amount (LCA) and cosmic rays is shown in Fig. 1 However, variability in LCA correlates equally well with TSI or solar UV, and cannot be uniquely ascribed to a single mechanism when using globally-averaged data. This has led to suggestions that the cosmic ray-low cloud link is a result of a tropospheric circulation response to TSI or solar UV (Kristjansson et al. 2002). However, the ISCCP cloud data now span two solar cycles which allows for a qualitative comparison over the "22 year" cycle. This cycle is the result of a change in the Sun's magnetic polarity between consecutive solar cycles and affects the shielding of GCR as they enter the heliosphere. Under solar minimum conditions this results in a pointed GCR peak during the late 1980's and a more rounded GCR peak during the 1990's, a feature which is also apparent in LCA, but not in TSI or UV (see Fig. 4). Another way to distinguish between these processes is to utilize the property that cosmic rays arriving at Earth are additionally modulated by the geomagnetic field whereas solar irradiance is not. Recent observational evidence indicates that the solar cycle amplitude in LCA, over the period 1984–2000, increases polewards and possesses a similar latitudinal dependence to that found in cosmic ray-induced ionization (CRII) of the troposphere (Usoskin et al. 2004). This supports a physical mechanism involving cosmic rays rather than solar irradiance.

Ionization from GCR possibly influences the atmospheric aerosol distribution on which clouds form, and/or affects the global atmospheric electric circuit with the potential to enhance aerosol-droplet collision efficiencies at cloud boundaries. Below the current status of both mechanisms is discussed together with a presentation of the important aspects of low cloud formation.

2. Ion mechanisms in the lower atmosphere

Ions produced through the nucleonic cascade of cosmic rays in the troposphere rapidly interact with atmospheric molecules and are converted to complex cluster ions (Gringel et al. 1986;Hoppel et al. 1986). There are two loss terms for the ions; ion-ion recombination and ion-aerosol attachment. Ion production rates in the lower troposphere mean that the average lifetime of a small ion is up to 350 s. The ions act as charge carriers in the Global electric circuit which can generate charge build-up at cloud boundaries, and affect the processing of CCN/cloud droplets (Electroscavenging). Ions are also thought to play a role in stabilizing an initial atmospheric cluster until it is large enough to continue growing via neutral growth mechanisms into a particle/CCN (Ion Induce Nucleation). In addition, ions will attach to pre-existing aerosol nucleated through neutral processes, once charged there is a potential the aerosol's continued growth is enhanced (Charge Assisted Growth). These mechanisms are likely to have an impact on the number of aerosols acting as cloud condensation nuclei (CCN) at typical atmospheric super-saturations of a few percent [Viggiano and Arnold 1995]. In the following the important aspects of Ion Induced Nucleation, Charge Assisted Growth, and Electroscavenging will be discussed.

2.1 Ion Induced Nucleation (IIN)

Atmospheric aerosols are either released from the surface or are nucleated in-situ within the atmosphere. Bursts of nucleation have frequently been observed in the lower troposphere (Kulmala, 2004), and are thought to be an important source of CCN, particularly over maritime regions. The nucleation of new aerosol in the atmosphere is the result of a competition between condensation/coagulation growth processes and evaporation. Under typical atmospheric conditions H2SO4 and H2O are thought to play an important role in the initial stages of new aerosol formation. However, initial clusters involving only a few H2SO4 molecules are more unstable than much larger clusters, due to the lower binding energies at the initial cluster surface. It is easier for a molecule to escape from a curved surface than a plane surface, thus the stability, which is related to curvature of a droplet, is size dependent. To over come this initial lack of stability through dissociation H2SO4 molecular concentrations, and hence collision rates with the initial ion clusters, must be high (>10^8 cc). However, typical atmospheric concentrations of the gas phase H2SO4 are too low (<10^7 cc) for clusters of a few molecules to be stable, thus the initial clusters should tend to evaporate and nucleation ought to be inhibited. Since observations indicate that in-situ nucleation is an important source of atmospheric aerosol, some additional process must be operating to stabilise the clusters during their initial growth until they are large enough to be stable in their own right and are considered nucleated. It is currently an open question as to how the initial clusters are stabilised until they reach this critical size. One mechanism that has been gaining increasing attention is the role of ionisation during the early stages of nucleation, known as Ion Induced Nucleation (IIN). Since the majority of atmospheric ions are generated by cosmic rays, IIN processes could be important for understanding a sun-climate link.

2.1.1 Modelling the Physics of IIN

A number of theoretical models have been developed to explore IIN, and there are currently two models of particular note referred to here as Yu's model (Yu and Turco, 2000; Yu, 2006), and Lovejoy's model (Lovejoy et al 2004). A brief overview of the physics included in each of these models is given below.

Both models describe the full kinetic behaviour of cluster production at molecular resolution for a H2SO4/H2O system under atmospheric conditions. The evolution of the size spectrum and composition of neutral and charged clusters are followed from a few molecules up to large particles (um). All clusters are allowed to grow through condensation and coagulation, and shrink through evaporation; small ion clusters are additionally lost through recombination or ion-aerosol attachment (figure 1). Nucleation on ions is favoured because the charged clusters are more stable thermodynamically, and their growth rate enhanced and evaporation lowered due to the interaction between ions and polarizable vapour molecules. After the initial growth of small ion clusters, recombination generates large neutral clusters, but they will shrink due to evaporation if smaller than the critical size. The nucleation rate is then determined by the number of clusters exceeding the critical size after recombination. This is partly dependent on atmospheric ambient conditions and partly on the cluster thermodynamics described in the model. Thus IIN is essentially the ability of ions to help maintain the initial cluster, limit evaporation, and enhance initial cluster growth before recombining to produce a stable neutral aerosol.

