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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Spatial features of rain frequency change and pollution and associated aerosols" *by* Y. Lin et al.

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Interactive comment on "Spatial features of rain frequency change and pollution and associated aerosols" by Y. Lin et al. Anonymous Referee #2 Received and published: 28 March 2011 In this manuscript, the authors examined changes in rain frequency and pollution and associated aerosols by using multi-satellite observations over East Asia during 1998-2009 (AOD from MODIS is from 2000-2009), with a focus on the spring season. They found that the change in rain frequency is associated with changes in pollution produced aerosols and long-range transport mineral dust. Cloud fraction from satellite observations and NCEP reanalysis data are also used to establish the causality between the change in rain frequency and changes in aerosols. The topics and results are interesting, and substantial efforts are made to analyze data and in revising

the manuscript (I noted that this is a resubmission of Lin et al., 2010, ACPD). However, uncertainties in satellite data are not acknowledged, and the causality between the change in rain frequency and changes in aerosols implied by the authors are still not adequately supported by their data. The short time period and large variations from year to year are also troubling. I would recommend the publication of the paper after my following concerns are addressed:

Lin et al.: We thank the reviewer for very thorough and constructive comments, which have helped to improve the quality of the paper in both sciences and writing. Below are our responses to those comments. The response follows each comment.

Major comments: 1. Uncertainties in satellite retrievals. The authors did not adequately acknowledge the uncertainties in satellite data. One example is about the fine mode AOD. The total AOD from MODIS does not show significant trend during 2000-2009 over Shanghai (Fig. 2), but the fine mode AOD does. In fact, the decreasing trend in the fine mode AOD over polluted regions is the key finding in this paper. However, the retrieval of the fine mode AOD is much less reliable than the total AOD, especially over the land areas, as this is the case in the current study. Uncertainties in the retrieval of the fine mode AOD are not discussed at all in the current manuscript. It is equally important for the authors to discuss whether these satellite data is suitable for trend analysis. For example, though the same instrument and the same algorithm are applied for measuring MODIS AOD and fine mode AOD, how about changes in surface properties or any drift in instrument accuracy that may affect the retrievals during 2000-2009? The same argument can be also applied to TRMM PR measurements. These issues should be discussed in detail, and the authors should also more prominently acknowledge these issues in abstract and conclusion. Lin et al: Yes, there are several issues, such as sampling bias (Zhang and Reid, 2009), and perturbations in sensor calibration (Levy et al., 2010; Remer et al., 2005; Zhang and Reid, 2010), may induce the artificial tendency in the MODIS AOD product and introduce biases to the ten-year trend of AOD. We added some discussion about these issues to the abstract and conclusion.

Ichoku et al. (2002) stated that the mean AOD from MODIS aerosol retrievals compared favorably with AERONET, and their standard deviations reveal some influence of surface effects on the MODIS aerosol retrievals over land, especially when AOD less than 0.2. However, the excellent agreements between Terra MODIS and Aqua MODIS AOD retrievals (Remer et al., 2006) and MODIS and MISR AOD retrievals (Kahn et al., 2009) add overall confidence to the climate-scale MODIS products, and suggest that the MODIS AOD retrievals are likely reliable. This conclusion is further reinforced by the long-term statistical validation of MODIS AOD against AERONET AODs (Remer et al., 2005). Zhang and Reid (2009, 2010) examined biases in satellite AOD trend analysis due to radiometric calibration, cloud contamination, and sampling biases, concluding that it is reasonable to use monthly mean AOD values from Terra/Aqua MODIS to study the decadal aerosol optical depth trend. Levy et al. (2010) estimated an overestimation of $\sim$0.005 prior to 2004 and an underestimation of similar magnitude afterwards based on the comparison of MODIS and AERONET. However, as the trend of AOD over Asia is generally increasing, such biases with time may change the magnitude of the trend but not the pattern of special-temporal changes of AOD.

The retrieval of fine mode AOD is much less reliable than total AOD (Hyer et al., 2011; Kahn et al., 2009; Remer et al., 2008). However, since the trend of AOD was used here as a linkage between gaseous pollution and precipitation. Many previous studies showed a steady increasing trend of NO2 (Richter et al., 2005, and others). Considering that NO2 and aerosol are often from same sources and NO2 acted as an important precursor of secondary formation of aerosol (elaborated in the answer of question below), the consistency of increasing trend of NO2 and fine aerosol may add the confidence that the trend of fine AOD over East China may be true.

