Chapter 3
Acid Deposition

Hiroyuki Sase

Abstract Acid deposition is a regional issue in Asia. Emission levels of acidic substances, such as SO_2 and nitrogen oxides (NO_X), are still very high, although SO_2 emissions in China have started declining. The Acid Deposition Monitoring Network in East Asia (EANET), the regional network covering Northeast and Southeast Asia, has been monitoring acid deposition and its effects on forests and inland waters. Wet deposition levels of sulfur and nitrogen in EANET countries are significantly higher than those in Europe and the United States, although total deposition, as well as dry deposition, has not been sufficiently evaluated in the region, particularly in forested areas. Soil acidification is a potential risk for plant growth in Asia. Soil acidification has been observed in areas receiving high acid deposition in China and Japan. Observational data on acid deposition and soil chemical properties should be accumulated for forest areas to ascertain the risk for plants in the region.

Keywords Acid Deposition Monitoring Network in East Asia (EANET) • Wet deposition • Dry deposition • Soil acidification

3.1 Introduction

Due to rapid economic growth in recent decades, emissions of acidic substances, such as SO_2 and nitrogen oxides (NO_X), have been increasing in Asian countries. Anthropogenic SO_2 emissions in East Asia (Japan, China, and the Republic of Korea) have been increasing significantly since the 1950s, from approximately 3,000 Gg SO_2 y^{-1} in the 1950s to 35,000 Gg SO_2 y^{-1} in 2005. Emissions in Southeast Asia have also been gradually increasing, while emissions in Europe and North America peaked in the 1970s/1980s and rapidly decreased thereafter (Smith et al. 2011). According to Ohara et al. (2007), from 1980 to 2003, the Asian emissions of SO_2 and NO_X increased by 119 % and 176 %, respectively. In particular, NO_X

H. Sase

Asia Center for Air Pollution Research, 1182 Sowa, Nishi-ku, Niigata City 950-2144, Japan
e-mail: sase@acap.asia

emissions in China increased by 280 % from 1980 to 2003 (Ohara et al. 2007). Updated emission inventories in Asia (Kurokawa et al. 2013) continue to show high growth rates from 2000 to 2008, with rates of 34 % for SO_2 and 54 % for NO_X. SO_2 emissions in China peaked in 2006 and started decreasing gradually thereafter (Lu et al. 2011), while NO_X emissions were still increasing as of 2010 (Zhang et al. 2012). The emission levels of both SO_2 and NO_X in Asia remain high compared with those in the 1950s, and therefore the actual emission trend of SO_2 and the growth rate of NO_X emissions should be carefully monitored henceforth.

Atmospheric deposition of acidic substances, hereafter referred to as "acid deposition", is a concern for the general public as "acid rain", which has been blamed for causing soil acidification (e.g., Hallbäcken and Tamm 1986), forest decline (e.g., Schulze 1989), and the decline of freshwater fish populations (Wright et al. 1976) in Europe and North America. Therefore, the monitoring of acid deposition and its effects on terrestrial ecosystems has also been conducted at the national level in Japan and China since the 1980s (Zhao et al. 1988; Tamaki et al. 2000) and at the regional level by the Acid Deposition Monitoring Network in East Asia (EANET) since 2001 (EANET 2014). Relevant studies on soil and inland water acidification have been conducted, particularly in Japan and China. In this chapter, we review the current status of acid deposition and soil acidification, as a potential inhibitor of plant growth, in Northeast and Southeast Asia, based on EANET data and other observational studies.

3.2 Acid Deposition Monitored by a Regional Network in East Asia

The EANET is an intergovernmental regional network that was formed by 13 countries in Northeast and Southeast Asia. The EANET started its preparatory-phase activities in 1998 and regular-phase activities in 2001. It has been monitoring wet deposition (precipitation), dry deposition (gaseous and particulate matter), soil chemical properties, forest vegetation (tree growth and species composition of understory vegetation), inland water chemistry, and biogeochemical processes in forest catchments. Monitoring data from 2000 onwards are disclosed to the public on the EANET website (www.eanet.asia).

