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Acidity in Precipitation and Solar North-South Asymmetry

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We are motivated by both the accumulating evidence for the connection of solar variability to the chemistry of nitrogen oxide in the atmosphere and recent finding that the Galactic cosmic-ray (GCR) influx is associated with the solar north-south asymmetry. We have analyzed the measured pH in precipitation over the 109 stations distributed in the United States. We have found that data of pH in precipitation as a whole appear to be marginally anti-correlated with the solar asymmetry. That is, rain seems to become less acidic when the southern hemisphere of the Sun is more active. The acidity of rain is also found to be correlated with the atmospheric temperature, while not to be correlated with solar activity itself. We have carried on the analysis with two subsamples in which stations located in the east and in the west. We find that the pH data derived from the eastern stations which are possibly polluted by sulfur oxides and nitrogen oxides are not correlated with the solar asymmetry, but with the temperature. On the contrary, the pH data obtained from the western stations are found to be marginally anti-correlated with the solar asymmetry. In addition, the pH data obtained from the western stations are found to be correlated with the solar UV radiation. We conclude by briefly pointing out that a role of the solar asymmetry in the process of acidification of rain is to be further examined particularly when the level of pollution by sulfur oxides and nitrogen oxides is low.

Keywords: Acid precipitation, solar north-south asymmetry, data analysis

1. INTRODUCTION

Though acidic rain was discovered in 1853, it was not until the late 1960s that scientists began widely observing to study the phenomenon (Likens et al. 1979). Acid rain is a rain or any other form of precipitation that causes water and soil to become more acidic. It has harmful effects on plants, aquatic animals, and infrastructure. Acidity is measured on the pH scale ranges from 0 to 14 with 0 being acid, 7 as neutral, and 14 as alkaline. Distilled water, once carbon dioxide is removed, has a neutral pH of 7. On the other hand, because carbon dioxide and water in the air react together to form carbonic acid, a weak acid, even unpolluted rain has an acidic pH, but usually no lower than 5.6. Occasional pH readings in rain and fog water of well below 2.4 have been reported in industrialized areas.

Acid rain is caused by emissions of sulfur dioxide and nitrogen oxide by industrial and transportation sources, which react with the water molecules in the atmosphere to produce nitric acid (HNO_3) and sulfuric acid (H_2SO_4) through the processes of oxidation and hydrolysis. Besides, volcanoes and forest fires may naturally emit sulphur dioxide (SO_2) and every time there is a flash of lightening nitrogen oxides can also be roduced (Likens & Butrler et al. 1981, Wolff 1995). The actual pathways can be influenced by the prevailing atmospheric conditions, such as, sunlight, temperature, humidity, and the presence of hydrocarbons, nitrogen dioxides, particulates in the atmosphere (Wisniewski & Kinsman 1982, Parungo et al. 1987, Cotter et al. 2003, Frey et al. 2005). Acidity in precipitation is actually a complex function of atmospheric aerosols and gases and of physical and chemical processes of hydrological cycles.

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Such attributes of its process initiated numerous investigations in various aspects. For instance, attempts have been made over the years to link solar variability to chemistry of nitrogen oxide which are oxidized to form nitric acid (Logan 1983, Aikin 1994, Garcia 1994), as the effect of nitrogen oxide (NO_x) on the local air quality and acid rain is as important as that of SO_2. Even though nitrogen oxide (NO_x) has been conventionally considered to be less crucial than sulfur dioxide (SO_2) in a role, we note that emissions of nitrogen oxides are of increasing importance due to strict controls on emissions of sulfur containing compounds these days. By using the ice core drilled at Talos Dome (Antarctica) the 11-year and Gleissberg solar cycles are shown to be present with the variability of 10 – 25 % in nitrate content in the pre-industrial epoch (Zeller & Parker 1981, Zeller & Dreschhoff 1995, Patris et al. 2002, Traversi et al. 2012). In fact, Galactic cosmic-ray (GCR) particles whose influx is modulated by the solar magnetic activity change not only the characteristics of the atmosphere but also the rate of various physical-chemical processes, which makes them one of the most plausible agents linking the solar magnetic activity to various phenomena (Friis-Christensen & Lassen 1991, Marsh & Svensmark 2000, Pudovkin 2004, Haigh 2007, Bazilevskaya et al. 2008). Ney (1959) first suggested that GCRs could affect the electrical environment in the troposphere, and give rise to an 11-year cycle in thunderstorm activity. Influx of cosmic energetic particles into the atmosphere greatly also disturbs the chemical composition from the upper stratosphere to the lower thermosphere (Crutzen 1975, Garcia et al. 1984, Reid et al. 1991, Vitt & Jackman 1996, Vitt et al. 2000, Ogurtsov et al. 2004, Jackman et al. 2005, Sinnhuber et al. 2012). Most important are changes to the budget of atmospheric nitric oxides and to atmospheric reactive hydrogen oxides. Large solar proton events can have a quite sporadic yet very significant impact on the stratospheric nitrogen oxide (NO_x) budget (Storini & Damiani 2007, Damiani et al. 2010, Funke et al. 2011). The study of responses of atmospheric ionization and the global electric circuit to varying GCRs demonstrates that the atmospheric transparency is associated with GCR flux (Wilcox et al. 1973, Tinsley 2000, Boberg & Lundstedt 2002, Roldugin & Tinsley 2004, Troshichev 2008). It has been also shown that GCRs are related to temperature, pressure, and tropospheric dynamics in general (Pudovkin et al. 1997, Cho et al. 2012).