Since launch in December 1997 till 2009, all components of the PR had shown no instrument issues (Yasuhisa Iida et al., 2010). The long-term hardware stability of the PR is verified from the small monthly variation (less than 0.05 dB) of sea surface

scattering cross sections in no rain conditions (K. Okamoto et al., Long term trend of ocean surface normalized radar cross section observed by TRMM precipitation radar, paper presented at XXVIIth General Assembly, Int. Union of Radio Sci., Maastricht, Netherlands, 2002). The orbit of TRMM was boosted from 350km to 402.5 km in August 2001. However, by comparing the PR data with surface rain gauges, DeMoss and Bowman (2007) and Nakazawa (2009) stated that the change in the PR bias due to the orbit boost is not statistically significant. Furthermore, we used the ratio of raining pixels to total sampling pixels. Using such a relative parameter also minimizes the systemic bias and retrieval uncertainties of PR rain rate retrievals.

2. The short time period Here I want to underline the point made by the previous reviewer #2 about the short time period used in this study. The large variations in rain frequency from year to year make the short time period especially troubling. As shown in Figure 2, if we only use the rain frequency data from 2000 to 2009, the same period as the MODIS AOD observations, we will not see any significant trend in rain frequency during this period over Shanghai (Fig. 2). The decreasing trend in rain frequency over Shanghai in this paper is mainly caused by the large rain frequency in the first two years examined (1998 and 1999). I would expect the trend in rain frequency in Figure 3 will also change if the authors exclude 1998 and 1999 in their analysis. Statistically significant changes in rain frequency are another key finding of the current manuscript. Lin et al.: Many studies based on a longer term surface rain gauge measurements show similar precipitation trends (Huang et al., 2007; Qian et al., 2010). However, those long-term surface rain gauge measurements are sparse. As aerosols and precipitation have large spatial and temporal variability, remote sensing from satellites delivers the most reliable spatial information about their regional and global distribution. The TRMM PR is only satellite sensor that directly measures precipitation. As TRMM PR was launch on November 1997 and provides data from December 1997 to present, a time series of twelve years (from 1998-2009) is the most long term we may use to study precipitation properties from TRMM PR. Although the interannual variability of precipitation is large and TRMM PR measurements are not longer than we would hope, the better

spatial coverage of TRMM PR provides an additional dimension, in terms of pollution-aerosol-precipitation interaction. Hence, we focused our investigation on the spatial features of rain frequency change induced by pollution and associated aerosols from the spatial-temporal perspective by utilizing multi-satellite observations over East Asia. Also, the basic trends in precipitation during the overlap period between long-term surface rain gauge measurements and TRMM PR observation are consistent with each other, which give us the confidence that extending the period of observation for a few years may not change our conclusions. Certainly we will extend our study when more years of TRMM PR measurements become available.

3. The causality between changes in aerosols and changes in rain frequency. The causality issue was raised by previous reviewers for Lin et al., 2010, ACPD. The authors took a substantial effort to address this issue in the revision, but to my opinion, it is not quite successful yet. For example, one key argument the authors used is the spatial distribution of correlation coefficient between rain frequency and AOD (Figure 7). But the area of stronger negative correlation does not really overlap with the regions with strong reduction in rain frequency (Figure 3) (See below for my comments on this). Another example is cloud fraction (Figure 4). As pointed out by previous studies, the positive correlation between cloud fraction and AOD in satellite observations can be caused by many non-indirect-effect factors (See below for my comments on this). It will be more appropriate to examine the trend of cloud droplet effective radius and cloud liquid water path from MODIS during 2000-2009. Change in droplet effective radius will be the first response of clouds due to aerosol indirect effects, and changes in liquid water path will then be expected from the 2nd aerosol indirect effects. Lin et al.: Thanks for the suggestion. The correlation of fine mode AOD with warm cloud droplet effective radius of three regions over China was further compared (Figure S1) and added to the revised manuscript as Figure 4. The liquid cloud droplet effective generally decreases with the fine mode AOD over the Yangtze River region and Yellow River Region, while there is no significant correlation in the background region. Furthermore, the cloud effective radius over the polluted regions is generally smaller than

the background region.

