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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impact of the East Asian summer monsoon on long-term variations in the acidity of summer precipitation in Central China" *by* B. Z. Ge et al.

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We are grateful to Dr. Aas for the valuable comments on our manuscript. Following are our responses.

Comment 1. It is interesting that you may find a "teleconnection" between pH and summer monsoon. Though maybe it is possible to use a simpler correlation, i.e. is it the monsoon itself or actually just a relation between pH and precipitation volume? High precipitation since more diluted acidification. This doesn't need to be shown with eigen-vectors etc, though changing precipitation volume may be because changing

weather pattern. It is always a danger to use very complicated statistics which the reader may have difficult to follow. At least some of the data used as input for statistical analysis need to be better presenter.

Reply to comment 1: There are many factors that affect the pH of precipitation, such as pollutants emission, transport and deposition, cloud chemistry process, and even the terrain can also influence the pH to some extent. Rainfall may affect the pH of course, since of dilution of acid pollutants and change of deposition. However, it is not a simple relationship that more rainfall corresponding to the high pH. It is very hard to investigate and/or obtain the simple relationship just between the pH and the local rainfall, because rainfall in different region and different season has different impacts to pH (Xie et al. 2009, Lin et al. 2009). East Asia summer monsoon is a comprehensive factor which can not only influence the wind field over eastern China and then change the transport of pollutants, but also impact the rainfall pattern over eastern and northern China. We take use of statistical analysis to explore the relationship between East Asia summer monsoon and the pH in Central China in order to interpret the changes of pH over Central China from physical mechanism. Rainfall is an important factor that influenced by summer monsoon. And we find out there is a teleconncetion between rainfall in MYRL and pH in Central China. This can testify our hypothesis that East Asia summer monsoon can affect the pH in Central China. In order to facility readers, we will add the representation of our input data in statistical method introduction section.

Comment 2. The representativety of the data from CMA-ARMN network need to be discussed more carefully, because this has an effect on both the spatial and temporal analysis: I.e.: 1)ãĂĄHow are these measured: bulk or wet only samples; daily, weekly or monthly samples; which components are measured, only pH?? Why not the ion content? What is the site characteristic? Is it a mixture of urban and rural site? Only rural wet only samples should be used. 2)ãĂĄ74 sites are used. Have all these sites data for whole period 1992-2006, or is some of the site for shorter time periods? If not the sites with less data should be excluded.

Reply to comment 2: The polyethylene polymer bucket of 40 cm diameter was used for wet-only sample collection at sites, and event samples were collected before 2005, daily precipitation samples were collected during 2006. Precipitation pH and conductivity are measured at site within 4 hours after the sample collected. The CMA-ARMN network stations share the facilities of the meteorological observation stations, which are normally located in suburb of a city or a town, thus much less directly influenced by urban emissions. In details, 37 among the 74 sites are located in relatively sparsely populated areas, including 3 GAW (Global Atmosphere Watch) regional stations and 3 weather stations on top of mountains, the rest 37 are city or near-city sites. Although the sites are of different characteristics, they are mixed and distributed in different regions. In this paper, we focus on variations of precipitation acidity and meteorological parameters in larger regions and the relationship between these quantities. The quality of data from all the 74 sites was checked by series procedures (Tang et al., 2010) and is believed meeting our purpose. Among the 74 sites data, only 5 sites missed measurements of 1 year, and a station missed 2 years. This small gap in the data series may not significantly influence the trends analysis (Tang et al., 2010).

Comment 3. It is not clear how the averaging is not (done?) for each region. Is it volume weighted mean for all the sites lumped together in each region? If so, it would be interesting to know about the spread/variability and the representativity as mentioned above.

Reply to comment 3: The averaging of pH for each region is volume weighted mean for all sites in the region. Most of stations are distributed equidistantly in the focused Central and East China regions. The distribution of stations is shown in Figure 1. We will add "Most of stations are distributed nearly equidistantly in Central and East China, where acid rain occurs at high frequencies. Although stations in Northwest China are sporadic and unevenly distributed, our results may not be significantly distorted since acid rain occurs only very scarcely in that region. "in our revised manuscript in line 10 page 8, section 3.1.1. The results for pHVWA, together with the arithmetic mean pH,

rainfall, and the number of stations, are listed in Table 2 in our revised manuscript. As can be seen in the table, the lowest pH value is in SW and YRM, while the highest is in NW and NE for both pH averaging methods. In this study, we focus on YRM and CT, and call them Central China.

Comment 4. How is the map of the measured pH been done? Is it volume weighted averages for each site for the whole time period that has undergone statistical kriging?

Reply to comment 4: Yes, in average pH patterns during 1992$\sim$2006, 1992$\sim$1999 and 2000$\sim$2006, the volume weighted average for each site is done. After this, we converted the pH into H+ ion concentration and interpolated into 0.5$\times$1 degree data using Cressman interpolation method. We will add this to our revised version.

Comment 5. When you discuss significantly change between the two periods (end of point 3.1.1), how is that done. Is it and average for each period which is compared? You have used the Mann Kendall test for trend analysis? And if so you need more points, at least 4 to define confidence interval?

