We would like to first thank the anonymous reviewer for the comments to help make better our manuscript. We address the reviewer's comments below. The original comments are in **bold font** and our responses are in *italic font*.

Abstract, L13. Given that L2.0 AERONET data comparisons against the aircraft data are limited to 1 point, the SSA conclusion must be removed. Figure 3 must also be removed as scatter plots with 1 point on them are not relevant or informative.

The conclusion about the discrepancies between the aerosol properties retrieved from AERONET and from in-situ aircraft measurements has been rewritten, so now it states that the discrepancies between the aerosol properties retrieved from AERONET and from in-situ aircraft measurements are smaller for the AOD, while the biggest discrepancies are for the other derived aerosol properties.

Figure 3 has been removed.

Introduction, Para 1: Given our current understanding of aerosol direct radiative forcing and WMGHGs (IPCC, 2007), it is not correct to same that they are the same magnitude. The aerosol direct effect has been assessed as -0.50 ± 0.4 Wm⁻² (IPCC, 2007, Chapter 2), while the radiative forcing from well mixed greenhouse gases is assessed as $+2.63\pm0.26$ Wm⁻² (IPCC, Chapter 2). Please tone this down.

The reviewer is correct, on a global scale, but on a regional scale the aerosol forcing can be of similar or greater magnitude. Nevertheless, this paragraph has been rewritten, so now it states that aerosols affect the Earth's radiative balance both directly (by scattering and absorbing solar radiation) and indirectly (through their action as CCN in cloud formation), and that they have been identified as one of the greatest sources of uncertainty in the interpretation of the Earth's climate, on both global and regional scales.

P29005, L6. These simple estimates of the direct radiative forcing are only relevant for cloud-fee skies. Insert 'cloud-free sky' before 'direct aerosol radiative forcing'.

Done.

L15. Sentence is too long. however -> However

Done.

P29006, L3. AOD is a wavelength dependent quantity, suggest inserting a subscript lambda after AOD, i.e. AOD_{λ}

We have inserted a sentence in section 1 clarifying the wavelengths: "Unless otherwise noted, values of spectral aerosol optical properties, including AOD, discussed in this paper all refer to a wavelength of 550 nm."

P29007, L13. Having a heater upstream of the nephelometers will certainly reduce the relative humidity. However, it may also cause volatalisation of some aerosol particles, particularly nitrate

A sentence has been added in section 2.1 reporting the calculated loss of light scattering in the heated nephelometer as 1-8%, based on ammonium nitration volatilization experiments (Bergin et al., 1997) and the seasonal contribution of ammonium nitrate to light scattering based on IMPROVE measurements at Bondville (Hand et al., 2011).

Bergin, M. H., Ogren, J. A., Schwartz, S. E., and McInnes, L. M.: Evaporation of ammonium nitrate aerosol in a heated nephelometer: Implications for field measurements, Environ. Sci. Technol., 31, 2878-2883, 1997.

Hand, J. L., Copeland, S. A., Day, D. E., Dillner, A. M., Indresand, H., Malm, W. C., McDade, C. E., Moore, C. T., Pitchford, M. L., Schichtel, B. A., Watson, J. G., 2011: IMPROVE, Spatial and Seasonal Patterns and Temporal Variability of Haze and its Constituents in the United States, ISSN 0737-5352-0787. Available at http://vista.cira.colostate.edu/improve/Publications/Reports/2011/2011.htm.

P 29008, L5. I note that the flights were performed between June 2006-October 2008. Kasatochi erupted on 7-8 August 2008 injecting ~ 1.5TgSO2 into the stratosphere (Kravitz et al., 2010). The dynamical evolution of the plume was such that the plume would have reached Illinois quite shortly after (mid-August). Here's a figure from Kravitz et al, with a line at 40N. You'll see a measured AOD of around 0.005 (at 750nm) on a zonal average. However, the AOD will be many times (factor of 10-100 greater) this in the centre of the plume just subsequent to the eruption :- Fortunately you should have missed the majority of the impacts of

the plume. It would be worth noting that the time period that you've chosen is pretty much outside the period of significant volcanic stratospheric AOD as this as one of the possibilities that you raise is aerosol above the maximum flight level of the aircraft.

This issue is addressed in section 4.3.

Yu et al. (2010) reported seasonally-averaged profiles of aerosol extinction coefficient for the Eastern US in 2007, and showed that aerosols between 5 and 10 km asl made a negligible contribution to AOD for that year. An examination of the CALIPSO 'level 3' mean extinction profile for June 2006-October 2008 in a 2 degree latitude by 5 degree longitude grid box, roughly centered on Bondville, also reveals negligible extinction above 4.6 km (J. Tackett, personal communication).