Please see attached Figure S1: The liquid cloud droplet effective radius and fine AOD over Yangtze River Region(30-34°N, 118-122°E, grid over ocean is excluded), Yellow River Region(32-36°N, 111-115°E), and background region (32-36°N,100-104°E) with the pristine atmosphere

The temporal trend of cloud effective radius over India- Myanmar is also added to Figure 5. Ice could effective radius in this region shows a decreasing trend. If the cloud effective radius in 2000 is excluded, the decreasing trend would be much more significant.

However, the correlation between liquid cloud water path with fine AOD in China, and the temporal trend of cloud water path over the India-Myanmar region are all insignificant.

Please see attached Figure S2: The trend of ice cloud fraction and ice cloud effective radius over India-Myanmar region.

Given the short time period (10 years), the larger noise and small signal in the observationtal data, I doubt the authors can ever firmly establish the causality between changes in aerosols and changes in rain frequency. Given these challenges, it is important for the authors to more prominently acknowledge the correlation study nature of this paper, and to caution the readers for the causality in both the abstract and the conclusion. Lin et al.: Thanks a lot for the suggestions. The uncertainty of satellite data was acknowledged in the abstract and conclusion in the revised manuscript. The abstract and conclusion was further revised to make it clearer that the paper is an investigation of the spatial and temporal correlations between loading of pollutants and rainfall change. The possible links are speculated in this paper but need further evidence from more robust statistical study, specific case study, and detailed modeling investigation and so on.

Specific comments: P. 8748, abstract: the abstract should acknowledge the uncertainty in satellite data, and cautions the readers about the causality, since the causality is not adequately supported by their data yet. Lin et al.: Thanks for the comments. The abstract and conclusion was revised according to the comments.

P. 8750, l. 20: please specify the months included in the spring season. Lin et al.: The months (March, April and May) included in the spring season was specified in the revised manuscript.

P. 8751, l. 3: Is the rain rate date used in Figure 1 and Figure 2 the same? If it is the same, why the rain rate is larger in 1999 than in 1998 in Figure 1, but it is larger in 1998 than in 1999 in Figure 2? Lin et al.: Thanks a lot for the question. Yes, there were some difference between Figure 1 and Figure 2 in the original manuscript. The rain rate data used in original Figure 1 included all four seasons in a year, while in original Figure 2, only the rain rate in spring was used. The rate rain data in original Figure 1 was based on a $1^\circ \times 1^\circ$ averaged value, while original Figure 2 was based on a $0.5^\circ \times 0.5^\circ$ averaged value. We changed both Figures 1 and 2 in the revised manuscript. The revised Figure 1 has minor ticks in the x-axis to make it more readable. The revised Figure 2 uses the same averaged domain as the revised Figure 1 ($1^\circ \times 1^\circ$). The trend was little changed, especially the trend of rain frequency. The comparisons of the averaged rain frequency and rain amount of $1^\circ \times 1^\circ$ and $0.5^\circ \times 0.5^\circ$ grids at the site are shown in Figure S3. The much smaller change of rain frequency may collaborate that rain frequency is a parameter which may minimize the systemic bias and retrieval uncertainties of PR rain rate retrievals.

Please see attached Figure S3: The comparison of the averaged rain amount and rain frequency of $1^\circ \times 1^\circ$ and $0.5^\circ \times 0.5^\circ$ grids

P. 8751, l. 7: What is the unit of PR and Gauge in Table 1? Lin et al.: The unit of PR and Gauge in Table 1 are "mm/per season", which was added to the title of Table 1.

P. 8752, l. 24: The authors stated that "the dramatic increase in NO2 concentration

implies a substantial enhancement of atmospheric aerosol loading.' High NO2 concentrations does not necessarily lead to high aerosol loading. As shown in Zhang et al. (2009) (Table 2), though NOx emission increased substantially from 2001 to 2006, the emission of PM2.5 particles increased quite moderately from 2001 to 2006. Lin et al.: The ambient aerosol loading is contributed by both primary emissions and secondary products from photochemical reactions. The significant correlation between NO2 and AOD was detected in various regions in the world, especially over industrial regions (Veefkind et al., 2011). NO2 not only shares the common emission sources with aerosol, such as fossil fuel combustion, but also acts as an important precursor of atmospheric aerosol.