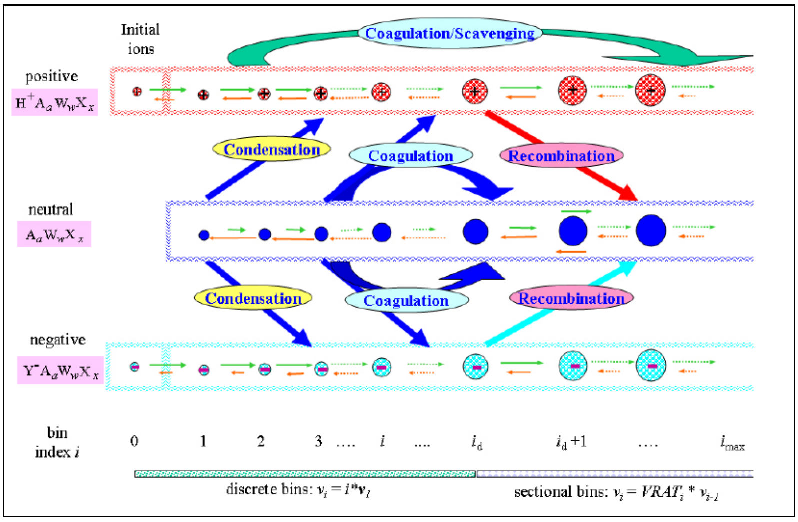

Figure 1: The kinematical models treat condensation, coagulation and recombination of charged and neutral clusters. Notice how the sizes of the small green arrows indicate increased probability for charged cluster coagulation and growth compared to neutral growth for small clusters. Yu's model predicts significant IIN under lower tropospheric conditions. Figure reproduced from Yu (2006).

Although the models of Yu and Lovejoy are similar, there is a major difference. Yu's model predicts significant growth of the cluster ions before recombination under lower Tropospheric conditions (potentially important for low clouds – see later) which provides a significant source of new particle generation in the presence of ionisation. However, under similar conditions Lovejoy's model only finds a negligible contribution to nucleation from ions, but a relatively large contribution at mid-upper Tropospheric conditions (figure 2). This appears to be a result of differences in the physical description of the cluster thermodynamics, and the treatment of positive ions which are explicitly allowed to evolve by Yu, but treated as a single species restricted to small ions in Lovejoy's model. However, there are large uncertainties in both these processes due to a lack of rigorous experimental data and lack of a suitably robust theoretical description of cluster thermodynamics that are valid under a wide range of atmospheric conditions.

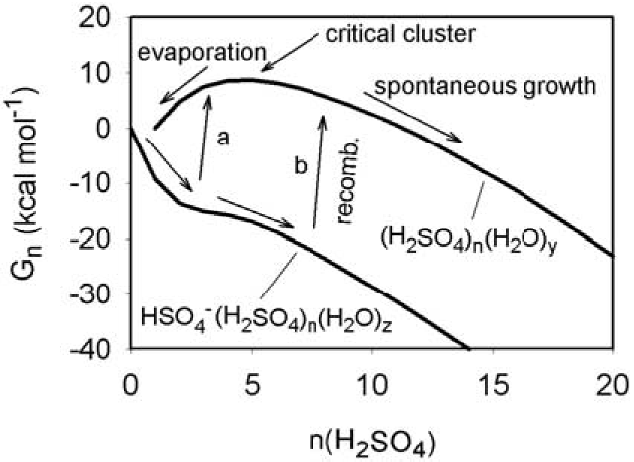

Figure 2: Gibbs free energy (G_n) versus number of H2SO4 molecules (n) in a cluster under upper tropospheric conditions. The upper branch indicates the energy barrier that must be negotiatied by neutral particles before nucleation can occur. The lower branch is the equivalent energy curve in the presence of an ion – note the lack of energy barrier. Lovejoy's model suggests that under lower tropospheric conditions the energy barrier in the presence of an ion is similar to the neutral case thus limiting IIN. Figure taken from Lovejoy et al (2004).

To summarise, both models indicate that, based on current nucleation theory, there is a significant role for ionisation in the nucleation of new particles in the atmosphere, but that it is unclear as to where in the atmosphere the largest contribution should be expected.

2.1.2 Observational Evidence for IIN

Growth of atmospheric ions has been observed in a number of in-situ situations. At a ground based station in Tahkuse, Estonia, Horrak et al (1994; 1998; 2000), detected the growth of small ions from 1.6nm - 6nm (referred to as intermediate ions), sizes well above the critical cluster size. Both positive and negative ions were observed to grow through the intermediate ion size, however, charge asymmetry existed with higher concentrations of negative ions. It was speculated that this was due to the preferential condensation of different atmospheric trace gases onto one or other polarity.

Positively charged ion growth events have been reported in the upper troposphere by (Eickhorn et al 2002). Available instrumentation limited observations to only positive ions in the upper troposphere, thus growth events in other parts of the atmosphere or on negative ions are not ruled out. Figure 3 is a plot of two average ion spectra from this campaign, one describing the background small ion distribution (< 1 nm), and the other significant ion growth events leading to the formation of massive ions (>1.6nm). Initial ion growth between 300-1000amu (<1.4nm) was consistent with the uptake of H2SO4 under upper tropospheric conditions with realistic gas concentrations (figure 3 dashed curve). However, condensation could not explain the growth of the massive ions. These largest ions were argued to have formed as the result of IIN producing large stable neutral particles through recombination, and then the subsequent attachment of a positive small ion rendering the particle detectable by the instrumentation.

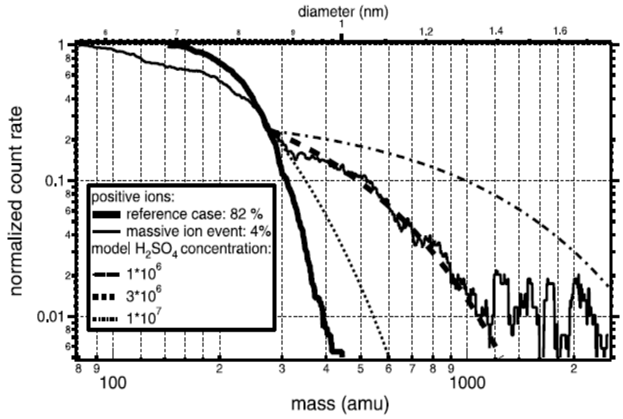

Figure 3: Positive ions (<2nm) versus normalised count rate detected in the upper troposphere (taken from Eickhorn et al (2002)). The thick solid curve represents the small ion spectrum typically found in 82% of the cloud free observations. The thin solid line indicates that massive ion growth was detected in 4% of the observations. Modelled ion spectra based on uptake of H2SO4 for various concentrations are indicated by the dashed lines.