In this contribution correlations between the pH in precipitation measured across the United States and the observed solar north-south asymmetry of sunspot area have been attempted. In addition to the solar north-south asymmetry we have also tried the total sunspot area, the solar UV variability, the atmospheric temperature in calculating correlations for comparison. We are motivated by both the accumulating evidence for the connection of solar variability to the chemistry of nitrogen oxide (NO_x) in the atmosphere as mentioned above and recent finding that the GCR influx is associated with the solar north-south asymmetry (Cho et al. 2011). It is natural on the basis of what we discussed above that one may attempt to relate the solar north-south asymmetry to the chemistry of nitrogen oxide (NO_x). It is indeed an interesting question of whether or not the observed pH in precipitation is associated with the solar north-south asymmetry since it is never attempted to our best knowledge. When looking for a relation between anthropogenic nitric acid (HNO_3) and sulfuric acid (H_2SO_4) emissions and the acid content of precipitation one is to draw a conclusion with due care considering other possible components including unknown ingredients yet. Trend analysis such as the one presented here is a useful tool in many aspects of the scientific consideration of acid rain and its effects. To the extent that the available data permit, several hypotheses on why trends are found may be evaluated. It is beyond the scope of this study, however, to provide a new model or a theoretical translation. We are to focus here our current presentation on reporting what we have found.

This paper begins with descriptions of data and calculated correlations between the measured pH data and the solar north-south asymmetry in Section 2. We discuss the results by comparing with other outcomes in Section 3. Finally, we summarize and conclude in Section 4.

2. CORRELATIONS BETWEEN PH AND SOLAR NORTH-SOUTH ASYMMETRY

In Fig. 1, we show the spatially averaged value (thin line) of the measured pH over the stations distributed in the United States for the time interval from September in 1986 to January in 2008 and its running average (thick line) with the 7 year-long moving window as a function of time. We have adopted pH data from the National Atmospheric Deposition Program (NADP) website[1]. The NADP established in May 1978, at the Illinois State Water Survey, University of Illinois, maintains three networks with more than 350 deposition monitoring sites. The NADP National Trends Network (NTN) has 250 sites in the United States located away from point sources of pollution. Each

[1]http://nadp.sws.uiuc.edu/NTN

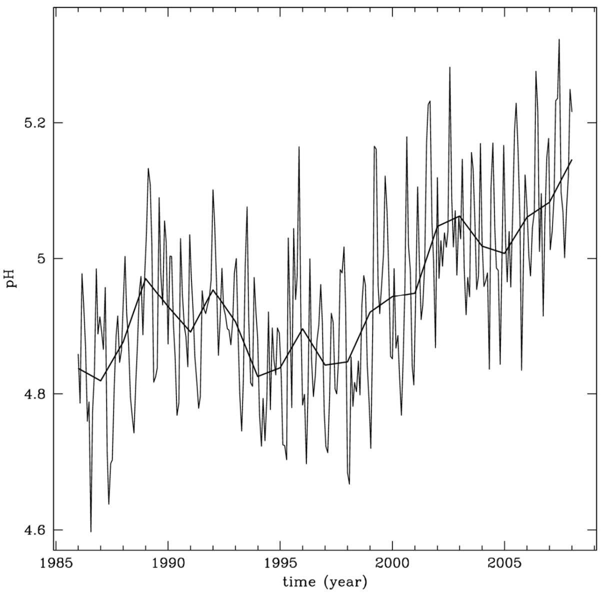

Fig. 1. Spatial average value (thin line) of measured pH over the United States for the time interval from September in 1986 to January in 2008 and its running average (thick line). The length of the moving window is 7 years.