Due to the technology penetration within industry sectors and the implementation of new emission standard, the emission factor of particles decreased, especially for the coarse particles (Lei et al., 2010; Zhang et al., 2009). As a result, the increase of particle emissions was much less than what they would have been. While on the other hand, the emission factor of NO2 was little changed (Zhang et al., 2009), that is why the increase magnitude of NO2 was much larger than particles.

However, a large part of anthropogenic aerosol comes from the secondary transformation, especially in urban areas. Song et al. (2006) reported that secondary products and the emissions from coal combustion and biomass burning dominated PM2.5 in Beijing. Miracolo et al. (2010) reported that the photo-oxidation created substantial secondary particulate matter (PM) greatly exceeding the direct PM emissions.

NO2 is an important precursor of secondary formation of aerosol. The high SOR (SOR =nSO42-/(nSO42- + nSO2)) and NOR (NOR =nNO3-/nNO3- + nNO2) could be used to evaluate the degree of atmospheric conversion from SO2 to SO42- and from NO2 to NO3-. The high values of SOR and NOR over urban areas indicate the formation of secondary aerosols due to conversion from gaseous precursors is significant and important (Lin, 2002). NO2 could also play an important role in the formation of secondary organic aerosol. Thus, it may safe to say that "the dramatic increase in NO2 concentration implies a substantial enhancement of atmospheric aerosol loading."

P. 8753, l. 1: It is difficult to claim that AOD increases in recent year from Figure 2. If we exclude the first 2 year data and start from 2002, there is not evident increase in AOD. Lin et al.: Because the limited length of the dataset and large interannual variability, the increasing trend of total AOD is not that obvious in Shanghai if excluded the first 2 year data. However, both fine AOD and the column concentration of NO2 were steadily increasing, shown in Figure 2. Anthropogenic pollution usually contributed to the high aerosol optical depth over industrial area (Xia et al., 2005). The fine mode aerosols are mainly attributed to the particles produced by fossil fuel/biomass burning and/or gas-to-particle conversion mechanism (Kedia and Ramachandran, 2011), which was mostly composed by hygroscopic and/or absorbing particles. As stated in the paper and many previous studies (Kaufman and Koren, 2006; Kaufman et al., 2002), those particles play an important role in aerosol-cloud interaction and/or enhance the atmospheric stability, which will further change the precipitation characteristics.

P. 8753, l. 12-13: The first sentence in this paragraph is redundant, as this has been stated in Section 2. I suggest the authors to remove it. Lin et al.: We have removed the redundant sentence.

P. 8754, l. 12: why is 'particually the rain frequency', but not rain amount? References or explanations. Lin et al.: Brief explanations were added to the revised manuscript. ". . .. . .particularly the rain frequency. As shown in Fig. 2, the decreasing trend of TRMM rain frequency was larger than the decreasing trends of rain amount from both TRMM PR and surface gauge measurements."

P. 8754, l. 29: figure 3d-f: what is the unit for the AOD trend? Lin et al.: The unit of AOD trend is "year-1", which was added to the Figure 3e-f.

P. 8755, l. 4: 'MAM' is not defined. Lin et al.: The definition of 'MAM' (March, April and May) was added to the revised manuscript.

P. 8755, l. 11-12: It is fair to state that 'the spatial distributions of rain frequency trends were different from the mean rain frequency distribution' based on Figure 3h and i. It is more fair to say that 'the spatial distributions of rain frequency trends were close to the mean rain frequency distribution'. This will also affect the subsequent statement about large scale dynamical changes. Lin et al.: Rain formed in the atmosphere requires certain dynamical, thermodynamical, and microphysical conditions. Rain spatial distribution indicates region with favorable rain forming conditions. If there were substantial changes of large scale dynamics, the spatial distribution of rain frequency trends will dramatically differ from the mean rain frequency distribution. In our study, as reviewer pointed out that "the spatial distributions of rain frequency trends were close to the mean rain frequency distribution", it suggests that there was no substantial change of large scale dynamics. Our analysis using reanalysis data also demonstrated this. However, some shifts in rain bands associated with slight changes in large scale dynamics are possible. It could result the spatial distributions of rain frequency trends were "similar to" the mean rain frequency distribution. Our study indicated that it was not the case: the spatial distributions of rain frequency trends were different from the mean rain frequency distribution. Most importantly, the spatial distributions of rain frequency trends were more similar to the spatial distribution of pollution and associated aerosol changes.

