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Investigation on emission factors of particulate matter and gaseous pollutants from crop residue burning

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Abstract

Emission factors of particulate matter (PM), element carbon (EC), organic carbon (OC), SO_2, NO_x, CO, CO_2, and ten ions (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, F^-, Cl^-, NO_2^-, NO_3^-, SO_4^{2-}) were estimated from the domestic burning of four types of commonly produced crop residues in rural China: rice straw, wheat straw, corn stover, and cotton stalk, which were collected from the representative regions across China. A combustion tower was designed to simulate the cooking conditions under which the peasants burned their crop residues in rural China, to measure the emission factors. Results showed that wheat straw had the highest emission factor for the total PM (8.75 g/kg) among the four crop residues, whereas, corn stover and wheat straw have the highest emission factor for EC (0.95 g/kg) and OC (3.46 g/kg), respectively. Corn stover also presents as having the highest emission factors of NO, NO_x, and CO_2, whereas, wheat straw, rice straw, and cotton stalk had the highest emission factors of NO_2, SO_2, and CO, respectively. The water-soluble ions, K^+ and Cl^-, had the highest emission factors from all the crops. Wheat straw had a relatively higher emission factor of cation species and F^-, Cl^-, NO_2^- than other residues.

Key words: rural China; crop residues; combustion tower; emission factor

Introduction

Crop residues are one of the oldest domestic fuels. Currently, crop residues are the fourth largest energy resource after coal, oil, and natural gas. There are about one-half of the people in the world who use crop residues for domestic heating and cooking, especially in many parts of the developing world. Before 1979, the crop residue energy had covered about 70% of the rural energy consumption in China. Up to now, the crop residue energy is still an important part of the rural energy for cooking or heating in China (Edwards *et al*., 2004), even though the energy conversion efficiency is only 10% to 20% (Li *et al*., 2001; Yuan *et al*., 2002).

Crop residue burning results of emissions from particle matter (PM) (Andreae and Merlet, 2001; Reddy and Venkataraman, 2002), appear with a large carbonaceous fraction (Liousse *et al*., 1996; Turn *et al*., 1997), and inorganic water-soluble ions (Rau, 1989). The two carbonaceous aerosol types are organic carbon (OC), which mainly scatters radiation and cools the atmosphere (direct forcing) and element carbon (EC), which absorbs solar radiation and results in heating the atmosphere (Liousse *et al*., 1996; IPCC, 2001; Menon *et al*., 2002). The water-soluble inorganic ions (e.g., potassium, sodium, chloride, and calcium) and hygroscopic organic compounds in crop residue combustion particles would act as cloud condensation nuclei (CCN), leading to a net reduction of solar radiation received on the Earth's surface (indirect forcing) (Penner *et al*., 1992). Crop residue combustion also results in emissions of greenhouse gases (GHG) such as carbon dioxide (CO_2), the main driving force for the past global climate change (Zhang *et al*., 2000; IPCC, 2001; Andreae and Merlet, 2001).

Emissions from crop residues sources are known to be spatially and temporally inhomogeneous, needing better characterization. China has a large population, 70% of which lives in rural areas. Because of the large use of crop residues, the pollutant emissions from the combustion, such as, PM, NO_x, and CO, have contributed substantially to the regional environment pollution problems (Duan *et al*., 2001; Wu *et al*., 2001; Lan *et al*., 2002).

Previous research efforts have obtained some emission factors of crop residue combustion in China (Zhang *et al*., 2000) and other countries (Smith *et al*., 1993; Turn *et al*., 1997; Andreae and Merlet, 2001). Of the limited researches in China, emission factors of greenhouse gases and other airborne pollutants from different types of stoves were reported, but the experimental materials were

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collected from places near the laboratories, which were not representative of China.

A more accurate inventory for aerosols and gaseous pollutants from the crop residue combustion for China is needed as an input to the regional scale air quality and climate studies. The objective of this article was to develop realistic emission factors of aerosols and gaseous pollutants to represent crop residues burning in domestic cooking stoves.