Lee et al (2003) reported aircraft based observations of aerosol particles (both charged and neutral) in the size range 4 to 2000nm. They found very high concentrations of ultra-fine particles (< 9nm) with evidence of significant new particle nucleation in the upper troposphere/lower stratosphere (7-13km). In this campaign ions were not separately measured, however, Lovejoy's model (described in the previous section) was used to estimate the evolution of particle production and growth assuming IIN was an important mechanism. A remarkably good agreement was found between the simulations and observations up to 5 days after a nucleation event (figure 4). Other nucleation mechanisms not involving ionisation were ruled out due to the low concentration of trace gases at these altitudes.

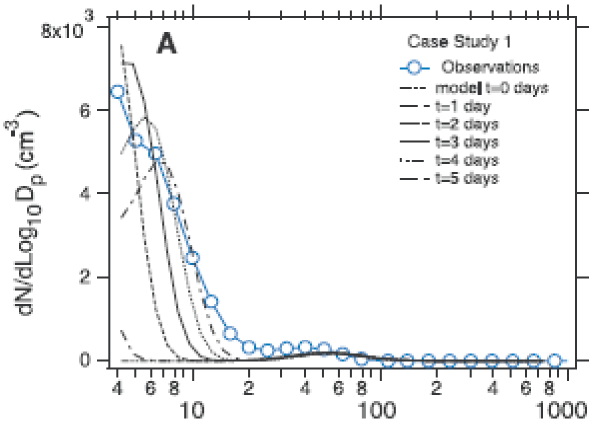

Figure 4: An example of observed particle growth in the upper troposphere and lower stratosphere measured from 4 nm and up to 2000 nm (blue curve), well above the nucleation size. Simulations using Lovejoy's IIN model (black curves) agree very well with the observations 5 days after a simulated nucleation event. Figure taken from Lee et al 2003.

2.1.3 Experimental Evidence of IIN

Until recently experimental efforts to explore the role of IIN had been performed under conditions that were not relevant for understanding atmospheric processes. Either unrealistically high levels of ionisation were present or the concentrations of gases were far from those observed in the atmosphere. The major limiting factor has been the difficulty in controlling within a laboratory environment the very low levels of reactive gases that are present in the atmosphere. However, recent experimental efforts have now begun to explore nucleation under realistic atmospheric conditions, two of which are described below.

Ionisation is not the only mechanism with the potential to stabilise the initial cluster growth. Other mechanisms have been proposed that involve the presence of trace amounts of various organic gases. The role of organic trace gases in stabilising the nucleation of H_2SO_4/H_2O particles has been investigated in an atmospheric pressure flow tube by Berndt et al. (2006). Furan was used as the test organic and it was found that nucleation of particles in the flow tube was independent of its presence (figure 5a). This suggests that organics are not necessarily needed, a null result which is consistent with the possibility of a mechanism involving ions. However, Berndt et al (2006) additionally measured the fractions of positively and negatively charged particles that were produced (figure 5b).

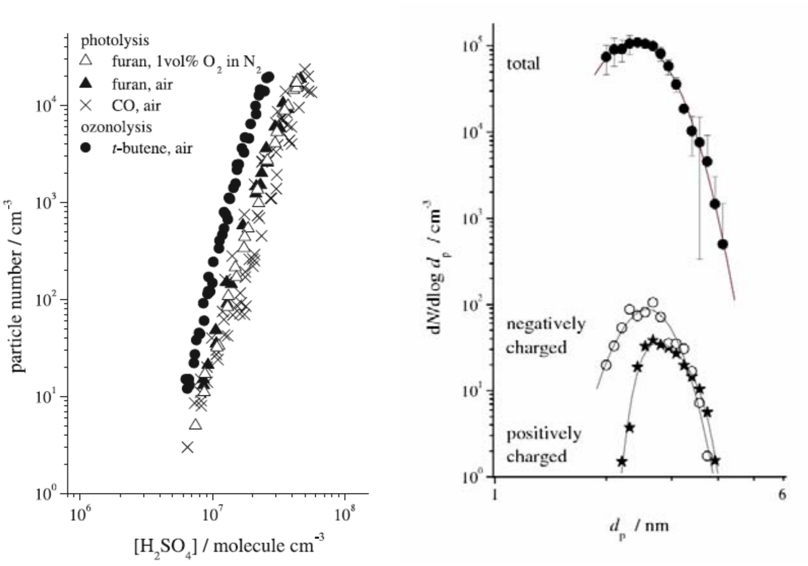

Figure 5: a – left panel) Number of H_2SO_4 molecules in a cluster versus particle concentration both with (triangles) and without (crosses) the presence of organics. The solid dots examine nucleation in the absence of UV illumination. In all cases little difference in nucleation and growth rates are observed. b – right panel) Particle diameter versus concentration for charged and total particle size distributions. Figures taken from Brandt et al (2006).

An excess of negatively charged particles was observed between 2-3nm. However, the charged particle concentration only represented a small fraction (< 1%) of the total particle concentration. Based on this observation, Berndt et al (2006) maintained that IIN was of minor importance under their experimental conditions. However, their instrumentation was only able to detect particles, charged or neutral, down to diameters >2nm. So it is unclear why they rule out the possibility that IIN could have nucleated the particles sub 2nm that, following recombination, would then appear neutral in the instrumental detection limit above 2nm. The charge distribution observed in their experiment could then be explained from ion-aerosol attachment where the neutral aerosol was initially formed under IIN; a similar argument was proposed by Eickhorn et al (2002) based on their in-situ observations of positive ions with diameters <2nm in the upper troposphere (see above).