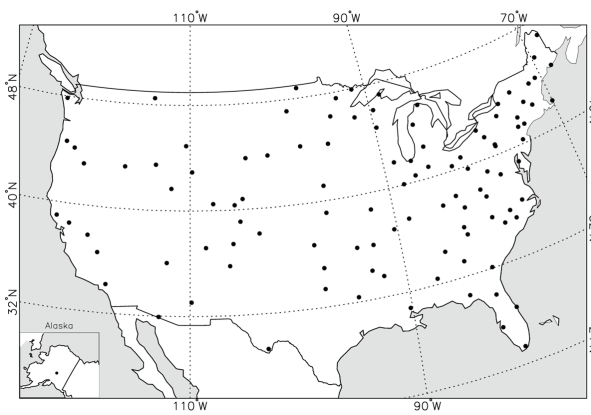

Fig. 2. Location of 109 stations of NTN in the United States where the monthly pH data are taken for the present analysis.

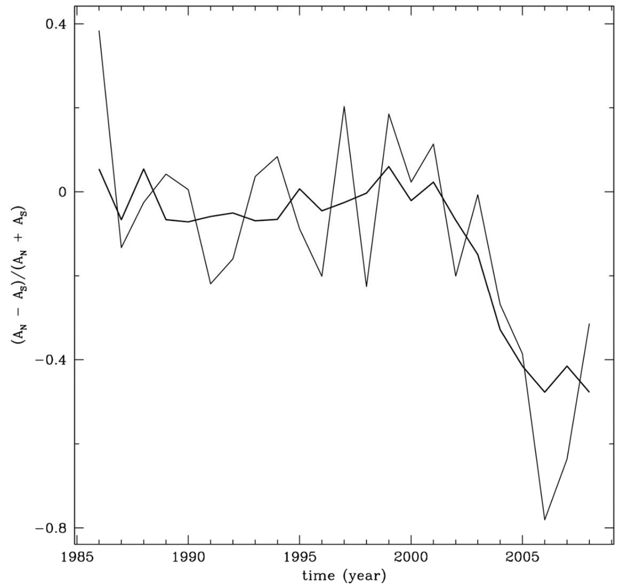

Fig. 3. Observed monthly solar north-south asymmetry (thin line) and its running average (thick line). The solar north-south asymmetry is defined as the difference of the sunspot area appearing in the solar northern and southern hemispheres normalized by their.

site has an automated precipitation collector and gage to gather samples only during rain or snowfall. For the present analysis the monthly pH data are taken from the 109 stations of NTN, whose locations are shown by filled circles in Fig. 2, in order to guarantee the spatial homogeneity.

According to the view that the normal pH of rain is a slightly acidic 5.6, the problem of acid rain seems widespread in the United States. Apparent decrease in the acidity of rainfall since the late 1990s may be due to the Acid Rain Program established by a series of amendments to the Clean Air Act Title IV calling for a total reduction of about 10 million tons of SO_2 emissions from power plants. It was implemented in two phases. Phases I and II began in 1995 and 2000, respectively. It is not straightforward, however, to find a clear relation when close examining the relation between anthropogenic sulphur emissions and the acidity of precipitation to pin-point instants of the decrease in the acidity of rainfall. For instance, NADP monitoring data in the eastern United States show that wet sulfate deposition has decreased an average of 30 % and nitrogen deposition has decreased as well, but to a lesser extent, since the early 1990s (see, e.g., NAPAP, 2005, 2011). It should be noted that this point of time is earlier than when the United States Congress passed these amendments and when the apparent decrease in the acidity of rainfall began.