3.2.1 Wet Deposition

From 2000 to 2009, the annual mean pH values of rainwater from 57 sites in 13 countries ranged from 4.18 to 6.94, with an arithmetic mean of 5.07 and a median of 4.94, according to the second EANET periodic report (EANET 2011). During this period, pH values below 4.30 were recorded at two sites (Jinyunshan and Haifu) in Chongqing, China, one site (Petaling Jaya) in Selangor, Malaysia, and one site
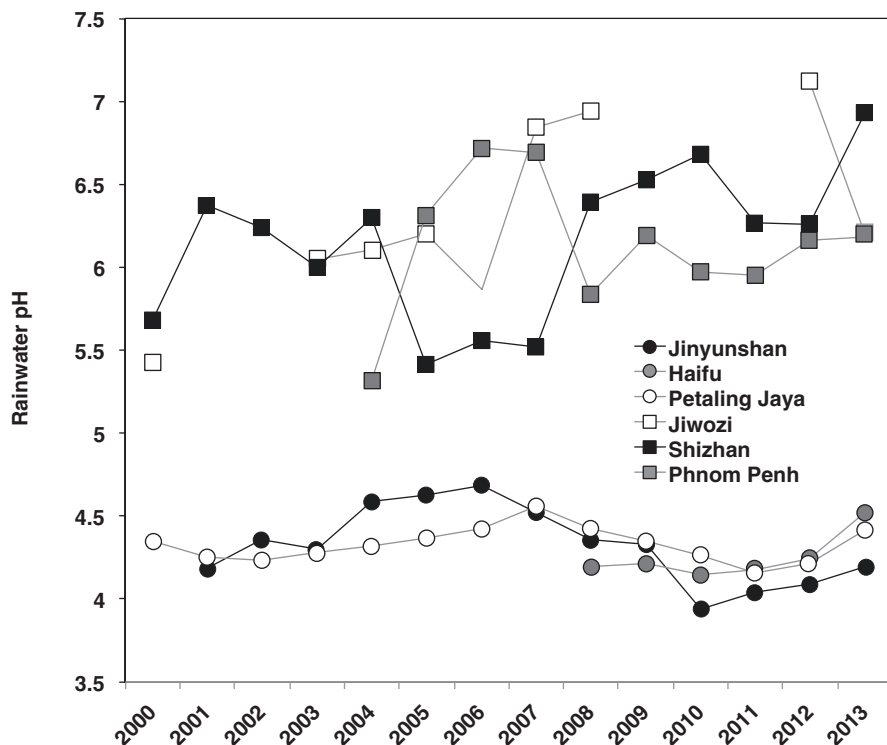
Fig. 3.1 Annual changes in rainwater pH at the EANET (Acid Deposition Monitoring Network in East Asia) sites. Sites with significantly high or low pH values are shown here – Jinyunshan and Haifu, in Chongqing, China; Petaling Jaya, in Selangor, Malaysia; Jiwozi and Shizhan in Xi'an, China; and Phnom Penh, Cambodia

(Kanghwa) in the Republic of Korea. In contrast, pH values above 6.5 were observed in two sites in Xi'an, China (Jiwozi and Shizhan) and one site (Phnom Penh) in Cambodia (EANET 2014). The annual variation of pH at these sites for the past decade is shown in Fig. 3.1 (after EANET 2014). Although the acidity of rainwater may fluctuate year by year depending on its chemical composition and precipitation amount, rainwater in China and Malaysia appears to have been significantly acidic for the past decade. However, at two sites in Xi'an, China, the rainwater was strongly neutral.