An experiment at our own institute (DNSC) has been designed to explicitly explore the role of ionisation in the initial nucleation process (Svensmark et al, 2006). The measurements were performed in a 7 m^3 chamber containing trace amounts of ozone, sulphur dioxide, and water vapour at concentrations relevant for the Earth's atmosphere. Ions were produced in the chamber by the naturally occurring galactic cosmic radiation and by the decay of the natural abundance of radon. The average production of ions could be additionally enhanced to between 1-60 ions/cc/s, levels that are relevant throughout the troposphere. Figure 6 indicates how aerosol concentrations (diameters > 3nm) are proportional to ion concentration within the chamber. Comparison with a simple growth model including condensation/coagulation processes provides an estimate of the nucleation rate, S, of particles that reach the critical cluster size. The results indicate that the production rate of new aerosol is proportional to the square root of the ion production rate.

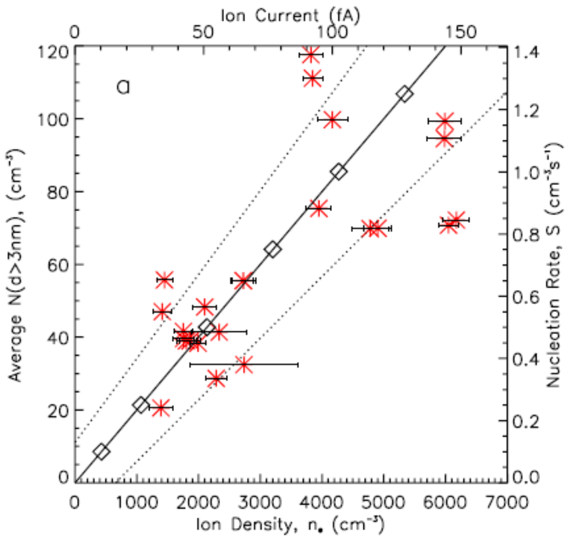

Figure 6: Ion density versus average aerosol concentration (diameter >3nm). The nucleation rate, S, on the right hand axis was estimated from a comparison with a simple growth model. The ion current on the upper axis was the physical parameter measured from which ion density was calculated. Figure taken from Svensmark et al 2006.

The timescale for the production of a critically sized cluster was tested in the presence of an electric field applied across the chamber. By varying the field strength the lifetime of ions in the chamber could be reduced. With a suitably large field IIN should be shut down as the ions would not have time to nucleate a new particle before their removal from the chamber. The maximum field strength was 12000 V/m which is equivalent to an ion lifetime of ≤ 2 secs, significantly less than the timescale for recombination (~300 secs) that limits the IIN mechanism described above. However, in the presence of this field particle concentrations were only reduced by 50%, and it was clear that significant nucleation involving ions was still occurring. The nucleation process under these experimental conditions definitely involved ions, not as a result of IIN, but through some other mechanism operating at a much faster time scale. It was speculated in Svensmark et al (2006) that the charge initially nucleated a cluster before detachment due to excitation by a thermal photon or the release of chemical energy within the initial cluster. The detached charge could then nucleate a new cluster, continuously repeating this cycle, until lost through recombination or aerosol attachment. These experimental results suggest that nucleation rates are limited by the residence time of a charge in a cluster necessary to produce a stable cluster. If, as the results suggest, this is much shorter than the recombination timescale, multiple nucleations from each individual charge is possible, leading to nucleation rates that are potentially much larger than the ion production rate. In contrast the maximum nucleation rate involving IIN would be limited by the ion production rate. So the proposed detachment mechanism acts as an amplification factor for new particle production involving atmospheric ionisation.

2.2 Charge Assisted Growth

The previous section has focused on the role of ions in the initial nucleation of aerosol particles. But ions may also play a role when they become attached to pre-existing neutrally stable aerosol particles that have formed without necessarily requiring ionisation. The presence of the charge on the aerosol will probably increase reaction rates with surrounding molecules, enhancing the growth to CCN sizes (~0.1um). However, aerosol that has become charged though ion-aerosol attachment is only a fraction (< 1%) of the total aerosol in the atmosphere (figure 5). It is unlikely that this effect would have a significant influence on CCN sized aerosol distributions affecting clouds.

2.3 Electroscavenging and the Global Atmospheric Electric Circuit

A further suggestion is that the amplification of cosmic rays on climate could be through changes in the global atmospheric electric circuit (Rycroft et al. 2000). Current flowing in the global atmospheric electrical circuit substantially decreased during the twentieth century, which has been quantitatively explained by a decrease in cosmic rays (CR) reducing the ionospheric potential as solar activity increased (Harrison 2002). This potentially affects aerosol-cloud interactions at the edges of clouds, e.g., Tinsley (2000), (see a review of possible mechanisms in Harrison and Carslaw (2003)). Highly-charged droplets are generated at cloud boundaries in the troposphere due to the weak vertical currents of the global electric circuit. Once these droplets have evaporated, highly charged CCNs remain, and the presence of this charge enhances collision efficiencies when interacting with other liquid droplets. The process of nucleation and evaporation repeats itself continuously and is thought to aid in the formation of ice particles in supercooled liquid water clouds; as a result it is referred to as ‘electroscavaging’ (Tinsley 2000).

It is however more unlikely that ‘electroscavaging’ has a global effect (see Harrison Carslaw article) on cloud generation and ultimately on global climate although there is some limited observational evidence to suggest that this process can have an additional influence on atmospheric dynamics (Roldugin and Tinsley 2004).

3. Low Clouds

3.1 Conditions for low cloud formation

Clouds form where atmospheric air cools below its dew point. In the atmosphere this most often happens by the approximately adiabatic expansion of rising air, but can also be due to radiative cooling (often resulting in fog) or the mixing of air masses with different temperatures and humidity. Low level clouds form when the vertical extent of rising air is limited to the lower few kilometers of the troposphere, with low cloud often capping a region known as the planetary boundary layer. Low clouds are often classified into Stratus or Cumulus clouds or some combination of the two known as stratocumulus. Stratus clouds appear in extensive flat layers covering 100's of kms in horizontal extent, and typically form when the vertical air motion arises from large scale convergence, frontal lifting or orographic lifting. Cumulus clouds are more 3-dimensional in nature with their vertical and horizontal dimensions of similar scale, typically <1km for low clouds that develop due to limited vertical convection of unstable air.