Yu, H., Chin, M., Winker, D. M., Omar, A. H., Liu, Z., Kittaka, C., and Diehl, T.: Global view of aerosol vertical distributions from CALIPSO lidar measurements and GOCART simulations: Regional and seasonal variations, J. Geophys. Res., 115, D00H30, doi:10.1029/2009JD013364, 2010.

P29008. L5 – L17. Are you sure that you were flying on flight levels? There is a technicality here that aircraft at low levels tend to fly on QNH rather than the standard atmosphere and 1013hPa surface pressue. If you are really flying on flight levels throughout then a more detail is required. Why? Because your dz determined from your pitot static tube pressure will be a function of the surface pressure. For example, if the 'real' pressure adjusted to ASL is 1030hPa rather than 1013 then you'll actually be flying at 1117m rather than 1000m (hydrostatic approximation with an assumed scale height of 7km) – you'll have to add about 10% or so onto the dz. This is potentially a source of error/bias in your calculations owing to you integrating your scattering (product of scattering * dz) if you are really flying flight levels as you suggest in your manuscript. Clarification is required.

Technically, "flight levels" (FL) don't begin until 18000 feet asl, i.e., FL180 is 18000 feet, so altitudes below 18000 feet don't have corresponding flight levels values. Therefore, since all flight operations were performed below 18000 feet asl, we were not actually flying on flight levels. Thus, we have changed the term "flight levels" to "flight altitudes".

Moreover, when pilots climb past 18000 feet asl, they adjust their altimeters to a sealevel pressure of 29.92 Hg. In other words, everybody uses the standard atmosphere above 18000 feet. Below 18000 feet, pilots set their altimeters on the ground to match the field elevation, or in-flight to a setting provided by air traffic controllers. In our case, changes in surface pressure are minimal for the distances we covered and the duration of our flights, so we expect the altitudes reported by our altimeter, set to the elevation of Willard Field near Bondville, to be accurate.

P29011, L 8-17. Fig 3 should be removed as it is unnecessary.

Done.

Section 4.1. I agree that there are possibly some problems related to relative humidity models etc. Having had a look at the hygroscopic growth factors from Figure 10, it is interesting to note that the Koloutsou-Vakakis et al (2001) parameterisation is strikingly similar to other measurements using tandem nephelometers. Haywood et al (2008, QJRMS) use airborne and surface based systems over the UK. The airborne system (green, light blue and dark blue) gives growth factors of around 1.4, 1.5, and 1.7 at RHs of 80, 85, and 90% RH, which seems strikingly similar to those shown in Figure 10. This study could be referred to give extra support to the K-V measurements. The real problem of non-linearity in the f(RH) versus RH comes when you get above 90%. At these high RHs, the ground based system (nephelometer combined with a visiometer) start to increase rapidly.

The reference Haywood et al. (2008) has been added to the text in order to give extra support to the use of the Koloutsou-Vakakis hygroscopic growth adjustment.

P29014, L 29. Only 59% of the particle mass is SO4 or VOC. Going back to the V-K et al paper, here is another 7% identified as nitrate. This could cause some trouble as the nitrate/ammonia/water is pretty unstable, particular at high temperatures. Heating prior to the nephelometer is likely to dissociate ammonium nitrate back to gas phase nitric acid and ammonia. If the V-K paper is representative, you could lose 7% of your aerosol anion mass (more aerosol total mass) simply by volatalising your nitrate. I think that this potential problem should be emphasised in the discussion, particularly as there is an author for both

studies on the author list: Bergin et al (1997), Evaporation of Ammonium Nitrate Aerosol in a Heated Nephelometer: Implications for Field Measurements

A sentence has been added in section 2.1 reporting the calculated loss of light scattering in the heated nephelometer as 1-8%, based on ammonium nitration volatilization experiments (Bergin et al., 1997) and the seasonal contribution of ammonium nitrate to light scattering based on IMPROVE measurements at Bondville (Hand et al., 2011).

Bergin, M. H., Ogren, J. A., Schwartz, S. E., and McInnes, L. M.: Evaporation of ammonium nitrate aerosol in a heated nephelometer: Implications for field measurements, Environ. Sci. Technol., 31, 2878-2883, 1997.

Hand, J. L., Copeland, S. A., Day, D. E., Dillner, A. M., Indresand, H., Malm, W. C., McDade, C. E., Moore, C. T., Pitchford, M. L., Schichtel, B. A., Watson, J. G., 2011: IMPROVE, Spatial and Seasonal Patterns and Temporal Variability of Haze and its Constituents in the United States, ISSN 0737-5352-0787. Available at http://vista.cira.colostate.edu/improve/Publications/Reports/2011/2011.htm.

P29017, L1: Typo 'phygroscopicity arameterizations'.

Corrected.

Section 4.2. It is troubling that the AAO measures 10-20% less scattering than measurements made at the ground. Generally, I'd believe ground based measurements more than aircraft measurements which are notoriously difficult to make accurately. If there is a discrepancy of this magnitude, this could pretty much explain the discrepancy between the AERONET and the AAO measurements. This again could be pointing the finger at nitrates being volatalised in the AAO nephelometer.