1 Experimental methods

1.1 Experimental design and sampling approach

A combustion tower was designed and installed in the laboratory to simulate the condition that peasants in rural China use, such as, crop residues for cooking in traditional brick stoves. The characteristics of the traditional brick stoves were described in detail by Edwards *et al*. (2004). For the design methodology, literature was referred to, which was described in detail elsewhere (Darley, 1977, 1979; Venkataraman and Rao, 2001) and summarized here. The tower is in the form of an inverted funnel with a cylindrical bottom, 1.2 m in diameter and 0.4 m high. From the top of the cylinder, the tower decreases to 0.2 m in a length of 1.0 m, and is topped with a stack 1.2 m in height. There is a combustion table (0.4 m × 0.4 m) in the form of net, which is made up of a steel bar with each angle having supports 0.1 m in height. The schematic sketch of the burning tower is shown in Fig.1, along with locations of sampling points. The sample site for gases, PM, and for recording temperatures and velocity of airflow is in the stack about 0.2 to 0.35 m below the top. A thermocouple temperature sensor was used for recording temperatures, and a Pitot tube was used for gas velocity measurements. Several factors may influence the estimation of the emission factors, such as, moisture content of the straw, combustion temperature, and ambient conditions. Therefore, the system is optimized to minimize combustion modification from the induced draft, ensuring that combustion temperature is not reduced by dilution from the draft.

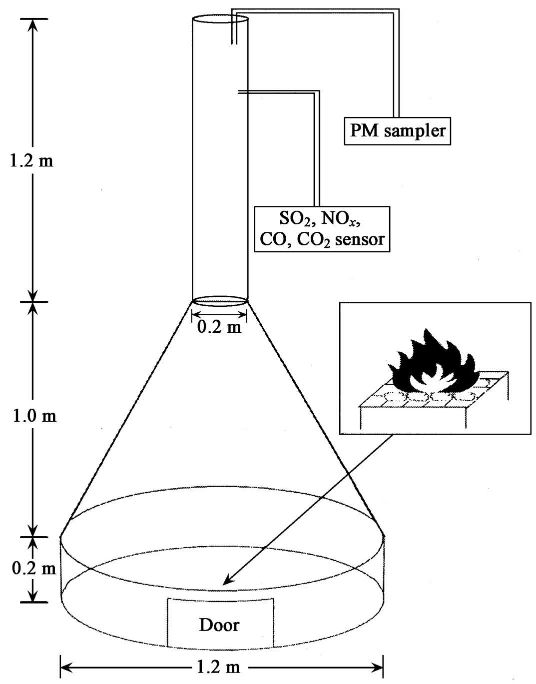

Fig. 1 Schematic diagram of burning tower with sampling attachment.

The water boiling test (Sinton *et al*., 1995; Zhang *et al*., 2000) is a standard international method of simulating a cooking task in a rural household. A known amount of water is boiled in a high power phase, and kept simmering during a low power phase. Burning cycles last approximately 10 to 20 min.

1.2 Source of experimental material

On the basis of the statistical data from MOA (Ministry of Agriculture of China, 2001) and NBSC (National Bureau of Statistics of China, 2001), the crop residues and stalk output in China has reached about 600 million tons in recent years (Cao *et al*., 2006), Among them, three kinds of cereal crops, that is, rice, wheat, and corn, account for about 19%, 17%, and 15% of the total grown area separately and the straw amounts produced account for about 18%, 21%, and 37.5% of the total amount separately. Of the straw, about 58.7% (Li *et al*., 1998) to 47% (Yevich and Logan, 2003) is used as fuel.

The straw used for this experiment was collected from the representative places in all parts of China, including wheat straw, rice straw, corn stover, and cotton stalk, as shown in Table 1. These straw types generally represent the crop residues being consumed in those regions. All crop residues were air dried at least one month before the experiment. The composition of the crop residues in China (C, H, O, N) are described in detail elsewhere (Zhang *et al*., 1997, 2000; Liu *et al*., 2001).

1.3 Experimental process

The experimental study was carried out at the Institute of Earth Environment in Xi'an, China, and lasted for 2 d. The combustion of crop residues from each place was repeated at least twice, so a total of 33 tests were conducted: 8 wheat straw, 14 rice straw, 9 corn stover, and 2 cotton stalk.