In Fig. 3, we show the observed monthly solar north-south asymmetry (thin line) and its running average (thick line) as a function of time. The solar north-south asymmetry is defined as the difference of the sunspot area appearing in the solar northern and southern hemispheres normalized by their sum, $(A_N-A_S)/(A_N+A_S)$, where A_N and A_S are the sunspot area appearing in the solar northern and southern hemispheres, respectively (Chang, 2008). Note that the solar northern and southern hemispheres are magnetically more active in general when sunspots appear more in the solar northern and southern hemispheres, respectively. That is, when values of the solar north-south asymmetry are positive the northern solar hemisphere is more active, and vice versa. In this particular example the length of the

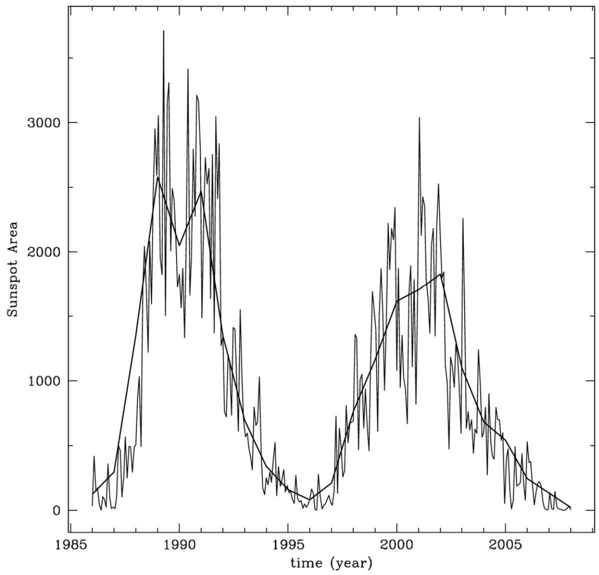

Fig. 4. Monthly and yearly average of the sunspot area in units of millionths of a hemisphere Thin and think lines represent monthly and yearly average, respectively.

moving window is 7 years. For the calculation of the solar north-south asymmetry we have taken sunspot area data for the time interval from September in 1986 to January in 2008 covering the solar cycles 22 and 23 from the National Aeronautics and Space Administration (NASA) website[2], in which monthly averages of the daily sunspot areas in units of millionths of a hemisphere are available for the solar northern and southern hemispheres separately.

We calculate a linear correlation coefficient *r* between the measured pH in precipitation and the observed solar north-south asymmetry, and the single-sided chance probability that *r* has an equal or larger value than its observed in the null hypothesis. There is a marginal anti-correlation found. That is, rain apparently seems to become less acidic when the southern hemisphere of the Sun is more active. The obtained correlation coefficient and chance probability are r=-0.59 with P=0.04. To examine additional statistical tests of data sets, we also employ the Spearman rank-order correlation test. It returns a correlation value r_s and the single-sided probability $P(r_s, N)$ that N pairs of uncorrelated variables would yield a value of r_s equally or more discrepant than the one obtained from the data set. We find again that the pH data appear to be anti-correlated marginally with the solar north-south asymmetry, that is, r_s=-0.45 with $P(r_s, N)$=0.11.

In Fig. 4, for comparison, we show the monthly and

[2]http://solarscience.msfc.nasa.gov/greenwch.shtml

Table 1. A linear correlation coefficient r and chance probability P, a Spearman rank-order correlation value r_s and its probability $P(r_s,N)$ for the relations between the measured pH over the stations distributed across the United States and parameters indicated in the first row.

	Asymmetry	Solar Cycle	UV radiation	Temperature Anomaly
r	-0.59	0.04	0.21	0.56
P	0.04	0.42	0.16	0.01
r_s	-0.45	0.10	0.38	0.55
$P(r_s,N)$	0.11	0.32	0.03	0.01

yearly average of the sunspot area in units of millionths of a hemisphere which appeared in the solar northern and southern hemispheres, A_N+A_S, as a function of time for the same period. Thin and thick lines represent the monthly average of the sunspot area and the yearly average of the sunspot area, respectively. The Schwabe cycles 22 and 23 are clearly seen. We repeat calculations of correlation coefficient and chance probability. In the case of the sunspot area as an index of solar activity, both the linear correlation coefficient and the Spearman rank-order correlation value consistently imply that there is no correlation between the pH in precipitation and the observed sunspot area. That is, the acidity of rain measured across the United States as a whole is not directly correlated with solar activity. The obtained statistical parameters are listed in Table 1.

3. DISCUSSION

Solar radiation in UV range and the atmospheric temperature are widely recognized as an important factor in the photodissociation process of the chemistry of acid rain. One of reasons for this is that the bulk of the acidity in rain comes from the reaction of sulfur dioxide (SO_2) with hydrogen peroxide in clouds and that the very hydrogen peroxide is formed from the photochemical reactions of volatile organic compounds. This mechanism is particularly important in summer, when most acid rain falls. In order to test this hypothesis we have carried out the same analysis with effective proxies of solar UV variation and the atmospheric temperature which are shown in Fig. 5 and 6.