Wet deposition levels of non-sea-salt (nss) SO_4^{2-}, dissolved inorganic nitrogen (DIN), and nss-Ca^{2+} at the EANET monitoring sites from 2009 to 2013 are summarized in Table 3.1. Two sites (Jinyunshan and Haifu) in Chongqing, and the Petaling Jaya site, had significant depositions of nss-SO_4^{2-} and DIN. This may contribute to the low pH values at these sites. The Shizhan site in Xi'an also had high deposition of nss-SO_4^{2-} and relatively high deposition of DIN (94 mmol $m^{-2} y^{-1}$; not shown in Table 3.1), although the rainwater pH was significantly high. The high level of wet nss-Ca^{2+} deposition may have contributed to neutralization of the acids.

Table 3.1 Wet deposition amounts of non-sea-salt SO_4^{2-}, dissolved inorganic nitrogen (DIN), and non-sea-salt Ca^{2+} from 2009 to 2013 in the EANET sites

Parameter	Percentile, mmol m^{-2} y^{-1} and site name with the mean value		
	10%	50%	90%
Non-sea-salt SO_4^{2-}	7.5	20	53
Site name (mean), "≤10%" or "90% ≤"	Listvyanka, RU (7.0)		Jinyunshan, CN (121)
	Ogasawara, JP (6.1)		Haifu, CN (113)
	Khanchanaburi, TH (5.3)		Bandung, ID (63)
	Chiang Mai, TH (4.7)		Shizhan, CN (60)
	Mondy, RU (0.74)		Petaling Jaya, MY (60)
DIN, $NO_3^- + NH_4^+$	19	56	133
Site name (mean), "≤10%" or "90% ≤"	Ochiishi, JP (19)		Haifu, CN (196)
	Ogasawara, JP (13)		Jinyunshan, CN (179)
	Listvyanka, RU (14)		Petaling Jaya, MY (178)
	Danum Valley, MY (10)		Bandung, ID (155)
	Mondy, RU (2.4)		Serpong, ID (133)
Non-sea-salt Ca^{2+}	3.7	12	42
Site name (mean), "≤10%" or "90% ≤"	Rishiri, JP (3.3)		Shizhan, CN (76)
	Nakhon Ratchasima, TH (3.2)		Haifu, CN (60)
	Ochiishi, JP (2.3)		Jinyunshan, CN (58)
	Ogasawara, JP (1.9)		Cuc Phuong, VN (57)
	Mondy, RU (0.97)		Mt. Sto. Tomas, PH (52)

After EANET 2014

Note: Total of 47 sites (whose annual data were available for at least 3 years)

CN China, *ID* Indonesia, *JP* Japan, *MY* Malaysia, *PH* Philippines, *RU* Russia, *TH* Thailand

At the sites in Chongqing, wet deposition levels of nss-Ca^{2+} were similar to those at the Shizhan site. However, as shown in Table 3.1, the deposition levels of nss-SO_4^{2-} at these Chongqing sites were almost double those at the Shizhan site, while the NO_3^- levels were more than double (70, 66, and 20 mmol m^{-2} y^{-1} at Haifu, and Jinyunshan, and Shizhan, respectively). Therefore, it seems that acidity derived from the strong acid anions in rainwater may not be sufficiently neutralized at these sites. In the sites in China, the deposition of strong acid anions and basic cations is significant, and therefore the balance between these ions is very important for evaluating the effects of acid deposition on ecosystems. The importance of cation deposition has already been suggested by previous studies (e.g., Dawei et al. 2001; Xu et al. 2001). Zhao et al. (2001) suggested that a high concentration of Ca^{2+} in the soil solution, which was derived from the high rates of Ca^{2+} deposition, controlled the toxicity of Al in the acidified soil of Chongqing. On the other hand, Larssen and Carmichael (2000) suggested that the effects of acid deposition on soil and vegetation would be manifested if the anthropogenic emission of calcium was reduced considerably faster than that of sulfur emission. Since sulfur emissions in China started declining after 2006, the emission trend of basic cations should also be monitored carefully.