Before each combustion test, the straw was first weighed with a balance and then put on the burning table and ignited. The weight of fuel to be combusted was about 0.5–1.9 kg for each test. After reaching steady burning in about half a minute, the sampling instrument and test equipment began to work at the same time (Jenkins *et al*., 1996). All the burning was ventilated naturally. Each combustion process lasted generally 6–15 min. After each combustion test, the ash of the burned straw was collected into a clean plastic bag for safekeeping.

1.4 Analysis methods

PM: quartz fiber filters of 47 mm in diameter (Whatman International Ltd., UK) were used for PM sampling. The filters were preheated at 800°C for 4–6 h and then placed in a desiccator for at least 24 h before using. The filters were carefully placed in the filter holders and used for sample

Table 1 Crop residues and stalk collect site

Straw type	Collect site	Collect reason
Wheat straw	Shannxi, Hebei	The wheat cultivated area of Huang-Huai-Hai Plain accounts for 60% of the cultivation of the whole country
Rice straw	Jiangsu, Hubei, Guangxi, Sichuan	The rice cultivated area in the Yangtze River valley accounts for 65.7% of the cultivation oof the whole country
Corn stover	Shandong, Jilin, Henan, Mongolia	The corn cultivated area of North-east China and Hebei, Shandong, Henan provinces accounts for 55% of the cultivation of the whole country
Cotton stalk	Xinjiang	The cotton cultivated area in Xinjiang Province accounts for 31% of the cultivation of the whole country

collection. After sampling, the filters were taken out of the holder and placed in a petri dish, desiccated for 24 h and weighed by electronic micro-balance with 1 μg sensitivity (ME 5-F, Sartorius Corp., Germany).

EC/OC was analyzed using the Thermal/optical carbon analyzer (Model 2001, Desert Research Institute, USA), with the help of the IMPROVE thermal/optical reflectance (TOR) protocol. The detailed procedures were given elsewhere (Chow *et al*., 1993). The detection limits for EC/OC were below 1.0 mg/m^3. The difference determined from replicate analyses was smaller than 5% for TC (total carbon) and 10% for EC/OC ratios.

Elemental composition: ten ions (NO_2^-, NO_3^-, SO_4^{2-}, F^-, Cl^-, K^+, Na^+, NH_4^+, Ca^{2+}, Mg^{2+}) in PM were analyzed by Ion chromatography (DX-600, Dionex Corp., Wommelgem, Belgium). Ionpac AS11-HC (4 mm, Dionex) column and Ionpac AG11-HC (4 mm, Dionex) were used for anion separation.

Accuracy and precision were checked by external standards. Precision in all instances was better than 5% RSD (residual standard deviation) and accuracy better than 4%. The detection limits were 5 μg/L for F^- and Cl^-, 15 μg/L for NO_3^-, NO_2^-, K^+, Na^+, NH_4^+, and 20 μg/L for SO_4^{2-}, Ca^{2+}, Mg^{2+}.

Gaseous pollutants: the concentrations of gaseous products, such as, SO_2, NO, NO_2, CO, and CO_2 in flue gas were measured by the online combustion analyzer (KM9106, Kane Corp., UK). The resolutions of SO_2, NO, and NO_2 were 1 ppmv, CO was 0.03%, and CO_2 was 0.3%. The detection range was 0–5000 ppmv for SO_2 and NO, 0–1000 ppmv for NO_2, 0–10000 ppmv for CO and 0–20% for CO_2. The instrument was started 3 min earlier in clean air to test the equipment, and then the normal operation began after steady burning was reached.

1.5 Computing method

Emission factors of gaseous species were calculated as the following equation and it was similar to that described by Jenkins *et al*. (1996):

$$E_i = \frac{10^{-3}}{m_{\rm fd}} \int_{t_0}^{t_f} A_s u C_i \frac{w_i}{22.4} {\rm d}t \tag{1}$$

where, E_i is the emission factor of species i, $m_{\rm fd}$ is the mass of crop residues consumed during each test, t_0 is the starting time of each test, t_f is the finishing time, $A_{\rm s}$ is the stack area (0.03 m^2), u is the average stack gas velocity, C_i is the sampling concentration of species i, and W_i is the molecular weight of species i.

