lattice fringes are absent. If we assume that 2000 atoms are displaced by the alpha-recoil nucleus, we calculate that the maximum dimension of an alpha-recoil track is 2 to 5 nm. The measured size of the aperiodic areas in the HRTEMs is 1 to 5 nm. These aperiodic areas are similar in general appearance to the amorphous cascade regions in Bi^+-implanted silicon (*26*).

The radiation-induced structural change and resulting fractures caused by alpha-decay in this zircon may be the type that could occur in actinide-bearing, polyphase, ceramic nuclear waste forms. Additionally, this fine-scale variation in the alpha-decay dose may account in part for discordant ages from zircons determined by U-Th-Pb methods (*9*). The system of microfractures provides pathways for the addition or removal of uranium, thorium, and lead. In addition, damaged areas will be removed preferentially as a result of the increased solubility of metamict zircon compared to that of crystalline zircon (*27*).

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30. Interference colors are divided into orders according to whether they result from retardations of 0 to 550 μm (first-order colors), 550 to 1100 μm (second-order colors), 1100 to 1650 μm (third-order colors), and so on.
31. Electron microprobe analyses and the transmission electron microscopy were completed in the Electron Microbeam Analysis Facility in the Department of Geology and Institute of Meteoritics at the University of New Mexico, supported in part by the NSF, NASA, DOE–Basic Energy Sciences, and the state of New Mexico. This work was supported by the U.S. Department of Energy, Office of Basic Energy Sciences (grant DE-FG04-84ER45099) and Sandia National Laboratories. The instrumental neutron activation analyses were completed at Los Alamos National Laboratory by the Research Reactor Group by S. R. Garcia (Los Alamos National Laboratories) and H. E. Newsom (University of New Mexico).

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Acid Rain: China, United States, and a Remote Area

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The composition of precipitation in China is highly influenced by fossil fuel combustion and agricultural and cultural practices. Compared to the eastern United States, precipitation in China generally has higher concentrations of sulfate, ammonium, and calcium. Wet deposition rates of sulfur in China are 7 to 130 times higher than those in a remote area in the Southern Hemisphere. In many areas of the world, significant ecological changes have occurred in ecosystems that have acid deposition rates substantially less than those currently existing in China.

THE IMPACT OF ANTHROPOGENIC activities on precipitation composition and the subsequent effects on aquatic and terrestrial ecosystems have been investigated in North America, Scandinavia, and Europe over the past decade (*1*). However, only recently have data been available on the composition of precipitation in China (*2–5*). These data indicate that the composition of this precipitation is also strongly influenced by anthropogenic activities. To evaluate the degree of influence and the potential ecological effects on aquatic and terrestrial ecosystems, we compared the precipitation composition in China, the eastern United States, and a remote area in the Southern Hemisphere (Katherine, Australia). The ecological implications of these observed differences in precipitation composition are presented here.

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We used data from the Global Precipitation Chemistry Project, two U.S. precipitation chemistry networks, and previously unpublished data from two precipitation networks in China to address the following questions: (i) What is the composition of precipitation in China compared with the eastern United States and a remote area of the world? (ii) What are the reasons for the observed differences in composition? (iii) What are the ecological implications of the composition of precipitation in China?

The data are from remote, rural, and urban-suburban areas. We define remote areas as those locations where precipitation chemistry is removed from the direct influence of anthropogenic activities (*6*). Rural areas are those influenced by anthropogenic activities but not surrounded by large population centers. Urban-suburban areas are those surrounded by population centers or small factories.

The data that we used to characterize the precipitation chemistry at a remote continental area (Table 1) were from event precipitation collected in wet-only samplers at Katherine, Australia (Fig. 1) (*7*). Data for the eastern rural United States (Table 1) are for 1984 and are from five sites where event sampling was done with wet-only collectors (*8*) (Fig. 1). Data for the eastern urban United States were obtained in New York City from monthly samples collected in wet-only samplers, as part of the Environmental Measurements Laboratory Urban Network (*9*).

Precipitation composition data in China (Table 1) are also from 1984 and are from the Guizhou area in the south and the Beijing area in the north (Fig. 1). Precipitation was collected at one urban site in the north and at one urban and three rural sites in the south. In both areas, it was collected on an event basis (*10*).