In Fig. 5, we show the Mg II core-to-wing ratio for the same period as a function of time. The Mg II core-to-wing ratio is one of the most widely used indices of solar activity in the ultraviolet region (Heath & Schlesinger 1986). The solar chromospheric activity in the UV region is of great importance to our understanding of both the physical properties of the Sun as a star, and of the solar influence on the terrestrial stratospheric chemistry. The Mg II core-to-wing index was first developed for the Nimbus 7 solar backscatter ultraviolet spectrometer as an indicator of solar UV flux temporal variation. The Mg II core-to-wing

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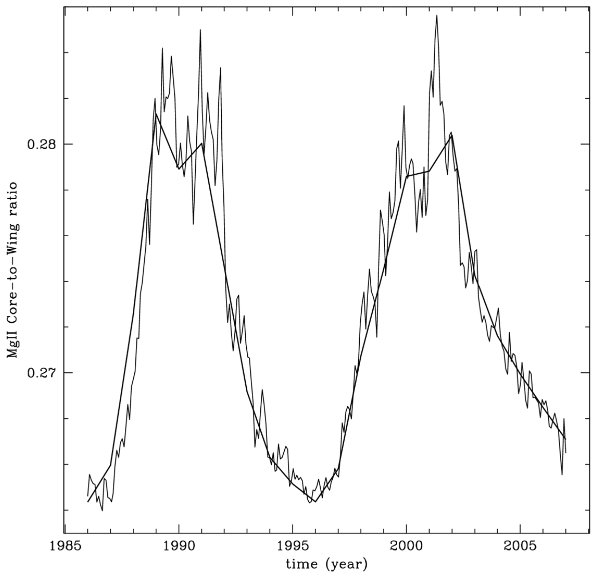

Fig. 5. Monthly and yearly Mg II core-to-wing ratio as an indicator of Solar UV flux variation. Thin and thick lines represent monthly and yearly average, respectively.

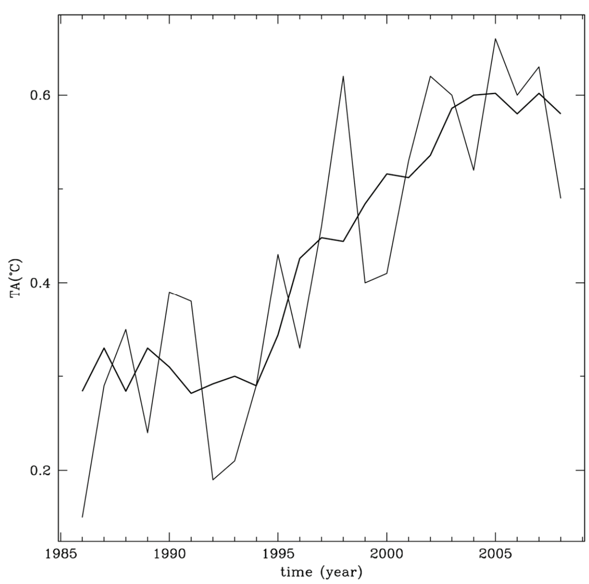

Fig. 6. Global temperature anomaly (TA) and its running average. Thin and thick lines represent monthly TA and running averaged TA, respectively. The length of the moving window is 7 years.

ratio is derived from the ratio of the H and K lines of the solar Mg II feature at 280 nm to the background or wings at approximately 278 nm and 282 nm. We have adopted the Mg II daily index version 9.1 from the National Geophysical Data Center (NGDC) database[3]. Thin and thick lines represent the monthly average of the ratio of the core to wing and the yearly average of the ratio, respectively. Note that the Schwabe cycles 22 and 23 can be also seen in phase. Having repeated calculations of correlation coefficients and chance probabilities we obtain both the linear correlation coefficient and the Spearman rank-order correlation value indicating that the acidity of rain is not correlated with the solar UV variation. This result agrees with that found with sunspot area data. It seems obvious in the sense that both of these are basically same kinds of proxies of solar activity. The obtained statistical parameters are listed in Table 1.