For PM or aerosols, an emission factor, E_i was calculated as:

$$E_{i,j} = \frac{1}{m_{\rm fd}} A_{\rm s} \overline{u} \frac{m_{k,i}}{v_0} \eta \frac{T_i}{T_s} \tag{2}$$

where, $E_{i,j}$ is the emission factor of PM or aerosols, $m_{k,i}$ is the sampling weight of species i, v_0 is the air pump work flow rate, η is the mass content of aerosol in PM, T_i is the absolute ambient air temperature, and T_s is the absolute stack gas temperature.

2 Results and discussion

Table 2 presents aerosols, ions, and gaseous pollutant emissions data for each straw type. The compositions of the PM emitted from crop residues are shown in Table 3. For easy comparisons, the emission factors, from literature, of crop residues burning, are presented in Table 4. The authors have estimated the error for each calculated emission factor by the coefficient of variation (CV, or the standard deviation divided by the mean).

2.1 Particulate matter

Varying emission of PM was observed in the study for different straws. The PM emission rate was 4.53 to 8.75 g/kg (Table 2), which closely matched the values reported in the literature (Table 4). Wheat straw had the highest emission factor of PM (8.75 g/kg), and for the others it was relatively low. The average emission factor of PM for four types of straws (6.21 g/kg) was lower than the values reported in the literature (Table 4).

EC emission rates were 0.42 to 0.95 g/kg, being relatively lower from wheat straw, and higher from corn stover (Table 2). OC emission rates were 1.83 to 3.46 g/kg with relatively lower contributions from cotton stalk, and higher from wheat straw. EC and OC emission factors closely matched the values reported in the literature (Table 4).

For the water soluble ions presented in PM, their computed emission factors were low. For cation species, such as, Na^+, K^+, NH_4^+, Mg^{2+}, and Ca^{2+}, their average emission factors varied from nd (not detected) to 0.837 g/kg (Table 2). The emission factor of K^+ was the highest, consistent with the values reported in the literature (Turn *et al*., 1997). For anion species, such as, NO_2^-, NO_3^-, SO_4^{2-}, F^-, Cl^-, the average emission factors varied from nd to 0.949 g/kg, and the emission factor of Cl^- was the highest, matching the values reported in the literature (Table 2). Wheat straw had relatively higher emission factors of cation species of F^-, Cl^-, NO_2^-. Rice straw had the highest emission factor of Mg^{2+} and NO_3^-, corn stover has the highest emission factor of Ca^{2+}, whereas, cotton stalk has

Table 2 Average emission factors for various straw types (g/kg)

Pollutant type	Straw	Wheat straw	Rice straw	Corn stover	Cotton stalk
Aerosol	PM	8.75±4.18	6.28±1.59	5.31±1.79	4.53±0.95
	EC	0.42±0.23	0.49±0.21	0.95±1.08	0.82±0.20
	OC	3.46±2.05	2.01±0.67	2.25±0.74	1.83±0.54
Cation	Na^+	0.098±0.097	0.077±0.035	0.095±0.060	0.102±0.009
	NH_4^+	0.011±0.024	0.003±0.008	0.001±0.002	nd
	K^+	0.837±0.377	0.715±0.533	0.180±0.080	0.735±0.058
	Mg^{2+}	0.005±0.004	0.006±0.005	0.001±0.002	nd
	Ca^{2+}	0.068±0.030	0.061±0.029	0.072±0.055	0.046±0.002
Anion	F^-	0.003±0.002	0.001±0.002	0.003±0.003	nd
	Cl^-	0.949±0.378	0.855±0.491	0.372±0.104	0.726±0.054
	NO_2^-	0.007±0.012	0.006±0.012	0.007±0.013	nd
	NO_3^-	nd	0.047±0.076	0.017±0.048	nd
	SO_4^{2-}	0.180±0.058	0.182±0.072	0.134±0.080	0.216±0.020
Gasous	SO_2	0.04±0.04	0.18±0.31	0.04±0.04	nd
	NO	1.93±0.90	3.09±0.91	3.25±0.72	2.28±0.21
	NO_2	0.35±0.11	0.33±0.17	0.34±0.13	0.22±0.02
	NO_x	2.28±1.00	3.43±1.08	3.60±0.85	2.49±0.23
	CO	57.78±24.75	67.98±25.58	67.64±13.01	105.82±6.02
	CO_2	1377.72±431.12	1674.12±452.26	2327.14±709.57	1345.42±108.02

nd: not detected. NO_x =NO + NO_2.