The deposition levels of S and N are quite high in EANET countries. Matsubara et al. (2009) compared the wet deposition of S and N between the monitoring networks in Japan, Europe (determined by the European Monitoring and Evaluation Programme; EMEP), and the United States (determined by the National Atmospheric Deposition Program; NADP) to assess the impacts of acid deposition on river water pH. According to the comparison, the 50th percentiles for the wet deposition of nss-S in 2003 were 8.01, 2.57, and 2.85 kg S ha^{-1} y^{-1} in Japan, Europe, and the United States, respectively. Those of DIN were 7.86, 5.19, and 3.61 kg N ha^{-1} y^{-1}, respectively. Compared with deposition levels in Europe and the United States, deposition levels in Japan were significantly greater. The 50th percentiles for wet deposition across all EANET countries corresponded to 6.21 kg S ha^{-1} y^{-1} and 7.84 kg N ha^{-1} y^{-1} (calculated based on the data in Table 3.1). Although the assessment year was slightly different, the wet deposition of nss-S and N in the EANET countries was significantly greater than that in Europe and the United States. Moreover, the 90th percentiles of wet DIN deposition were 16.4, 9.20, 6.51, and 18.6 kg N ha^{-1} y^{-1} in Japan, Europe, the United States, and the EANET countries, respectively. Monitoring sites in the EANET countries and Japan significantly exceeded the threshold values of N deposition, whereby NO_3^- concentration had increased in stream water: 10 kg N ha^{-1} y^{-1} (Wright et al. 1995) and 8 kg N ha^{-1} y^{-1} (MacDonald et al. 2002) in Europe and 7 kg N ha^{-1} y^{-1} (Aber et al. 2003) in the United States. Measured values also significantly exceeded the critical N loads for epiphytic lichens: 2.4 kg N ha^{-1} y^{-1} (Giordani et al. 2014) in Europe and 3–9 kg N ha^{-1} y^{-1} (Geiser et al. 2010) in the United States. Therefore, the effects of N deposition along with acid deposition should be carefully monitored in the EANET countries, taking into account forest ecosystem sensitivities.

3.2.2   Dry and Total Deposition

Dry deposition of gaseous and particulate matter is also an important deposition process in terrestrial ecosystems. To evaluate acid deposition in these ecosystems, total deposition should be estimated as the sum of dry deposition and wet deposition. In the EANET countries, it was recommended that dry deposition flux should be evaluated by the inferential method (EANET 2010), in which the flux can be calculated by multiplying the air concentrations of the pollutants by their deposition velocities. However, detailed meteorological data is necessary to estimate dry deposition velocities, and sufficient meteorological data has not been compiled for the EANET sites, except for those in Japan. Therefore, as of 2015, regional assessment of dry deposition and total deposition has not yet been conducted in the EANET countries. According to the EANET second periodic report (EANET 2011), the mean dry deposition amount and mean wet deposition amount of S compounds in the Japanese EANET sites for the period from 2003 to 2007 were approximately 20 and 35 mmol m^{-2} y^{-1}, and those of N compounds were approximately 28 and 48 mmol m^{-2} y^{-1}, respectively. Compared with levels in Europe (determined by EMEP) and

the United States (determined by the Clean Air Status and Trends Network; CASTNET), the total deposition levels of S and N compounds were significantly greater in Japan (EANET 2011). In particular, total S deposition was three times greater than that in Europe and the United States, due to high levels of precipitation and nss-SO_4^{2-}. Although dry deposition has not been evaluated, total deposition levels at some of the EANET sites, such as the sites in Chongqing and Malaysia, may be significantly greater than those at Japanese sites; this can be explained by the extremely high wet deposition levels (see Table 3.1). Therefore, in the future, regional assessment of total deposition in Northeast Asia and Southeast Asia should be promoted to evaluate the effects of acid and N deposition on terrestrial ecosystems.

