The authors thank the referee for the useful and constructive comments which help to improve the manuscript. Our point-to-point replies to the comments (in blue) are given below (the original comments are copied here in *Italic*). The manuscript has been revised accordingly.

Anonymous Referee #1

This manuscript evaluates the model-predicted aerosol, cloud and radiative-forcing changes between a binary homegeous nucleation (BHN) scheme and a "binary" ion-mediated nucleation (IMN) scheme. The model used is the CAM5 global climate model with online modal aerosol microphysics. The authors find a large change in the properties, particularly the cloud radiative forcings. The authors claim this to be the sensitivity of the model to ionization vs. no ionization. The manuscript has major issues that must be addressed before it is publishable. I cannot recommend that it be published in its current state.

Major comments
- While the CLOUD results in Kirkby et al. (2011) show a strong response of nucleation rates to ion formation rates in the absence of ternary species and the presence of ammonia, new CLOUD results show that the importance of ions on nucleation diminishes in the presence of many organics, and that continental BL nucleation rates cannot be reproduced without these organics. Since in previous studies, this IMN nucleation scheme used in this paper has been shown to generally predict the right order-of-magnitude for nucleation rates, and this IMN nucleation does not account for ammonia and organics, it must be getting the nucleation rates approximately right but for the wrong reasons. Furthermore, since the importance of ions is diminished in the presence of ammonia and organics, the BHN baseline for the ion-free atmosphere will strongly underpredict nucleation. While there is a mention of the important of these ternary species in the conclusions section and a mention that the results might change, the bulk of the paper frames the results as "the effect of ions on nucleation, CCN and clouds", while the results here are a clear overestimate of this effect.

ACPD follow up comments: As far as I know, the new CLOUD results are not published yet. However, in the Kirkby et al., (2011) Nature paper, none of the experiments (except for the one at 248 K) can reproduce the ambient nucleation rates (even when ammonia is added) (see figure 5 and the surrounding discussion). No organics were added to any of these experiments. Furthermore, the ambient nucleation rates in this figure extend to rates that greatly exceed the BL ion-pair formation rates. Regarding the boreal field campaigns, there is a difference between variations in organics at a single site causing variations in nucleation rates versus organics enhancing nucleation rates. In the Kirkby 2011 paper, the enhancements due to NH3 saturate at around 100 ppt. If the organics involved in nucleation are always high enough in concentration that their effect on nucleation is saturated, any variation in organics will not make a difference. This does not mean that the organics are not having a large effect on the nucleation rates.

As the referee pointed out in the following up ACPD comment, the "new CLOUD results" are not published yet. It is hard for us to address the results from un-published work. We are aware of a number of laboratory chamber studies which investigated nucleation processes at close to atmospheric conditions (e.g., Zhang et al., 2004; Berndt et al., 2005; Berndt et al., 2006; Hanson and Lovejoy, 2006; Benson et al., 2008; Metzger et al., 2010; Sipila et al., 2010; Enghoff et al.,

2011; Kirkby et al., 2011) but the results from various studies differ significantly (sometimes contradict each other), probably as a result of different levels of contamination (Kirkby et al., 2011) and/or sampling issues (Sipila et al., 2010). We would like to emphasize that laboratory results do not always reflect what occur in the real atmosphere and have to be confirmed or reproducible by other research groups. Further researches are apparently needed to reconcile these differences and assess the laboratory results (including the new CLOUD results) against field measurements.

While the role of organics in growing freshly nucleated particles is well established, their exact role in promoting initial formation of critical clusters remains to be clarified. For example, both Janson et al. (2001) and Sellegri et al (2005) concluded that, based on measurements made during two different boreal forest field campaigns (BIOFOR and QUEST), the oxidation products of terpenes were not the primary nucleating species observed at Hyytiälä. The main reasons for their conclusion include: (1) that the concentrations of the terpenes and their oxidation products where higher at night when no nucleation was observed; (2) that organic oxidation products were not significantly elevated during event days compared to non-event days based on the results from the BIOFOR campaign; and (3) that organic compounds including terpenes are generally lower during event days compared to non-event days based on QUEST data.

We agree with the referee that, "if the organics involved in nucleation are always high enough in concentration that their effect on nucleation is saturated, any variation in organics will not make a difference". However, it remains to be established if this is the case in the real atmosphere. Fortunately, we have other measurements that can provide useful insights about the dominant nucleation processes and species.