Reply to comment 5: We just compared the average value of pH in this two periods (e.g., 1992$\sim$1999, 2000$\sim$2006), and used T-test to check up if this difference is significant. The Mann- Kendall test we mentioned in our paper was done by Tang, et al.(2010) based on monthly volume-weighted pH values. We will add this information in our revised version. Comment 6. The CMAQ model may be ok. It seems like it is quite well documented for ozone and NO2, but for sulphate deposition the references are not well documented for external reader, i.e. Ohara (2010) is difficult to access. Further, the correlation between model results and what is measured in the urban EANET sites Hongwen and Guanyinqiao is not very useful. The site are not representative for large scale model with a grid of 50$\times$50 km or 100$\times$100 km (should be specified). As seen in figure 1, nitrate gives very poor correlation, and a log scale y axes hide some of the large biases. These sites may reflect the general emission changed in China, but they are not very good for spatial resolution. It would have been much more useful if the model could have been compared to rural sites, like the EANET site Jinyuanshan and Xiaoping. Further on, more than two sites are needed to verify the model for whole of China.

Reply to comment 6: Firstly, our model horizontal resolution is 80×80 km. Secondly, we agree that the log scale y axes hide some of biases. So, we re-do our validation of CMAQ model using observation data adding two rural sites (Jinyunshan and Xiaoping) and two GAW sites (Linan and Longfenshan) (See Figure 1 and Table 1 in our revised manuscript). As it shows, variations of nitrate concentrations are well reproduced by CMAQ model both at Guanyinqiao and at Jinyunshan (It looks better than log scale y axes although there exists some underestimation), with the latter being better. Sulfate seems to be underestimated in CMAQ model in some high concentration events at all sites. However, the underestimations in high concentration events are all in winter and not in our focus season summer. Moreover, both simulated and observed data have similar trends. Therefore, the modeled results can be used in our analysis.

Comment 7. As I see it the NO3 and SO4 deposition is mainly a model exercise, which is ok, but should be addressed more like that. I have some problems to see the relative importance of sulfate and nitrate to pH. In central China sulfate is much more dominant that nitrate to the pH, but in this paper these are treated as equal importance. Some discussion of which trend (SO2 and NOx emission) has had largest impact on the changes in pH could be interesting to include.

Reply to comment 7: Indeed, different pollutants have different impacts on the pH in different areas. However, in this paper we do not investigate the influence of sulfate and nitrate on the pH separately. To address the separate influences of different pollutants, much more in-depth studies should be done, and this would be our next objection.

We will add the following reference in our manuscript in page 19595, line 19, to support that air pollution was reduced after some measures implemented, but rebound after 2000. And also, we will add the reference in page 19595, line 24, to show that the

emission and transport of SO2 and NOx are the dominant causes of acid rain in China. Larssen, T., Lydersen, E., Tang, D., He, Y., Gao, J., Liu, H., Duan, L., Seip, H.M., Vogt, R.D., Mulder, J., Shao, M., Wang, Y., Shang, H., Zhang, X., Solberg, S., Aas, W., Økland, T., Eilertsen, O., Angell, V.M., Liu, Q., Zhao, D., Xiang, R., Xiao, J., Luo, J., 2006. Acid rain in China. Environmental Science and Technology, 15, 418–425.

References: Lin, C. C., J. X. Liu, Y. Y. Cai, B. L. Li, Z. L. Wang & B. B. Chen (2009) Study on the Relationship Between Meteorological Conditions and Acid Rain in Mid-Eastern Fujian. Bulletin of Environmental Contamination and Toxicology, 83, 180-187. Xie, Z. Q., Y. Du, Y. Zeng, Y. C. Li, M. L. Yan & S. M. Jiao (2009) Effects of precipitation variation on severe acid rain in southern China. Journal of Geographical Sciences, 19, 489-501. Larssen, T., Lydersen, E., Tang, D., He, Y., Gao, J., Liu, H., Duan, L., Seip, H.M., Vogt, R.D., Mulder, J., Shao, M., Wang, Y., Shang, H., Zhang, X., Solberg, S., Aas, W., Økland, T., Eilertsen, O., Angell, V.M., Liu, Q., Zhao, D., Xiang, R., Xiao, J., Luo, J., 2006. Acid rain in China. Environmental Science and Technology, 15, 418–425.

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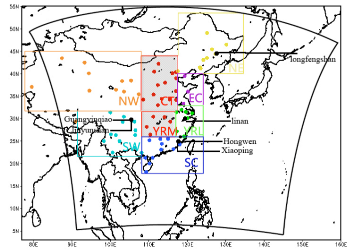

Figure 1. The modeling domain and measurement sites of CMA-ARMN.

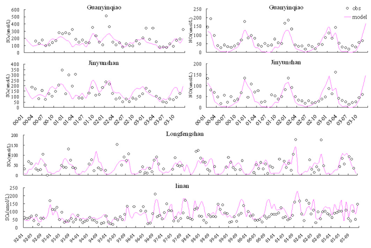

Figure 2. Time series of modeled and observed monthly volume-weighted mean concentrations of sulfate and nitrate for EANET sites and GAW background stations

Fig. 1.