Typical conditions for the development of low cloud occur within cold air outbreaks or due to temperature inversions just above the planetary boundary layer. Cold air outbreaks occur when large scale atmospheric motion leads to cold, dry air from land or sea-ice regions flowing out over the relative warm oceans. This situation often materializes behind cold frontal systems. The subsequent vertical transfer of heat and moisture from the ocean to the atmosphere leads to the formation of low cloud. Temperature inversions in the vertical profile of the atmosphere are also ideal situations for the formation of Low cloud. Over the vast sub-tropical oceanic regions this is particularly important as descending warm, dry air from the Hadley circulation meets weak updrafts of relatively cool, humid air. The subsiding air from above warms adiabatically creating a temperature inversion just above the Marine Boundary Layer (MBL) that limits the upward motion of moist air and caps the vertical extent of cloud formation.

While the large scale circulation of the atmosphere controls the vertical extent, the rate of cooling and the amount of condensation available in low clouds, the properties of cloud droplets, their in-cloud inter-actions and the rate of any subsequent precipitation is determined by much smaller scale microphysical processes. All droplets in low clouds form when water vapour condenses on atmospheric aerosol particles acting as CCN. Properties of the background aerosol distribution limit the number of droplets that are able to form, which in turn affects the growth rate and inter-actions of a cloud's droplet distribution. This ultimately regulates cloud lifetime and cloud radiative properties both of which play an important role in the Earth's radiation budget.

3.2 Multiple Cloud Regimes

It is becoming increasingly apparent from observations that when conditions for low cloud formation exist multiple cloud regimes are possible (Stevens et al, 2005), i.e., stratus, stratocumulus, and cumulus. These regimes appear dependent on the local dynamical and microphysical properties within the MBL, where clouds have formed, rather than changes in the large scale circulation of the atmosphere (Petters et al, 2006). Evidence is beginning to suggest that the number of CCN not only determine cloud droplet concentrations, but also play an important role in the microphysics maintaining the different cloud regimes. Evolution between the regimes is not continuous but occurs abruptly, in steps, as feedbacks involving precipitation and microphysics re-organize themselves over a matter of hours (Wood and Hartmann 2006). Rosenfeld et al (2006) suggest that this is in response to changes in concentrations of the background aerosol acting as CCN passing through various thresholds. When close to one of these thresholds a small perturbation in CCN concentrations can lead to considerable changes in cloud properties with a potentially large impact on Earth's climate. Below is an overview of the microphysics involved and the role aerosols/CCN play in maintaining the different low cloud regimes.

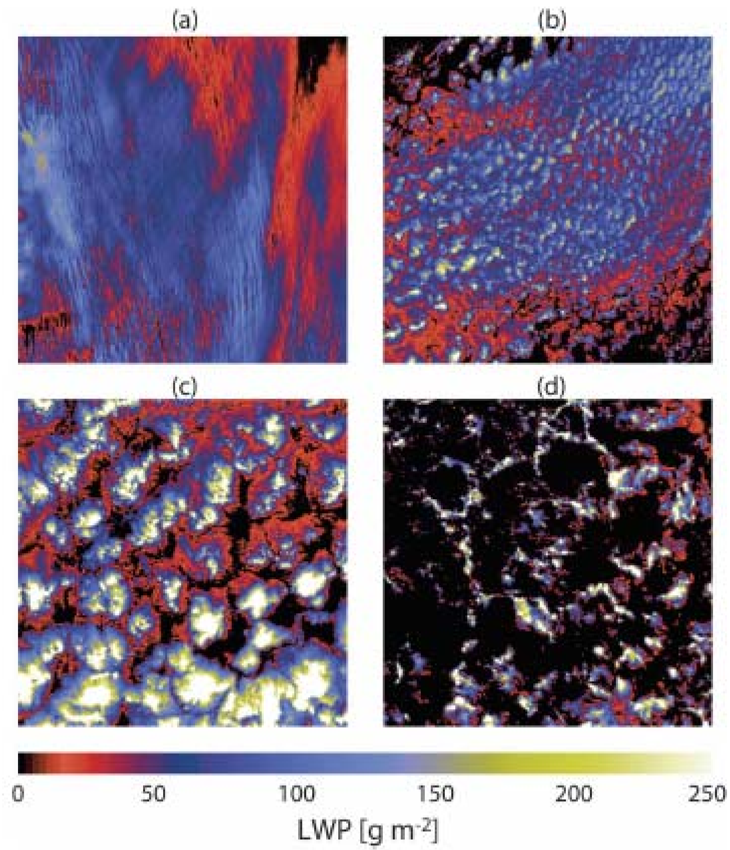

Figure 7 – MODIS scenes of Cloud Liquid Water Path's (LWP) providing examples of different cloud regimes, where a) Unstructured stratus, b) closed cell stratocumulus, c) transition to open cell, and d) open cell cumulus. Figure taken from Wood and Hartmann (2006).

Satellite images have provided numerous examples of the multiple cloud regimes, which are particularly evident over the sub-tropical oceans (figure 1). Studies indicate that the dynamical properties of the MBL typically evolve into Mesoscale Cellular

Convection (MCC), where the horizontal scale of the convective cells is much larger than the depth of the MBL (< a few kms). Rosenfeld et al (2006) have identified three regimes once the MBL has evolved into MCC: closed cells (100% cloud fraction) driven by cloud top radiative cooling, open cells (40% cloud fraction) driven by radiative heating from the surface, and a super-clean state (~0% cloud fraction). They propose that the transition between these states is not continuous but occurs abruptly in two steps at low concentrations of CCN. Woods and Hartmann (2006), when comparing both satellite observations and NCEP reanalysis data, show that while the horizontal cell size of MCC scales with the MBL depth, the transition of MCC from closed to open circulation is not strongly affected by large scale atmospheric properties. Although the physical mechanism generating and sustaining these different states is currently unknown, the ability of precipitation to form appears to be key for the evolution from closed to open MCC (Stevens et al, 2005).