The concentrations and wet deposition rates of SO_4^{2-} in precipitation in China are generally larger than in the United States. In one instance, Guiyang City, the concentrations are about six times higher than in New York City (Fig. 2). For several reasons, the higher concentrations and deposition are related to coal combustion. In China, coal is used as a primary fuel for electricity generation, home heating, and cooking. The coal is primarily burned in medium- and small-sized furnaces and small household stoves with short chimneys. Controls on the release of sulfur to the atmosphere are minimal (*3, 4*). The sulfur content of coal in the Guizhou Province is 3 to 5% (*5*), which accounts for the very high concentrations of SO_4^{2-} in precipitation in Guiyang City and the high concentrations in the adjacent rural areas. In Beijing, the sulfur content of the coal is about 1%, which probably accounts for the lower SO_4^{2-} content of the precipitation there compared to that of Guiyang City. These high concentrations of SO_4^{2-} in precipitation in China are not unique to these two cities. Zhao and Sun reported even higher SO_4^{2-} concentrations in precipitation from urban areas of Beijing and Tianjin (*3*).

On the basis of available data, in China the precipitation concentrations and wet deposition rates of NO_3^- are generally lower than in the United States (Fig. 2). The differences are caused by different patterns of fossil fuel use. In the United States, the high concentrations and deposition of NO_3^- are caused about equally by emission of nitrogen oxides (NO_x) from point and transportation sources (*11*). In China, the emission of NO_x to the atmosphere from fossil fuel combustion is primarily due to coal burning. Because of the low number of motor vehicles in China, they do not contribute significantly to NO_3^- in precipitation except in a few large cities (*4*). Of the areas discussed in this report, Beijing has the highest NO_3^- concentrations in precipitation; it also has the greatest density of motor vehicles (*12*).

The precipitation concentrations and wet deposition rates of Ca^{2+} are substantially greater in China than in rural and urban-

Table 1. The volume-weighted average (VWA) composition of precipitation and annual deposition at locations in China, the United States, and a remote area in Australia. The data from China and the United States (*8–10*) are from 1984. The data from Katherine, Australia, are for the period 1980–1984 (*7*). The number of storms analyzed is listed as *n*. The VWA concentrations are calculated from the sampled storms. Precipitation amount is total precipitation at the collection site. Collection efficiency is the percentage of total precipitation that was sampled. Deposition is calculated from the VWA concentrations and precipitation amount. The coefficients of variation of the VWA for China are in the range of 30 to 50%; for the United States and Katherine, Australia, the coefficients of variation are 5 to 15%. IEC, Institute of Environmental Chemistry.

Location	Area type	*n*	Precipitation amount (cm)	Collection efficiency (%)	VWA concentration (μeq liter^{-1})					Deposition (eq ha^{-1} year^{-1})				
					H^+	SO_4^{2-}	NO_3^-	NH_4^+	Ca^{2+}	H^+	SO_4^{2-}	NO_3^-	NH_4^+	Ca^{2+}
China														
Beijing (north)														
IEC	Urban-suburban	13	32.3	76	0.6	91.4	28.0	117	102.5	1.9	295	90.4	378	331
Guizhou (south)														
Guiyang City	Urban	47	117.1	80	95.2	447	10.0	60.4	248	1110	5230	117	707	2900
Shisun	Rural	79	105.2	99	18.6	76.9	7.8	18.9	62.3	196	809	82.1	199	655
Kaiyang	Rural	95	126.1	69	17.5	147	15.2	54.1	74.8	221	1850	192	682	943
Luizhang	Rural	102	107.5	86	12.3	119	7.3	21.8	99.3	132	1280	78.5	234	1070
United States														
New York City, NY	Urban	12	140.1	100	65.9	69.4	24.2	16.7	9.3	923	971	339	234	130
Ithaca, NY	Rural	79	125.4	98	58.4	51.4	27.7	14.0	5.2	732	644	347	176	65.2
Whiteface, NY	Rural	93	114.5	96	42.5	42.2	21.8	14.1	3.8	487	483	250	161	43.5
State College, PA	Rural	95	123.5	98	79.4	72.8	35.2	17.3	5.8	981	899	435	214	71.6
Charlottesville, VA	Rural	49	127.7	94	46.0	41.8	19.7	11.3	2.6	587	534	252	144	33.2
Champaign, IL	Rural	67	81.9	90	50.9	61.0	28.7	22.3	14.8	417	500	235	183	121
Australia														
Katherine	Remote	147	104.4	71	18.3	3.9	4.0	2.9	1.7	191	40.7	41.8	30.3	17.7