P. 8756, l. 4: Please clarify about Figure 4. What does each data point in Figure 4 mean? Is the fine mode AOD averaged over individual regions for the spring season? The correlation between fine mode AOD and warm cloud fraction is really small over Yangtze River region, similar to that over Background region. Also, cloud fraction has been shown to have a positive correlation with AOD in satellite data by many previous studies, and potential reasons can be due to cloud contamination in AOD retrievals, the swelling effects of aerosol by clouds, and so on (Quaas et al., 2010), and the positive relationship between AOD and cloud fraction does not necessarily mean the 2nd aerosol indirect effects. Lin et al.: Each data point in Figure 4 represents the mean fine mode AOD of each $1^\circ \times 1^\circ$ grid for the spring season in the region. Following the reviewer's suggestion, the correlation between fine mode AOD and liquid cloud effective radius was further investigated. A more robust anti-correlation between the two was detected over the Yangtze River region and Yellow River region than in the Background region.

P. 8756, l. 18-20: Can the authors elaborate why the coarse mode AOD increases over the India-Myanmar region? Lin et al.: The increase of coarse AOD over the India-Myanmar region may attribute to the large dust storm in northwest India, which starts in March-April in Rajasthan (Gautam et al., 2009; Prospero et al., 2002). Tripathi and Rajmani (1999) suggested that this was a much more intense source of dust in the recent past. On the other hand, the central dry zone of Myanmar is located in a subtropical semi-arid zone, which may also contribute to the increasing dust aerosol in this region.

Since the fine AOD over this region also showed a slight increase, it implies that intense biomass burning in spring in Myanmar may also contribute to the increasing of AOD in this region. However, since the lack of surface observation data and limit research of aerosol in this region, the possible reasons for the increasing AOD still need further investigation.

P. 8757, l. 16: R is 0.48 in Figure 6a. Lin et al.: The value was revised in the new manuscript.

P. 8757, l. 24-25: The statement "the wet scavenging has no dormant effect in the observed trend of AOD at long-term scales" is not supported by their data. Even if the wet scavenging has dormant effect in the observed trend of AOD, I do not see why the correlation between AOD trend and NO2 trend should not be positive. Please remove this statement here and in Section 4 (p. 8759, line 15-17). Lin et al.: We have removed those statements.

P. 8577, l. 25: Figure 6b). Should be X axis 'Fine mode AOD' or 'Fine mode AOD trend'? Lin et al.: The X axis in Figure 6b was "Fine mode AOD".

P. 8757, l. 27: R is 0.82 in Figure 6b. Lin et al.: The value was change to 0.82 in the revised manuscript.

P. 8758, l. 2: Figure 7. Figure 7b is not correct, as the spatial pattern of Figure 7b is not the same as Figure 7a. Also, I noted that Figure 7b and Figure 7 f are identical. Lin et al.: We have replaced Figure 7b with the correct one.

P. 8758, l. 8-9: The area of stronger negative correlation does not really overlap with the regions with strong reduction in rain frequency. Comparing Figure 3b with Figure 7d, it is clear that many areas with stronger reduction in rain frequency shows smaller or even positive correlation between rain frequency and fine mode AOD (e.g., Yangze River regions, and Pearl river regions). Please clarify this statement. Lin et al.: We changed those to: "In general, the temporal variation of fine mode AOD is anti-correlated to that of coarse mode AOD. The consistency of the temporal variation of total AOD with its component, fine mode AOD or coarse mode AOD, depends on which component dominates the total aerosol variation. In the Eastern China, there are more grids that show the consistency of negative correlations of rain frequency with fine mode AOD and with total AOD, suggesting the fine mode AOD dominates in the region. In contrast, in India-Myanmar region, the negative correlation with the coarse mode AOD is similar to that with the total AOD, as the coarse mode AOD dominates the component of aerosol in the region. "

P. 8758, l. 15-26: This paragraph does not provide enough evidence for its claim that 'the observed changes in precipitation were not related to the dynamical changes in the atmosphere'. The authors only examined two parameters, PW and DWVT, which do not cover the full sets of dynamical parameters. Lin et al.: We have changed the statement to "the observed changes in precipitation were not related to PW and DWVT, the major dynamical factors that may affect large scale precipitation change."