In Section 4.2 we have observed that AAO measures 10-20% less scattering than that measured on the ground, which can be explained by the fact that the aircraft inlet excludes larger aerosols or because there is less aerosol at the lowest flight level. Although our analysis of these two hypotheses is inconclusive, and we cannot reject any of them, they can only account for a small part of the discrepancy between the AOD from AERONET and AAO.

However, in Section 4.1 we have observed the high influence that the selected adjustment to ambient RH of the scattering coefficient has over the comparison between the AOD from AERONET and AAO (e.g., see Table 3). Thus, we can affirm that the largest portion of the observed AOD discrepancy is probably due to an incorrect hygroscopic growth parameterization.

The issue is not nitrate volatilization, as discussed above that only contributes a loss in scattering of 1-8%.

Section 4.3. The point made earlier about the aircraft operating on Flight Levels needs to be addressed.

As explained before, "flight levels" (FL) technically don't begin until 18000 feet asl, so altitudes below 18000 feet don't have corresponding flight levels values. Therefore, since all flight operations were performed below 18000 feet asl, we were not actually flying on flight levels. Thus, we have changed the term "flight levels" to "flight altitudes".

Section 4.5. One thing to note here is that the site is under an area with extremely high air traffic. In fact – this plot from IPCC suggests it is in an area which is a global maximum (IPCC, 1999, see below). This means that there will be significant aircraft emissions over the site at cruise altitude. This is also an area where contrails and contrail induced cirrus are very prevalent. Could the discrepancy be caused by the SKYRAD and AERONET methods including very disperse thin subvisible contrail cirrus? The optical depth offset of ~0.05 and the AERONET detection of more large particles (Fig 2, 4,5) might be related to this. Are the cloud screening algorithms really going to be able to detect and reject sub-visible cirrus with a visible optical depth of 0.05?

We have added a paragraph to section 4.5 discussing the potential for contamination by cirrus clouds not removed by the AERONET Level 2.0 screening algorithm.

Recent studies have quantified the contribution of tropical cirrus clouds to AERONET AOD measurements (Chew et al., 2011; Huang et al., 2011) as a bias of 0.03-0.05 in AOD (at 500 nm wavelength) and a shift of the Ångström exponent and fine mode fraction to smaller values (i.e., more large particles). These are clouds that were not removed by the AERONET Level 2.0 screening criteria. While the same criteria were applied to derive the Level 2.0 AOD data at Bondville, supporting lidar measurements are not available to assess whether the findings from the tropics are applicable to the AOD observations at Bondville. If the tropical findings apply to Bondville, cirrus clouds could explain much of the offset of 0.05 observed in the AOD regression analysis (section 3) as well as the observed differences in Ångström exponent and fine mode fraction.

Chew, B. N., Campbell, J. R., Reid, J. S., Giles, D. M., Welton, E. J., Salinas, S. V., and Liew, S. C.: Tropical cirrus cloud contamination in sun photometer data, Atmos. Environ., 45, 6724-6731, 2011.

Huang, J., Hsu, N. C., Tsay, S.-C., Jeong, M.-J., Holben, B. N., Berkoff, T. A., and Welton, E. J.: Susceptibility of aerosol optical thickness retrievals to thin cirrus contamination during the BASE-ASIA campaign, J. Geophys. Res., 116, D08214, doi:10.1029/2010JD014910, 2011.

Section 4.9. I don't think that anything can be inferred about the SSA given the paucity of the data.

We have changed the text to state the difficulty of verifying the AERONET retrieval algorithm at a site that is not highly polluted.

Conclusions. I'd like to see some more acknowledgement of the potential role of nitrate aerosol (e.g. Bergin et al. 1997), the role of stratospheric aerosol (Kravitz et al 2010, but also perhaps Solomon et al, Science, 2011, below), and the fact that the site is one of the potential hot-spots for sub-visual cirrus induced by aircraft. SSA should not be compared against given you've only 1 data point. It would be very useful too if the authors could suggest what additional measurements (either from the surface or from the aircraft) would be necessary for better determining the reasons behind the discrepancies.

The calculated loss of scattering due to nitrate volatilization is so small that it does not warrant highlighting in the Conclusions. We have not drawn conclusions based on the single SSA comparison point, other than to state that it points out the difficulty of validating AERONET SSA retrievals in areas that are not highly polluted. The possibility remains that there were sub-visible cirrus above the site, but observations to explore this possibility are lacking.

We have added a clause to the sentence where we conclude that the largest part of the discrepancy is probably due to an incorrect adjustment of light scattering to ambient

RH: "improved measurements of the aerosol hygroscopic growth factor would be needed to confirm this diagnosis."