Table 3 Average chemical composition in PM (%)

	EC	OC	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	F^-	Cl^-	NO_2^-	NO_3^-	SO_4^{2-}	Total
Wheat straw	4.80	39.54	1.12	0.12	9.56	0.06	0.77	0.03	10.85	0.08	–	2.06	68.99
Rice straw	7.80	32.01	1.22	0.05	11.39	0.10	0.97	0.01	13.61	0.09	0.74	2.89	70.88
Corn stover	17.89	42.37	1.78	0.01	3.39	0.02	1.35	0.06	7.00	0.13	0.32	2.53	76.86
Cotton stalk	18.10	40.40	2.26	–	16.23	–	1.03	0.01	16.02	–	–	4.77	98.82

Table 4 Published emission factors of crop residues burning (g/kg)

Emission factor	TSP	PM_{10}	EC	OC	Na^+	NH_4^+	K^+	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	SO_2	NO_x	CO	CO_2
Wheat straw	12[c] 5.82[d] 4.73[e] 13[h]	5.74[d]	1.2[c] 0.8[f]	2.2[f]	0.018[f]	0.19[f]	0.9[f]	0.004[f]	1.2[f]	0.021[f]	0.26[f]	0.47[d] 0.031[e]	2.33[d] 1.14[e] 2.79[h]	66.69[d] 61.1[e] 5[h]	1194.88[d] 2.1320[e] 1400[h]
Rice straw	5.3[c] 3.49[d]	3.46[d]	0.86[c] 0.5[f]	0.94[f]	0.021[f]	0.21[f]	0.48[f]	0.016[f]	0.94[f]	0.008[f]	0.12[f]	0.62[d]	2.84[d]	31.39[d] 48.7[i]	1162.15[d] 1101[i]
Corn stover	12[c] 6.31[d] 1.68[e]	6.21[d]	0.96[c] 0.75[f]	1.8[f]	0.003[f]	0.31[f]	0.73[f]	0.007[f]	1.4[f]	0.014[f]	0.1[f]	0.2[d] 0.015[e]	0.75[d] 1.27[e]	38.78[d] 40.3[e]	1313.71[d] 1160[e]
Cotton stalk															
Combined	11[a] 8.05[e] 13[g]		0.69[g] 0.78[f]	5.0[b] 1.9[f] 3.3[g]	0.024[f]	0.17[f]	0.77[f]	0.011[f]	0.92[f]	0.015[f]	0.24[f]	0.4[g] 0.22[e]	2.5[g] 7.0[e]	58[a] 92[g] 86.3[e]	1515[g] 1130[e]

[a]USEPA; [b]Streets *et al*., 2003; [c]Liousse *et al*., 1996; [d]Jenkins *et al*., 1996 ; [e]Zhang *et al*., 2000 (Brick stove); [f]Turn *et al*., 1997; [g]Andreae and Merlet, 2001; [h]Ortiz de Zárate *et al*., 2000; [i]Smith *et al*., 1993, 2000 (Traditional mud stove).

the highest emission factor of SO_4^{2-}.

The calculated average CV of emission factors were: ±31.9% for PM, ±58.9% for EC, ±38.7% for OC, ±54.1% for Na^+, ±228.3% for NH_4^+, ±43.0% for K^+, ±121.1% for Mg^{2+}, ±43.1% for Ca^{2+}, ±122.2% for F^-, ±33.2% for Cl^-, ±185.7% for NO_2^-, ±222.0% for NO_3^-, and ±35.2% for SO_4^{2-}. Calculated emission factors of NH_4^+ and NO_3^- have the highest CV.

2.2 Gaseous pollutant emission

The concentration of gaseous emissions from different straws increased quickly in the combustion tower only after 0.5 min of ignition and then remained in a relatively steady condition. The trend observed for rice straw, which was one type of straw used in the study, is shown in Fig.2. After the fire died out, gaseous emissions would maintain for 1.5 min, and then its concentration reduced gradually.