P. 8759-8760, conclusion and discussion: Need more discussion about the uncertainty associated satellite data, and the authors also need to more prominently acknowledge the correlation study nature of this paper, and to caution the readers for the causality in both the abstract and the conclusion. Lin et al.: Thanks for all your comments. We added more discussion about measurement uncertainty, and correlation and causality issues

Technical corrections: P. 8749, l. 16: 'of and' ! 'of'. Lin et al.: It was corrected in the revised manuscript.

P. 8753, l. 25: 'that' ! 'than'. Lin et al.: It was corrected in the revised manuscript.

References:

Quaas, J., Stevens, B., Stier, P., and Lohmann, U.: Interpreting the cloud cover – aerosol optical depth relationship found in satellite data using a general circulation model, Atmos. Chem. Phys., 10, 6129-6135, doi:10.5194/acp-10-6129-2010, 2010. Zhang, Q., Streets, D. G., Carmichael, G. R., He, K. B., Huo, H., Kannari, A., Klimont, Z., Park, I. S., Reddy, S., Fu, J. S., Chen, D., Duan, L., Lei, Y., Wang, L. T., and Yao, Z. L.: Asian emissions in 2006 for the NASA INTEX-B mission, Atmos. Chem. Phys., 9, 5131-5153, doi:10.5194/acp-9-5131-2009, 2009.

DeMoss, J. D., and Bowman, K. P.: Changes in TRMM rainfall due to the orbit boost estimated from buoy rain gauge data, Journal of Atmospheric and Oceanic Technology, 24, 9, 1598-1607, 2007.

Gautam, R., Hsu, N. C., Lau, K. M., and Kafatos, M.: Aerosol and rainfall variability over the Indian monsoon region: distributions, trends and coupling, Annales Geophysicae, 27, 9, 3691-3703, 2009.

Huang, Y., Chameides, W. L., and Dickinson, R. E.: Direct and indirect effects of anthropogenic aerosols on regional precipitation over east Asia, Journal of Geophysical Research-Atmospheres, 112, D3, doi:10.1029/2006JD007114 , 2007.

Hyer, E. J., Reid, J. S., and Zhang, J.: An over-land aerosol optical depth data set for data assimilation by filtering, correction, and aggregation of MODIS Collection 5 optical

depth retrievals, Atmospheric Measurement Techniques, 4, 3, 379-408, 2011.

Ichoku, C., Chu, D. A., Mattoo, S., Kaufman, Y. J., Remer, L. A., Tanre, D., Slutsker, I., and Holben, B. N.: A spatio-temporal approach for global validation and analysis of MODIS aerosol products, Geophysical Research Letters, 29, 12, -, 2002.

Iida, Y., Kubota, T., Iguchi, T., and Oki, R.: Evaluating sampling error in TRMM/PR rainfall products by the bootstrap method: Estimation of the sampling error and its application to a trend analysis, Journal of Geophysical Research-Atmospheres, 115, doi:10.1029/2010JD014257, 2010.

Lin, J. J.: Characterization of the major chemical species in PM2.5 in the Kaohsiung City, Taiwan, Atmospheric Environment, 36, 12, 1911-1920, 2002.

Kahn, R. A., Nelson, D. L., Garay, M. J., Levy, R. C., Bull, M. A., Diner, D. J., Martonchik, J. V., Paradise, S. R., Hansen, E. G., and Remer, L. A.: MISR Aerosol Product Attributes and Statistical Comparisons With MODIS, Geoscience and Remote Sensing, IEEE Transactions on, 47, 12, 4095-4114, 2009.

Kaufman, Y. J., and Koren, I.: Smoke and pollution aerosol effect on cloud cover, Science, 313, 5787, 655-658, 2006.