In Fig. 6, we show the global temperature anomaly (thin line) and its running average (thick line) for the same period as a function of time. In this plot the length of the moving window is 7 years. A data set for the global temperature anomaly has been derived from the Hadley Center of the United Kingdom Met Office[4]. They provide the monthly averaged temperature anomaly of 5°×5° grids which are globally spread, and defined by a temperature deviation from the mean value over the period of 1951-1980. We repeatedly calculate the linear correlation coefficient r and the Spearman rank-order correlation coefficient r_s with the chance probabilities between the pH in precipitation and the temperature anomaly. The pH data appear to be marginally correlated with the temperature. That is, the linear correlation coefficient and chance probability are r=0.56 with P=0.01. The Spearman rank-order correlation value is again found to be r_s=0.55 with $P(r_s, N)$=0.01. The obtained statistical parameters are listed in Table 1.

In Fig. 7, we show similar plots to Fig. 1, except that the upper panel and the lower panel result from stations of the United States in the east and in the west with respect to the longitude of 85°W. In the western United States, aside from the heavily polluted area such as Los Angeles basin, the alkaline dust in the air neutralizes the acid. More crucially, in the western United States the amount of emissions is much smaller than in the eastern part of the United States. One may expect that the trend of temporal pH distribution in precipitation could exhibit different behaviors in two separate regions whose average pH level is different. To examine this idea we divide the group of stations into two, that is, stations located in the east and in the west with respect to the longitude of 85°W, so that the pH data set is separated into two subsets in terms of acidity. This division may be somehow arbitrary. But according to the spatial distribution of data of pH in precipitation over the United States provided by a report of the NAPAP the longitude

[3]http://www.ngdc.noaa.gov/stp/solar/solaruv.html

[4]http://www.metoffice.gov.uk/climate-change

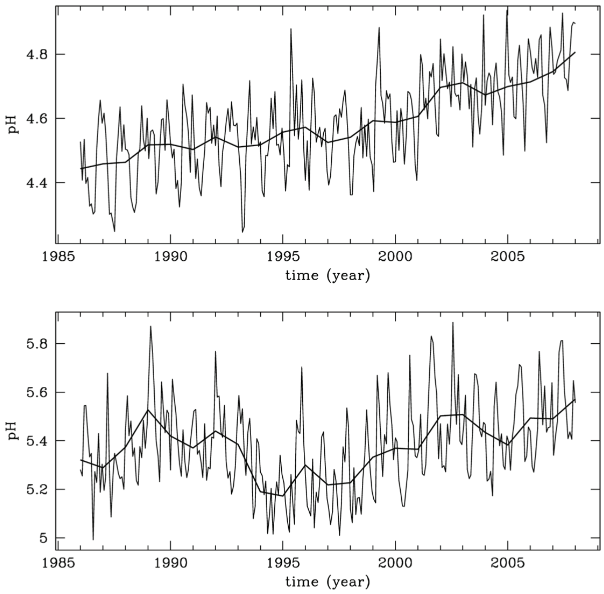

Fig. 7. Similar plots to Fig. 1, except that the upper panel and the lower panel result from stations of the United States in the east and in the west with respect to the longitude of 85°W.

Table 2. Similar to Table 1, except the pH is only averaged over the stations distributed in the eastern United States.

	Asymmetry	Solar Cycle	UV radiation	Temperature Anomaly
r	0.42	0.21	-0.01	0.75
P	0.12	0.16	0.47	0.00
r_s	0.01	-0.22	0.05	0.77
$P(r_s,N)$	0.48	0.15	0.40	0.00

Table 3. Similar to Table 1, except the pH is only averaged over the stations distributed in the western United States.

	Asymmetry	Solar Cycle	UV radiation	Temperature Anomaly
r	-0.41	0.32	0.42	0.23
P	0.13	0.06	0.02	0.13
r_s	-0.69	0.25	0.47	0.22
$P(r_s,N)$	0.02	0.12	0.01	0.15

of 85°W corresponds to the rough boundary where the pH level changes abruptly (e.g., NAPAP 2005, 2011). We continue to investigate the correlations of the pH data with several factors we have analyzed using two subsets of the pH data hoping to see how man-made sources of acid rain or natural cause can be differentiated in the results. The pH data obtained in the eastern stations, which are reckoned among region heavily polluted by sulfur oxides and nitrogen oxides, appear serious in that the value of pH is well below 5.6. On the other hand, the pH data obtained in the western stations suggest that rain is almost clean in the sense that the average of the measured pH is close to that of clean rain, that is, ~ 5.4.