suburban areas of the eastern United States (Fig. 2). Three reasons for these differences follow: calcareous soils (in the north), small- and medium-sized furnaces that emit alkaline material and that lack particulate emission controls, and the extensive use of calcareous building materials (lime, limestone, and concrete) in populated areas of China (*3, 5, 13, 14*). The high Ca^{2+} concentrations in the precipitation of China affect the acidity of the precipitation (*15*).

The concentrations of NH_4^+ in the precipitation and the rates of wet deposition of NH_4^+ are generally greater in China than in the eastern United States. The two urban-suburban areas in China have higher concentrations than rural areas sampled (Fig. 2 and Table 1). The probable reasons for the higher concentrations and wet deposition rates in China are the widespread agricultural use of excretory wastes generated by the dense human population and the release of NH_3 from the high *p*H soils of the north (*4, 5, 15*).

The *p*H of precipitation is about 4 to 5 in southern areas and about 6.5 in northern areas of China (*3–5*). Compared to the eastern United States, these *p*H values are high given the high levels of SO_4^{2-} in China. The high *p*H values are due to the larger concentrations of bases (NH_3 and airborne particles, for example, $CaCO_3$) in the atmosphere of China (*5*). We suggest that these bases neutralize some of the precipitation acidity caused by H_2SO_4 and HNO_3 from fossil fuel combustion. Thus, the measured hydrogen ion concentration, $[H^+]$, of precipitation reflects the acidity after neutralization by atmospheric bases and is not a measure of the original acidity. We estimate the approximate upper limit of the original precipitation acidity by adding the equivalent concentrations of NH_4^+ and Ca^{2+} in the precipitation to the measured H^+ concentration. We assume that the NH_4^+ and Ca^{2+} in precipitation were originally present as NH_3 and $CaCO_3$. Available data on the mineralogy of soil and atmospheric particles suggest that much of the Ca^{2+} in the atmosphere was originally in the form of $CaCO_3$ (*16*). The calculated estimates of the original acidity, identified as $[H^+] + [NH_4^+] + [Ca^{2+}]$, are compared to the measured acidity $[H^+]$ (Table 2). In the south of China, the $[H^+] + [NH_4^+] + [Ca^{2+}]$ estimates of the original acidity are about four to ten times larger than $[H^+]$. In the northern site (Beijing) the $[H^+] + [NH_4^+] + [Ca^{2+}]$ estimate of the original acidity is more than two orders of magnitude greater than $[H^+]$, reflecting the greater concentrations of bases in the northern atmosphere. Therefore, if bases were not present in the atmosphere, the *p*H of precipitation would be about 3.5, substantially more acidic, on an annual average, than the values for the eastern United States.

Compared to the United States, China has higher concentrations of SO_4^{2-}, NH_4^+, and Ca^{2+} in precipitation. There is also greater spatial variability in the precipitation composition of China (Fig. 2). This variability may be due to the large number of ground-level emission sources in China (*5*).