3.2.1 The role of precipitation in cloud microphysics

As a cloud begins to form in an updraft of air, the droplets nucleate on CCN and grow through condensation and coalescence. If the droplets reach a size that can no longer be suspended in the updraft then they will begin to fall as precipitation. The number of CCN determines both the concentration and size of cloud droplets, and hence will regulate precipitation rates. Precipitation not only limits the amount of water present in a cloud but also limits the importance of entrainment processes. Entrainment is the mixing of environmental air in to a cloud's boundaries. The entrained air is often much drier than the cloud's own air and thus leads to cloud drop evaporation, and hence cooling, where the two air masses meet. This effect is often referred to as evaporative cooling and drives downdrafts of air into the cloud, further drying the cloud from above. These entrained downdrafts require sufficient cloud water to be present for them to become heavy enough, through evaporative cooling, to penetrate deep into the cloud. So there is a balance between precipitation limiting the cloud water available to drive entrainment and the drying out of the cloud from the ensuing downdrafts. The entrained air also contains a source of CCN from outside of the cloud, replenishing those CCN that were lost through precipitation [Rosenfeld et al, 2006]. By maintaining the concentrations of CCN, cloud drop growth is limited, which helps avoid a run away effect where fewer CCN lead to larger cloud droplets and hence faster precipitation rates.

If precipitation reaches the surface at a rate faster than surface evaporation then the MBL will become drier, the amount of cloud water will decrease, and entrainment will be reduced. However, precipitating particles do not necessarily reach the surface. Depending on their size it is possible droplets will evaporate in the MBL below the cloud, often referred to as drizzle. This will moisten the MBL, which helps maintain the available cloud water and hence strengthen entrainment. (In extreme cases the cooling of the MBL will stabilize the rising air, which ultimately could lead to a decoupling of the cloud from the MBL and its moisture source at the surface). Since the number of CCN available in the atmosphere has an impact on precipitation, externally forced changes in CCN have the potential to modulate this self-regulating cloud mechanism and ultimately the properties of the clouds that form.

3.2.2 CCN driven transition from cloudy to super-clean states

The importance of CCN/precipitation can be demonstrated by considering the transition between the three cloudy states under conditions of MCC. Closed cells are characterized by almost total cloud cover through the broad rising motion at the cell center with narrow regions of cloud thinning due to the downdrafts at the cell walls. Radiative cooling at cloud tops drives this relatively weak circulation generating low precipitation rates at the surface, subsequently enhancing entrainment rates and leading to a rapid replenishment of CCN. If the number of CCN decreases drizzle rates will begin to increase, cooling the lower MBL through evaporation and stabilizing the broad updrafts at the cell core. At some critical point the MBL will have stabilized to such an extent that the cloud layer decouples from the moisture source below. Rapidly, precipitation and drying from entrainment processes will lead to the break up and loss of the cloud at the cell core. These clear regions allow for radiative cooling from deep in the lower MBL, and the subsequent sinking of air in the cell core. Heat fluxes from the ocean surface are then able to drive the MBL from below with a complete reversal in circulation leading to strong updrafts at the narrow cell walls and weaker downdrafts in the now clear cell core. The transition to the open cell regime is now complete with clouds forming only at the cell walls. Stronger updrafts lead to higher precipitation rates, with the corresponding weaker entrainment rates suppressing an important source of CCN replenishment.

Such a scenario is supported by simulations. These indicate that following the onset of precipitation closed cell MCC evolves into cumulus like narrow updrafts at the cell walls accompanied with broader, gentler downdrafts in the core (Stevens 1998). Due to the persistent drizzle in these open cells, CCN are scarce which prevents the build-up of stratiform cloud layers. The reduction in CCN accelerates the drizzle process, reducing the amount of moisture vertically transported within the cloud layer and thus reduces the altitude of cloud tops. The abundance of precipitation modulates both dynamical and microphysical cloud properties, with the scarcity of CCN maintaining the open cell structure. Case studies indicate that under certain conditions CCN sources via entrainment rates (from above) or from sea-to-air rates (from below) are too low to maintain cloud cover (Petters et al, 2006). If the concentrations of CCN are low enough this can result in a run-away effect where the depletion of CCN leads to an increase in precipitation and the near clearing of clouds generating the super-clean state. However, this super-clean state would support the formation of low cloud if concentrations of CCN were lifted above the run-away threshold. Rosenfeld et al (2006) suggest that satellite observations demonstrating the presence of ship tracks, which introduce a massive increase in CCN locally, in an otherwise clear sky environment is evidence in support of this super-clean state (figure 2).

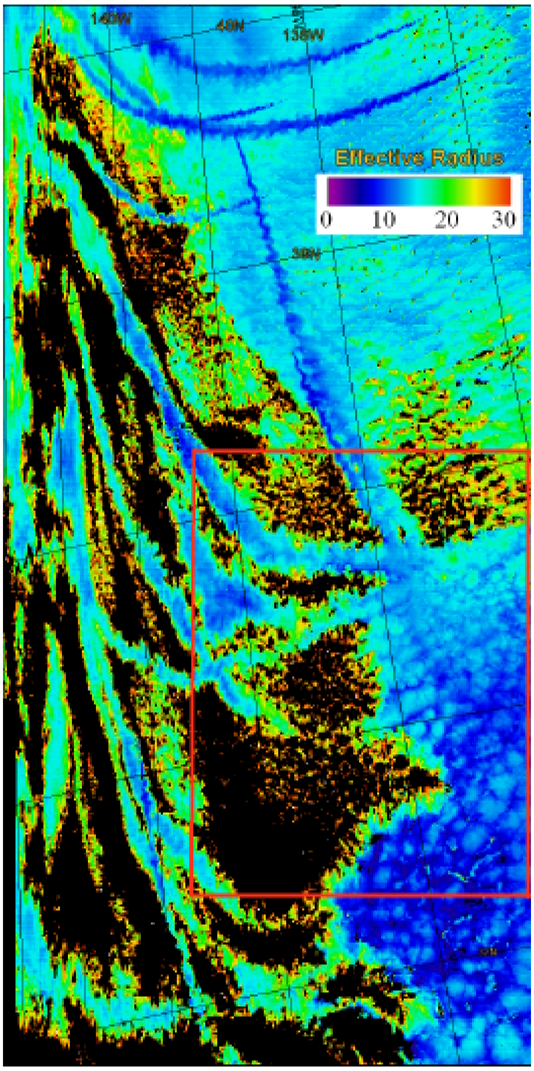

Figure 8 - MODIS scenes of effective cloud drop radius demonstrating the increase in cloud drop size from closed to open cells. Streaks of Ship tracks are clearly present within the otherwise cloud free super-clean state in the left-hand side of the image. Figure taken from Rosenfeld et al (2006).