Kaufman, Y. J., Tanre, D., and Boucher, O.: A satellite view of aerosols in the climate system, Nature, 419, 6903, 215-223, 2002.

Kedia, S., and Ramachandran, S.: Seasonal variations in aerosol characteristics over an urban location and a remote site in western India, Atmospheric Environment, 45, 12, 2120-2128, 2011.

Lei, Y., Zhang, Q., He, K. B., and Streets, D. G.: Primary anthropogenic aerosol emission trends for China, 1990-2005, Ieee Power and Energy Soceity General Meeting 2010, 931-954, 6545, 2010.

Levy, R. C., Remer, L. A., Kleidman, R. G., Mattoo, S., Ichoku, C., Kahn, R., and Eck,

T. F.: Global evaluation of the Collection 5 MODIS dark-target aerosol products over land, Atmospheric Chemistry and Physics, 10, 21, 10399-10420, 2010.

Lin, J. J.: Characterization of the major chemical species in PM2.5 in the Kaohsiung City, Taiwan, Atmospheric Environment, 36, 12, 1911-1920, 2002.

Miracolo, M. A., Hennigan, C. J., Ranjan, M., Nguyen, N. T., Gordon, T. D., Lipsky, E. M., Presto, A. A., Donahue, N. M., and Robinson, A. L.: Secondary aerosol formation from photochemical aging of aircraft exhaust in a smog chamber, Atmos. Chem. Phys. Discuss., 10, 11, 27893-27924, 2010.

Nakazawa, T., and Rajendran, K.: Interannual Variability of Tropical Rainfall Characteristics and the Impact of the Altitude Boost from TRMM PR 3A25 Data, Journal of the Meteorological Society of Japan, 87, 317-338, 2009.

Prospero, J. M., Ginoux, P., Torres, O., Nicholson, S. E., and Gill, T. E.: Environmental characterization of global sources of atmospheric soil dust identified with the Nimbus 7 Total Ozone Mapping Spectrometer (TOMS) absorbing aerosol product, Reviews of Geophysics, 40(1), 1002, doi:10.1029/2000RG000095,, 2002.

Qian, Y., Gong, D. Y., and Leung, R.: Light rain events change over North America, Europe, and Asia for 1973-2009, Atmospheric Science Letters, 11, 4, 301-306, 2010.

Remer, L. A., Kaufman, Y. J., and Kleidman, R. G.: Comparison of three years of Terra and Aqua MODIS aerosol optical thickness over the global oceans, Ieee Geoscience and Remote Sensing Letters, 3, 4, 537-540, 2006.

Remer, L. A., Kaufman, Y. J., Tanre, D., Mattoo, S., Chu, D. A., Martins, J. V., Li, R. R., Ichoku, C., Levy, R. C., Kleidman, R. G., Eck, T. F., Vermote, E., and Holben, B. N.: The MODIS Aerosol Algorithm, Products, and Validation, Journal of the Atmospheric Sciences, 62, 4, 947-973, 2005.

Remer, L. A., Kleidman, R. G., Levy, R. C., Kaufman, Y. J., Tanre, D., Mattoo, S., Martins, J. V., Ichoku, C., Koren, I., Yu, H. B., and Holben, B. N.: Global aerosol climatol-

ogy from the MODIS satellite sensors, Journal of Geophysical Research-Atmospheres, 113, D14S07, doi:10.1029/2007JD009661, 2008.

Song, Y., Xie, S. D., Zhang, Y. H., Zeng, L. M., Salmon, L. G., and Zheng, M.: Source apportionment of PM2.5 in Beijing using principal component analysis/absolute principal component scores and UNMIX, Science of the Total Environment, 372, 1, 278-286, 2006.

Tripathi, J. K., and Rajamani, V.: Geochemistry of the loessic sediments on Delhi ridge, eastern Thar desert, Rajasthan: implications for exogenic processes, Chemical Geology, 155, 3-4, 265-278, 1999.

Veefkind, J. P., Boersma, K. F., Wang, J., Kurosu, T. P., Krotkov, N., Chance, K., and Levelt, P. F.: Global satellite analysis of the relation between aerosols and short-lived trace gases, Atmos. Chem. Phys., 11, 3, 1255-1267, 2011.