As shown in Table 2, the pH data derived from the eastern stations appear not to be correlated with the solar north-south asymmetry, but with the temperature. In the case of the solar north-south asymmetry, the linear correlation coefficient and Spearman rank-order correlation value contradict each other so that their correlation values are less reliable. On the other hand, the linear correlation coefficient and chance probability for the temperature data are r=0.75 with P≈0.00. The Spearman rank-order correlation value for the temperature data is again found to be r_s=0.77 with $P(r_s, N)$≈0.00. The fact that the pH data set is not correlated with the solar north-south asymmetry may be understood in the sense that sulfur and nitrogen compounds from electricity generation plants in the eastern United States are rather influential. Table 3 summarize the obtained statistical parameters resulted from the pH data obtained from the western stations. The pH data obtained from the western stations appear to be anti-correlated with the solar north-south asymmetry. The linear correlation coefficient and chance probability for the solar north-south asymmetry are r=-0.41 with P=0.13. The Spearman rank-order correlation value for the solar north-south asymmetry data is again found to be r_s=-0.69 with $P(r_s, N)$=0.02. We speculate this property of anti-correlation of the pH data with the solar north-south asymmetry is due to natural traits. In addition, we note that the pH data obtained in the western stations are seemingly correlated with the Mg II core-to-wing ratio as a proxy of the solar UV radiation. The linear correlation coefficient and chance probability for the solar UV radiation are r=0.42 with P=0.02. The Spearman rank-order correlation value for the solar UV radiation is again found to be r_s=-0.47 with $P(r_s, N)$=0.01. We wonder this is also a part of natural characteristics. However, it should be pointed out that results from sunspot data are insufficient to support the idea that the pH data are correlated with the solar UV variation. Since the solar UV variation is an effective index of solar activity, if the pH data are correlated with the solar UV variation then it is expected to be correlated with a typical index of solar activity, such as, sunspot area.

4. SUMMARY AND CONCLUSION

It is widely accepted that acid rain is caused by emissions of sulfur dioxide and nitrogen oxide from man-made sources. Actual pathways, however, can be influenced by the prevailing atmospheric conditions, such as, sunlight, temperature. Here we have attempted to associate the pH in precipitation measured across the United States with the observed solar north-south asymmetry of sunspot area, which is one of important features of solar variability. We have analyzed the measured pH in precipitation over the 109 stations of the National Trends Network of the National Atmospheric Deposition Program distributed in the United States for the time interval from September in 1986 to January in 2008.

What we have found are mainly as follows:

(1) The data set of pH in precipitation measured in stations across the United States as a whole appears to be marginally anti-correlated with the observed solar north-south asymmetry. The obtained correlation coefficient and chance probability are r=-0.59 with P=0.04. The Spearman rank-order correlation value for the solar north-south asymmetry data is found to be r_s=-0.45 with $P(r_s, N)$=0.11. That is, rain apparently seems to become less acidic when the southern hemisphere of the Sun is more active. On the contrary, the acidity of rain measured across the United States as a whole is not directly correlated with the observed sunspot area, or solar activity.

(2) Since solar radiation in UV range and the atmospheric temperature are widely recognized as an important factor in the photodissociation process of the chemistry of acid rain, for comparison, we have repeatedly calculated correlation coefficients of pH in precipitation with the solar UV variability and the atmospheric temperature. The acidity of rain is not correlated with the solar UV variation. On the other hand, the pH data only appear to be correlated with the temperature.

(3) One may expect that the trend of temporal pH distribution in precipitation could exhibit different behaviors in two separate regions whose average pH level is different. To examine this idea we divide the group of stations into two, that is, stations located in the east and in the west with respect to the longitude of 85°W. The pH data derived from the eastern stations appear not to be correlated with the solar north-south asymmetry, but with the temperature. It may be because sulfur and nitrogen compounds from electricity generation plants in the eastern United States are rather influential. On the contrary, the pH data obtained from the western stations appear to be anti-correlated with the solar north-south asymmetry. Additionally, the pH data obtained from the western stations are again found to be correlated with the solar UV radiation.

We, therefore, conclude by pointing out that a role of the observed solar north-south asymmetry in the process of acidification of rain is to be further examined particularly when the level of pollution by sulfur oxides and nitrogen oxides is low.

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