Another result of this sudden cleaning of the atmosphere is that it clears the way for new particle nucleation (Petters et al, 2006). Thus the number of CCN not only regulates the extent of cloud fraction and cloud top height/thickness, but also influences the potential for new particle formation. Further, the humidity of air just above the MBL can modulate the threshold of transition between these 3 states. For example the entrainment of drier air reduces the amount of water available for precipitation, lowering the threshold level of CCN required for the transition from closed to open cells, and accelerating the transition to the super clean state.

This extreme sensitivity of MBL cloud cover to changes in atmospheric aerosol will have significant consequences for the global radiation budget such that any process having even a minimal impact on CCN will have an amplified climate impact through the processes of the MBL.

4. Summary

The aim has been to try and understand mechanisms explaining a possible link between solar variability and changes in Earth's climate. This report has focused on just one possible mechanism, the role of ionization, and its potential impact on cloud properties. There is no question that the majority of ionization in the lower atmosphere below 35km originates from cosmic rays that are modulated by solar activity. There is also a wide body of evidence indicating that climate and the intensity of cosmic rays arriving at Earth have varied simultaneously over a range of timescale from days to millions of years (for details see TN-WP103/ESA, (Marsh et al, 2005) and references therein). However, it remains an open question as to whether a suitable mechanism exists to generate a significant impact on Earth's climate from solar modulated cosmic rays.

It has been shown here through a combination of models, observations and recent experimental efforts that cosmic ray induced ionization has an impact on aerosol nucleation and growth under atmospheric conditions. A strong case has been made for the role of IIN, with simulations showing consistency with in-situ observations of ion growth. However, it is uncertain whether the lower troposphere is sensitive to the IIN process or not. Experimental efforts have indicated that aerosol concentrations in the lower troposphere are sensitive to ionization and suggest that a mechanism is operating at much faster timescales than indicated by IIN. It is, however, still an open question as to whether the generation of atmospheric aerosol, via ionisation, can grow to cloud condensation nuclei sizes, and whether these nuclei are capable of significantly influencing cloud properties by modifying cloud droplets, cloud optical properties and precipitation.

Yu's model suggests that CCN in the lower troposphere and marine boundary layer (below 5 km) are most sensitive to changes in ionisation. Under such conditions, an increase in GCR would lead to an increase in the number of aerosols, CCN and cloud droplets and hence a decrease in cloud droplet sizes. Ferek et al. (2000) have shown that an increase in aerosol concentrations due to ship exhaust can lead to drizzle suppression. This clearly has an impact on cloud microphysics and in turn implications for cloud properties. However, ship tracks are a large perturbation locally, whereas a possible GCR - CCN mechanism will be a small perturbation globally. If ionization from GCR can be shown to have a similar affect on the lower tropospheric aerosol distribution, and subsequently prolong a cloud's lifetime, it would be consistent with the cosmic ray - low cloud correlation (Marsh and Svensmark, 2000). An effect from ionisation on clouds would provide the link to climate with their strong impact on the radiative budget of the Earth.

Although the majority of recent work in this field has emphasized mechanisms involving ions-aerosol-CCN-clouds, it does not rule out a potential effect from the Global electric circuit and the role of charge build-up at cloud boundaries in the aerosol-cloud droplet recycling processes. However, limited progress has been made in this area over recent years and it is uncertain what magnitude such an effect might have.

It is clear that more work and validation campaigns are needed to improve our understanding of a mechanism involving ions, and to determine the potential implications for climate. CLOUD (an international collaboration of research groups in atmospheric science) will very shortly begin to explore all currently proposed atmospheric mechanisms involving ions with an aerosol chamber at CERN, Geneva, Switzerland (http://cloud.web.cern.ch/cloud/). This will hopefully provide the experimental constraints for developing a robust theory to describe the role of ions in atmospheric processes.

References

Bazilevskaya G. A., Observations of variability in cosmic rays, *Space Sci. Rev.*, 94, 25–38, 2000.

Berndt T., Böge O., and Stratmann F., Formation of atmospheric H2SO4/H2O particles in the absence of organics: A laboratory study, *Geophysical Research Letters*, vol. 33, l15817, doi:10.1029/2006gl026660, 2006

Carslaw K. S., Harrison R. G. and Kirkby J., Cosmic Rays, Clouds, and Climate *Science* 29 November 2002,Vol. 298. no. 5599, pp. 1732 – 1737, DOI: 10.1126/science.1076964, 2002

Eichkorn, S., Wilhelm, S., Aufmhoff, H., Wohlfrom, K. H. & Arnold, F. 2002 Cosmic ray-induced aerosol-formation: first observational evidence from aircraft-based ion mass spectrometer measurements in the upper troposphere. *Geophysical Research Letters* 29. art no 1698. (doi:10.1029/ 2002GL015044), 2002

Ferek, R.J., T. Garrett, P.V. Hobbs, S. Strader, D. Johnson, J.P. Taylor, K. Nielsen, A.S. Ackerman, Y. Kogan, Q. Liu, B.A. Albrecht, and D. Babb. Drizzle suppression in ship tracks. J. Atmos. Sci. 57, 2707-2728, doi:10.1175/1520-0469(2000)057, 2000

Gringel, W., J. A. Rosen, and D. J. Hofmann, Electrical structure from 0 to 30 kilometers, in *Studies in Geophysics: The Earth's Electrical Environment*, pp. 166–182, Natl. Acad., Washington D. C, 1986

Harrison, G., Twentieth century secular decrease in the atmospheric potential gradient, *Geophysical Research Letters*, 29(14), 2002.

Harrison, R. G. and K. S. Carslaw, Ion-aerosol-cloud processes in the lower atmosphere, *Reviews of Geophysics,* 41(3), 2003.