Gaseous emissions of various types of straws are given

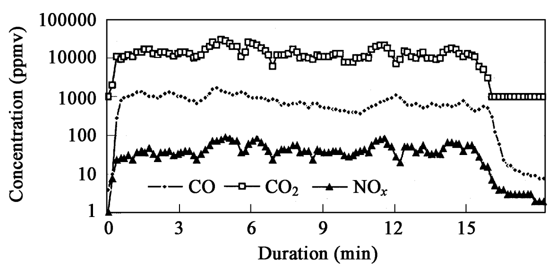

Fig. 2 Concentration of various gaseous emissions during a case of crop residue burning in combustion tower.

in Table 2. Except for SO_2, computed emission factors from straw closely matched the values reported in literature, especially the values for the household stoves in China (Zhang *et al*., 2000). SO_2 was lower than the values reported in literature. Corn stover had the highest emission factors of NO, NO_x and CO_2, whereas, wheat straw, rice straw, and cotton stalk has the highest emission factors of NO_2, SO_2, and CO, respectively.

It was interesting to note that the emission factor of SO_2 was about one-fifth of the value from literature. As high-concentration fertilizers with very low sulfur content were largely used in China, the Chinese soil was in the state of lower sulfur content (Cui and Wang, 2002) and consequently the content of sulfur in the crop residues was relatively lower. Result analyses showed that the difference in emission factors of SO_2 was large for each straw and rice straw was noticeably higher than the others. Based on the existing data (Zhang *et al*., 1997; Liu *et al*., 2001), elemental compositions of crop residues in China had little difference, about 38%–45% of C, 5%–6% of H, and 0.15%–2.6% of N, but the mass content of S was very low (nd–0.2%). It was also found that the emission factors of SO_2 from crop residue combustion in South China were clearly higher than those in North China. That might be because of acid rain depositions, which were relatively serious in South China (Wang *et al*., 2000).

The CV of emission factors of gases were: ± 124.1% for SO_2, ± 26.9% for NO, ± 32.6% for NO_2, ± 27.0% for NO_x, ± 26.3% for CO, and ± 24.2% for CO_2. These values for uncertainties are similar to the estimates made by Zhang *et al*. (2000).

2.3 Particle matter chemical composition

The average mass fraction in the PM is presented in Table 3. For the carbonaceous aerosols, EC mass content in the PM is 4.8%–18.1% and OC is 32.01%–42.37%. As it is known from earlier studies that the majority of PM (about 50%) emission from crop residue combustion is carbonaceous aerosol (Turn *et al*., 1997), these results in China reinforce the findings (Table 3).

For water soluble species, the mass content in the particulate has a great difference. With the exception of cotton stalk, Cl^- has the highest mass content with values ranging from 7.0%–16.02%, followed by K^+, SO_4^{2-}, and Na^+. Other species present with less than 1% in the particulate. Cotton stalk has the highest mass contents of Na^+, K^+, Cl^-, and SO_4^{2-}, but other species have very little NH_4^+. Mass content for wheat straw is higher in the PM. Corn stover has the highest mass content of Ca^{2+} and F^-, rice straw has the highest mass content of Mg^{2+} and NO_2^-.

3 Conclusions

Emission factors have been experimentally determined for aerosols, gaseous pollutants, and water-soluble ions from the simulated domestic burning of four types of crop residues in China: rice straw, wheat straw, corn stover, and cotton stalk. Combustion tower experiments indicate that wheat straw has the highest emission factor of PM (8.75 g/kg), corn stover has the highest emission factor for EC (0.95 g/kg), and wheat straw has the highest emission factor of OC (3.46 g/kg). For the water-soluble ions, the emission factor of K^+ and Cl^- are the highest; Wheat straw has relatively higher emission factors of cation species and F^-, Cl^-, and NO_2^-. Corn stover has the highest emission factors of NO, NO_x, and CO_2. Wheat straw, rice straw, and cotton stalk have the highest emission factors of NO_2, SO_2, and CO, respectively. However, the emission factor for SO_2 is about one-fifth of the values from the literature. As there is a general lack of understanding concerning emissions from rural China, this study has improved local knowledge and measurement of emissions of aerosols and gases from the domestic burning of crop residues in China, and has paved the way for a more accurate estimate of emissions that have air quality and climate impacts in China.

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