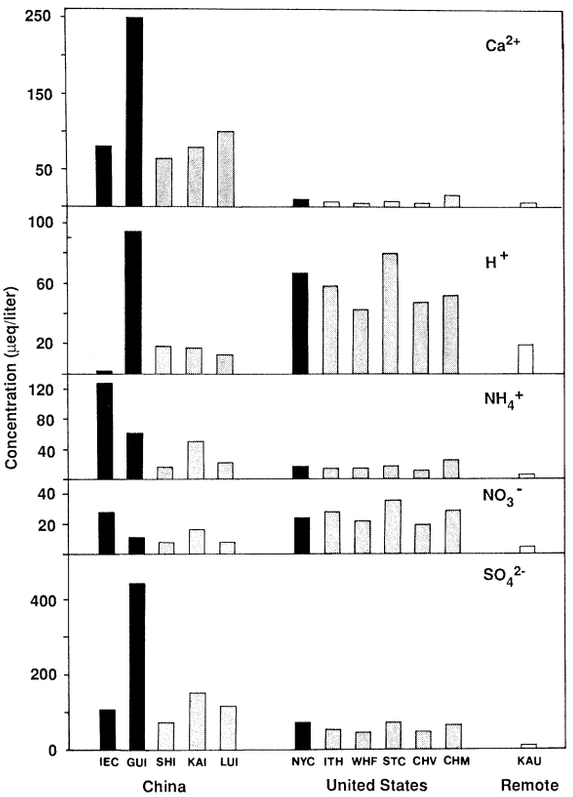

Fig. 2. The composition of precipitation in China, the United States, and a remote area in Australia. The solid bars represent urban-suburban areas; the shaded bars, rural areas; and the open bars, the remote area. Abbreviations are given in the legend to Fig. 1.

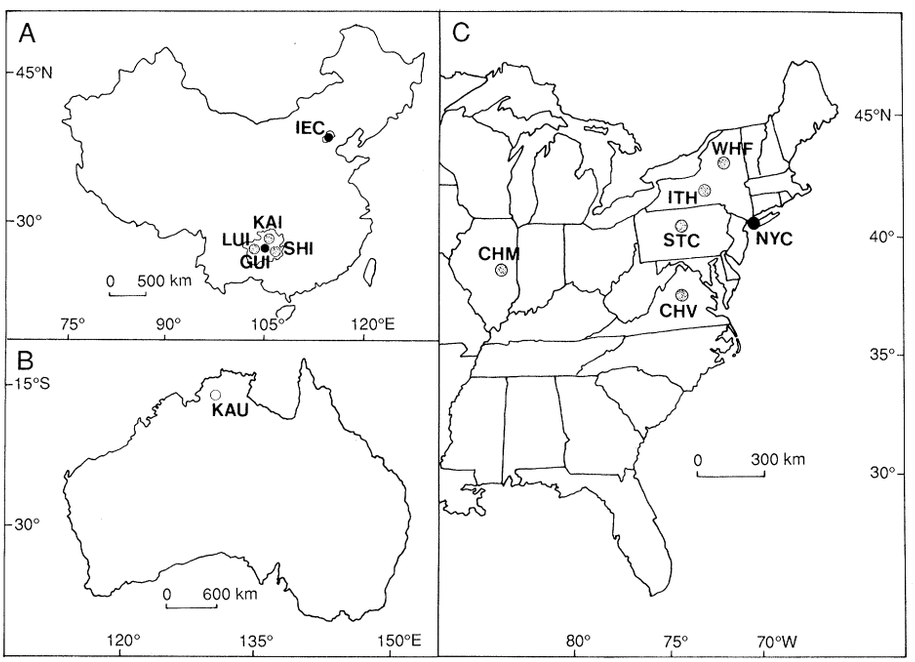

Fig. 1. Locations in (**A**) China, (**B**) Australia, and (**C**) the United States where precipitation was collected. The solid circles represent urban-suburban areas; the shaded circles, rural areas; and the open circle, a remote area. Abbreviations: IEC, Institute of Environmental Chemistry, Beijing; GUI, Guiyang City, Guizhau; SHI, Shisun, Guizhou; KAI, Kaiyang, Guizhou; LUI, Luizhang, Guizhou; NYC, New York City, NY; ITH, Ithaca, NY; WHF, Whiteface, NY; STC, State College, PA; CHV, Charlottesville, VA; CHM, Champaign, IL; KAU, Katherine, Australia.

Table 2. A comparison of the measured H^+ concentration $[H^+]$ of precipitation to the original acidity. IEC, Institute of Environmental Chemistry.