Hoppel, W.A., R.V. Andersen, and J.C. Willet, Atmospheric Electricity in the planetary boundary layer, in Studies in Geophysics: *The Earth's Electrical Environment*, p159-165, Natl. Acad. Press, Washington D.C., 1986.

Horrak U., Salm J. and Tammet H., Bursts of intermediate ions in atmospheric air., *Journal of Geophysical Research,* 103 D12, 13909-13915, 1998

Horrak, U., Salm, J., & Tammet, H.. Statistical characterisation of air ion mobility spectra at Tahkuse observatory: Classifcation of air ions. *Journal of Geophysical Research* D, 105, 9291–9302., 2000

Horrak, Iher, Luts Salm and Tammet, Mobility spectrum of air ions at Tahkuse Observatory, *Journal of Geophysical Research* ,Vol 99, No. d5, pages 10,697-10, 700, May, 1994

Kristjánsson, J.E., A. Staple, J. Kristiansen and E. Kaas: A new look at possible connections between solar activity, clouds and climate, *Geophysical Research Letters*, Vol. 29, No. 23, 2107. 2002

Kulmala M.,Kerminen V. ,Anttila T.,Laaksonen A., and O'Dowd C., Organic aerosol formation via sulphate cluster activation, *Journal of Geophysical Research*, vol. 109, d04205, doi:10.1029/2003JD003961, 2004

Lovejoy, E.R., J. Curtius, and K.D. Froyd, Atmospheric ion-induced nucleation of sulfuric acid and water, *Journal Of Geophysical Research*, 109, D08204, doi:10.1029/2003JD004460, 2004.

Lee S.-H., Reeves J. M., Wilson J. C., Hunton D. E., Viggiano A. A., Miller T. M., Ballenthin J. O. and Lait L. R., Particle Formation by Ion Nucleation in the Upper Troposphere and Lower Stratosphere, *Science* 26 September 2003: Vol. 301. no. 5641, pp. 1886 – 1889, DOI: 10.1126/science.1087236, 2003

Marsh, N.D. and H. Svensmark (2000a): Cosmic Rays, Clouds, and Climate. *Space Science Review*, 94, 215-230., 2000

Marsh, N.D. and H. Svensmark (2000b): Low Cloud Properties influenced by Cosmic Rays. *Physical Review Letters*, bind 85(23), side 5004-5007. 2000

Marsh, N. and H. Svensmark, Galactic cosmic ray and El Nino Southern Oscillation trends in International Satellite Cloud Climatology Project D2 low-cloud properties, *Journal of Geophysical Research-Atmospheres,* 108(D6), 2003.

Marsh, N., Svensmark, H., and F. Christiansen, ISAC Literature Study: Climate Variability Correlated with Solar Activity, *TN-WP103/ESA*, 2005.

Petters, M. D., J. R. Snider, B. Stevens, G. Vali, I. Faloona and L. Russell, 2005: Accumulation mode aerosol, pockets of open cells, and particle nucleation in the remote subtropical Pacific marine boundary layer. *Journal of Geophysical Research*, 111, D02206, 2005.

Roldugin, V. C. and B. A. Tinsley, Atmospheric transparency changes associated with solar wind-induced atmospheric electricity variations, *Journal of Atmospheric and Solar-Terrestrial Physics*, 66(13-14), 1143-1149, 2004.

Rosenfeld, D., Y. J. Kaufman and I. Koren, Switching cloud cover and dynamical regimes from open to closed Benard cells in response to the suppression of precipitation by aerosols, *Atmos. Chem. Phys.*, 6, 2503–2511, 2006

Rycroft, M.J., Israelsson, S., Price, C., The global atmospheric electric circuit, solar activity and climate change. *Journal of Atmospheric and Solar-Terrestrial Physics* 62, 1563–1576. 2000

Svensmark, H. and Friis-Christensen. Variation of cosmic ray flux and global cloud coverage – a missing link in solar-climate relationships. *Journal of Atmospheric Solar-Terrestrial Physics.*, 59: 1225–1232.1997

Svensmark et al,, Experimental Evidence for the role of Ions in Particle Nucleation under Atmospheric Conditions, *Proceedings of Royal Society A, Physical and Engineering Sciences* , 2006 (in press)

Stevens, B., Vali, G., Comstock, R. Wood, K., vanZanten, M. C., Austin, P. H., Bretherton, C. S., and Lenschow, D. H.: Pockets of Open Cells (POCs) and Drizzle in Marine Stratocumulus, *Bull. Amer. Meteorol. Soc.*, 86, 51-57, 2005.

Stevens, B., W. R. Cotton, and G. Feingold: A critique of one- and two-dimensional models of boundary layer clouds with a binned representations of drop microphysics. *Atmospherical Research*, 47-48, 529–553. 1998

Tinsley, B. A., Influence of solar wind on the global electric circuit, and inferred effects on cloud microphysics, temperature, and dynamics in the troposphere, *Space Science Reviews,* 94(1-2), 231-258, 2000.

Usoskin, I. G., et al., Reconstruction of solar activity for the last millennium using 10Be data, *Astron. Astrophys*., 413, 745– 751. 2004

Viggiano A.A., and F. Arnold, Ion chemistry and composition of the Atmosphere, in *Handbook of Atmospheric electrodynamics*, vol.1, ed: H. Volland, p1-26, CRC Press, 1995.

Wood, R. and Hartmann, D. L.: Spatial variability of liquid water path in marine low cloud: The importance of mesoscale cellular convection, *Journal of Climate*, in press, 2006.

Yu, F., Binary H2SO4-H2O homogeneous nucleation rates based on a kinetic quasi-unary model: Look-up tables, *Journal of Geophysical Research,* doi:10.1029/2005JD006358, in press. 2006

Yu, F., and R. P. Turco, Ultrafine aerosol formation via ion-mediated nucleation, *Geophysical Research Letters*, 27, 883. 2000

Yu, F., and R. P. Turco , From molecular clusters to nanoparticles: The role of ambient ionization in tropospheric aerosol formation, *Journal of Geophysical Research*, 106, 4797, 2001