As pointed out in the paper, Yu and Turco (2011) demonstrated that the state-of-the-art multi-instrument field measurements (including overcharging ratios of freshly nucleated particles) taken in a boreal forest appear to strongly support the dominance of IMN mechanism, which is further supported by the most recent cluster mass spectrometer measurements at the site showing the absence of small neutral clusters (Jokinen et al., 2012). These results appear to indicate that neutral nucleation process may be not as important in the real atmosphere (at least in the boreal forest) as shown in various laboratory chamber studies. It remains to be studied if the new CLOUD results the referee mentioned (i.e., "the importance of ions on nucleation diminishes in the presence of many organics") are consistent with the field measurements taken in the boreal forest where a lot of organics are known to be around. If neutral nucleation (involving ammonia and organics) is significant in the boreal forest (where ammonia concentration is generally well above 100 ppt and concentrations of condensable organics are known to be high), we expect to see the undercharging of freshly nucleated particles and abundance of small neutral pre-nucleation clusters. As mentioned earlier, so far the measurements of overcharging ratios and concentrations of small neutral clusters appear to indicate the opposite.

We agree that the exact mechanisms of new particle formation in the atmosphere remain to be further investigated. The main objective of this manuscript is to assess the possible effect of ionization based one neutral and one ion-mediated nucleation schemes. We would like to emphasize that both BHN and IMN schemes used in this study are constrained by laboratory measurements (Yu, 2010). While species other than H_2SO_4 and H_2O may enhance neutral nucleation, they may enhance ion-mediated nucleation as well. We have added some further discussions about the uncertainty in nucleation processes and implications.

- Why is the a solar modulation in ion formation rates not included here? It says that it is being saved for future work, but the current paper is extremely thin (1 table, 2 figures and only 2 simulations in an ACPD paper). There is really no good reason why ion formation perturbation runs shouldn't be included here. I'd feel this comparison may be somewhat better represented by the model than the present comparison since the BHN lower limit is unrealistically low for simulations without ions (see above), and the IMN scheme generally predicts ok nucleation rates (at least when its in the GEOS-Chem model). This paper would be far more complete and informative if it included these simulations.

We have carried out additional simulations and now the effect of ionization perturbation associated with solar cycle is included. Based on the present CAM5 simulation, the 5-year mean impacts of solar cycle induced changes in ionization rates on CCN and cloud forcing are small (~ -0.02 W/m^2) but have larger inter-annual (from -0.18 to 0.17 W/m^2) and spatial variations. It appears that positive and negative perturbations cancel each other and it is unclear the underlying mechanism of such cancellation is unclear. Further research with much longer simulations is needed to assess the impact of model internal variations and uncertainties on the solar cycle signals.

- As far as I know, this is the first time that the IMN scheme has been tested in the CAM model with the MAM3 microphysics scheme. This is a very different scheme than the IMN scheme in GEOS-Chem with APM (which includes a large number of size bins for the growing nucleated particles). Please add evaluation of CN and CCN concentrations.

This is a good suggestion. We have added evaluation of CCN concentrations which are directly relevant to the aerosol indirect radiative forcing. Because of the lack of the nucleation mode in the present version of CAM5-MAM3 and the dominance of CN by small nucleation mode particles in many regions, it is hard to directly compare observed CN (particles larger than ~ 3 or 10 nm) with CAM5-MAM3 predicted total CN (summation of particles in Aitken, accumulation, and coarse modes). Therefore, we didn't evaluate CN concentrations in the present study.

- I am very concerned that there is no nucleation mode in MAM3 and that Kerminen and Kulmala is used for growth all the way until the Aitken mode. What upper diameter is used in Kerminen and Kulmala in this work? This may lead to large errors since Kerminen and Kulmala assumes that the current growth and coagulation conditions are constant and instantly grows the particles to the upper size. Nucleation rates positively correlate with high growth rates and lower coagulation sinks, and growth rates are likely to decrease and coagulation rates are likely to increase while growing to larger sizes. Thus, instant growth to sizes larger than a few nm will lead to an overprediction in the number of nucleated particles that survive growth to these larger sizes. Please add some evaluation of this bias or add a nucleation mode.