Location	$[H^+]$ (μeq/liter)	$[H^+] + [NH_4^+] + [Ca^{2+}]$ (μeq/liter)
	Guizhou (south)	
Guiyang City	95.2	404
Shisun	18.6	100
Kaiyang	17.5	146
Luizhang	12.3	133
	Beijing (north)	
IEC	0.6	220

The characteristics of China's precipitation—high *p*H and large and variable levels of SO_4^{2-} and Ca^{2+}—are very similar to that observed in Tennessee in the United States, between about 1915 and the early 1920s. MacIntyre and Young reported that the SO_4^{2-} concentrations of precipitation were 539 μeq/liter in the middle of the city of Knoxville, 282 μeq/liter 2.4 km from the city center, and 88 μeq/liter at a farm about 11 km from the city's center (*17*). The high SO_4^{2-} concentrations were matched by nearly equivalent amounts of bases. MacIntyre and Young postulated that the high concentrations of SO_4^{2-} and bases in precipitation were due to atmospheric emission of fossil fuel combustion products (*17*). The rapid decrease in concentrations away from the city center was believed to be caused by ground-level emission from short smokestacks and the rapid settling of the large particles. We believe this situation in Tennessee is similar to that which exists currently in China. If future pollution control in China involves the use of tall smokestacks to reduce local pollution problems, then it is expected that differences in precipitation composition in rural and urban areas will decrease and the magnitude of the regional impact of anthropogenic activities on precipitation composition will increase, as is currently the case in the United States.

The measured acidity of precipitation in China is low compared to the amount of SO_4^{2-} and NO_3^- present. However, for two reasons the low acidity does not adequately reflect the potential effects of atmospheric deposition on those ecosystems that receive it. First, SO_4^{2-} is a better indicator to use than H^+ because H^+ can be neutralized by bases (for example, NH_3 and $CaCO_3$). The reduction of the acidity, however, does not necessarily reduce the potential for acidification of aquatic and terrestrial ecosystems (*6*). A solution of $CaSO_4$ can cause short-term acidification of surface waters as a result of cation-exchange reactions in acid soils. Additions of $(NH_4)_2SO_4$ can acidify soils and surface waters by nitrification, a common process in soils (*18*). Second, data are presented here only for wet deposition. To accurately assess effects, total deposition must be known. The influence of dry deposition relative to wet deposition is likely to be much greater in China than in the United States because of significantly higher atmospheric concentrations of sulfur compounds in China (*3–5*).

To determine the enrichment of SO_4^{2-} that occurs in precipitation in China as a result of human activity, we compared the concentration of SO_4^{2-} in precipitation from a remote area in the Southern Hemisphere to the SO_4^{2-} level in precipitation of China (Table 1). The SO_4^{2-} in precipitation in China is enriched from 20 to 100 times relative to that in Katherine, Australia (Fig. 2). Similarly, NH_4^+ and NO_3^- enrichments in China vary from about 7 to 21 and 2 to 4, respectively, relative to Katherine, Australia; the Ca^{2+} enrichments in China are about 40 to 140 relative to Katherine, Australia. Because of the lack of data from both China and remote areas, however, these enrichments should be viewed only as estimates. The enrichments are substantial, however, and raise concern over potential ecological effects in China. Experience in North America and Europe has shown that excessive deposition of SO_4^{2-}, NO_3^-, and NH_4^+ (if oxidized to NO_3^- in the soil process of nitrification) can acidify soil and water systems (*1, 11, 18*). On the basis of studies in North America and Europe, it has been suggested that sulfate deposition of less than about 200 to 300 eq ha^{-1} $year^{-1}$ is required to protect sensitive aquatic ecosystems (*19*).

Thus, the extremely high deposition rates for SO_4^{2-} in China (295 to 5230 eq ha^{-1} $year^{-1}$) (Table 1) represent a major ecological concern. Aquatic ecosystems with low alkalinities, considered to be sensitive to acidification, are present in some areas of China, especially the south (*5*), but detailed surveys of alkalinities in small lakes and headwater streams are generally not available. Therefore, the potential for ecological deterioration in China is unknown at this time. However, in other areas of the world, significant and severe ecological changes have occurred in terrestrial and aquatic ecosystems at levels of atmospheric deposition of sulfur- and nitrogen-containing compounds that are less than those currently existing in China.

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