Xia, X. A., Chen, H. B., Wang, P. C., Zong, X. M., Qiu, J. H., and Gouloub, P.: Aerosol properties and their spatial and temporal variations over North China in spring 2001, Tellus Series B-Chemical and Physical Meteorology, 57, 1, 28-39, 2005.

Zhang, J., and Reid, J. S.: A decadal regional and global trend analysis of the aerosol optical depth using a data-assimilation grade over-water MODIS and Level 2 MISR aerosol products, Atmospheric Chemistry and Physics, 10, 22, 10949-10963, doi:10.5194/acp-10-10949-2010, 2010.

Zhang, J. L., and Reid, J. S.: An analysis of clear sky and contextual biases using an operational over ocean MODIS aerosol product, Geophys. Res. Lett., 36, L15824, doi:10.1029/2009GL038723, 2009.

Zhang, Q., Streets, D. G., Carmichael, G. R., He, K. B., Huo, H., Kannari, A., Klimont, Z., Park, I. S., Reddy, S., Fu, J. S., Chen, D., Duan, L., Lei, Y., Wang, L. T., and Yao, Z. L.: Asian emissions in 2006 for the NASA INTEX-B mission, Atmospheric Chemistry and Physics, 9, 14, 5131-5153, 2009.

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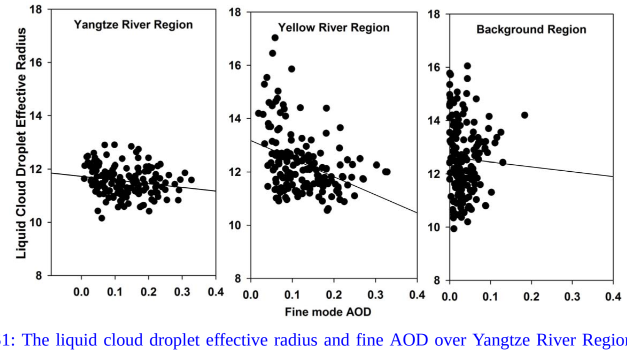

Figure S1: The liquid cloud droplet effective radius and fine AOD over Yangtze River Region(30-34ºN, 118-122ºE, grid over ocean is excluded), Yellow River Region(32-36ºN, 111-115ºE), and background region (32-36ºN,100-104ºE) with the pristine atmosphere

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Fig. 1.

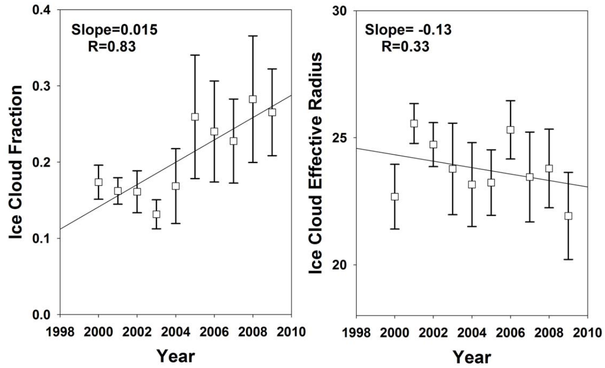

Figure S2: The trend of ice cloud fraction and ice cloud effective radius over India-Myanmar region.

1

Fig. 2.

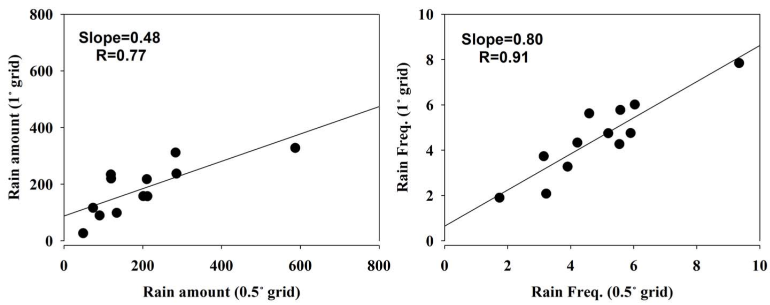

Figure S3: The comparison of the averaged rain amount and rain frequency of 1º×1º and 0.5º×0.5º grids

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Fig. 3.