The upper diameter used with the Kerminen and Kulmala parameterization is 12 nm. We agree that not treating the nucleation mode explicitly leads to less accurate simulation of the aerosol distribution. However, a mode treatment of the submicron aerosol (with no nucleation mode) has been used in previous studies (e.g., Wang et al., 2009; Wang and Penner, 2009). While such a treatment appears to overestimate total particle number (Wang et al., 2009; Anttila et al., 2010), the effect on larger size particles (and CCN) is less (Wang et al., 2009) as many of the smaller

Aitken mode particles are also lost by coagulation. Also, growth of new particles by condensation of organics is not included in our simulations, causing an underestimation of the contribution of new particle formation to CCN. As we show in Section 3.1 for the comparison of predicted and observed CCN, the present model overall under-predicts CCN, especially in the regions where the contributions of nucleated particles to CCN are expected to be significant. We have included discussion of these points in the revised manuscript.

Specific comments
page 17351 line 2: "coagulation (Aitken and accumulation modes)". Do these modes not coagulate with the coarse mode? Ignoring coarse-mode self coagulation is probably fine, but ignoring coagulation losses of the smaller particles with the coarse mode will lead to an overprediction in the probability of a nucleated particles growing to a CCN size.

Coagulation of Aitken and accumulation mode particles with coarse mode particles is not treated. The coagulation rate for 20-40 nm diameter (Aitken mode) particles with a 2 μm (coarse mode) particle is about 10 times greater than that with a 0.2 μm (accumulation mode) particle, but the accumulation mode number concentration is typically several hundred times greater than the coarse mode number, so Aitken-accumulation coagulation is generally much larger than Aitken-coarse. The coagulation loss of freshly nucleated particles, which is calculated with the Kerminen and Kulmala (2002) parameterization, does include loss to the coarse mode.

Page 17351 line 6: You describe cloud-droplet activation, but what about cloud microphysics? One of your outputs in the paper is precipitation, so the treatment of cloud microphysics is extremely important. Is this 1- or 2-moment microphysics? How is autoconversion treated? How are the treatments of warm and cold clouds different. Do CCN directly affect cold cloud microphysics?

We have added the following description:

Stratiform cloud microphysics is represented using the double-moment formulation of Morrison and Gettelman (2008), which predicts number and mass mixing ratios of cloud droplets and ice crystals and diagnoses number and mass mixing ratios of rain and snow particles. Autoconversion of droplets to rain depends on droplet number according to Khairoutdinov and Kogan (2000). Droplet nucleation depends on updraft velocity and the number, mean radius, and mean hygroscopicity of all aerosol modes according to Abdul-Razzak and Ghan (2000). Homogeneous and heterogeneous nucleation of ice crystals depends on aerosol size distribution through both homogeneous freezing of haze particles and cloud droplets and heterogeneous freezing of cloud droplets induced by mineral dust (Liu et al., 2007); Ghan et al. (2012) showed that homogeneous nucleation in CAM5 produces a significant longwave aerosol indirect effect. Liu et al. (2007) and Gettelman et al. (2010) describe the treatment of mixed-phase cloud microphysics, including the Bergeron-Fineisen process.

Page 17352 line 1: I agree that there are uncertainties in primary sulfate and in how ions might affect the number particles from sub-grid nucleation. However, omitting primary sulfate reduces

the number of CN, which makes your CCN more sensitive to regional nucleation. This needs to be discussed.

The key issue is that "primary" sulfate particles are not primary. They are secondary particles formed through nucleation in sub-grid plumes. Omitting primary sulfate does not necessarily reduce the number of CN if a suitable nucleation scheme is used. Luo and Yu (2011) showed a compensation effect of nucleation to primary sulfate emission. They found that adding primary sulfate emission does not improve the agreement between simulated and observed annual mean number concentrations of particles > 10 nm at 21 stations around the globe. We have added some discussion on this in the revised manuscript.

Page 17354 line 3: why is there a supplement with 1 figure and 1 paragraph? The paper is currently very short, just put this figure and text in the paper.

We have now put the supplement figure and text in the paper.

Page 17355 line 13: What does "_116%" represent here? This doesn't seem to be smaller than 45%. Also, I think you meant to say "is expected to BE relatively smaller".

Thanks for pointing this out. "116%" is a typo, should be "16%". We have changed the line to: " is expected to be relatively smaller (uncertainty ~16%)"

Page 17355 line 17: "which dominate precipitations." Total precipitation volume amount? Precipitation rates? Precipitation frequency?

Total precipitation volume amount. This is clarified in the text.

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