3.2.3 Observational Data in Forested Areas Obtained by the EANET: Relevant Studies

Acid deposition monitoring in the EANET has been conducted in open spaces, away from forested areas, to obtain representative data over a relatively large area. Therefore, to gain observational data in forested areas, such data should be gathered in the region. Atmospheric deposition and its effects on forest catchments have been studied by EANET scientists in different types of forests; namely, a temperate coniferous forest in Japan from 2002 (e.g. Kamisako 2008; Sase et al. 2008), a tropical dry evergreen forest in Thailand from 2005 (e.g., Yamashita et al. 2010; Sase et al. 2012), a tropical rainforest in Sabah, Malaysia, from 2008 (Yamashita et al. 2014), and a tropical rehabilitated rainforest in Sarawak, Malaysia, from 2012 (Sase et al. 2015). Although DIN may be subject to strong effects of canopy interactions, such as uptake or consumption in the forest canopy (Sase et al. 2008), the canopy interactions of S may be negligible. Therefore, the levels of S deposition collected by throughfall (TF) and stemflow (SF), which also include dry deposition amounts, can be considered as the total deposition in forested areas.

Atmospheric deposition amounts of S and DIN collected by TF and SF in the study forests are summarized in Table 3.2 (Sase et al. 2015). The deposition amounts

Table 3.2 Atmospheric deposition amounts of S and DIN in four different forests in Asia

Site	Kajikawa	Sakaerat	Danum Valley	Bintulu
Country and province	Niigata, Japan	Nakhon Ratchasima, Thailand	Sabah, Malaysia	Sarawak, Malaysia
Forest type	Japanese cedar	Dry evergreen forest	Tropical rainforest	Rehabilitated forest
Annual precipitation (mm)	2,281	1,488	2,700	3,500
S (kg S ha^{-1})	28.5	5.76	3.6	19
DIN (kg N ha^{-1})	16.6	7.9	6.2	11.8

After Sase et al. 2015
Note: Deposition amounts under the forest canopy, determined by throughfall (+ stemflow) method, are shown in the Table

of S and DIN were significant at the Kajikawa site in Japan, likely caused by the long-range transport of air pollutants from the Asian continent (Sase et al. 2012). Deposition levels at the Bintulu monitoring site, located in one of the most industrialized cities in Malaysia, were also high. Deposition levels measured in Sakaerat, Thailand, and the Danum Valley, Malaysia, were relatively low. However, the deposition pattern at the Sakaerat site showed a distinct seasonality as a result of well defined dry and wet seasons, with deposition peaks observed at the beginning of the wet season (Sase et al. 2012). At the Kajikawa site, the highest deposition levels were observed during periods of high snowfall in winter, due to strong seasonal westerly winds from the Sea of Japan (Kamisako 2008; Sase et al. 2008, 2012). Such seasonality of deposition and precipitation patterns may be closely related to phenology and biogeochemistry in ecosystems. Therefore, not only annual deposition levels but also their seasonal patterns should be carefully monitored to quantify the effects of acid deposition on forest ecosystems.

3.3 Soil Acidification in Asian Countries

The current level of acid deposition in EANET countries may not affect plant physiology directly. However, soil acidification is still a potential risk for plant growth in Asian countries. According to a model simulation by Hicks et al. (2008), acid-sensitive soil types were found in some areas of Northeast and Southeast Asia, where base saturation of the soil would decrease to less than 20% with acidification within 50 years, although the projection varied depending on future deposition scenarios. This finding strongly indicates that long-term observational data of acid deposition and soil chemical properties should be accumulated.

Soil has buffer systems; therefore, the addition of a strong acid does not directly result in a decrease in pH (Ulrich 1991). The amount of strong acid required to reduce the pH of a system to a reference pH value is termed the "acid neutralizing capacity (ANC)"; thus, soil acidification should be defined as a decrease in the ANC of the soil (Van Breemen et al. 1983). Decreases in the ANC due to inputs of strong acids derived from external sources are not necessarily reflected in changes to indicators of soil acidification, such as pH and exchangeable H^+ and Al^{3+}. Therefore, it takes a long time to detect soil acidification phenomena from field observations. In Europe (e.g., Hallbäcken and Tamm 1986; Nilssen1986; Schulze 1989) and the United States (Drohan and Sharpe 1997), soil acidification phenomena, such as decreases in pH and in Ca/Al and Mg/Al ratios in soil solutions, were observed by re-sampling forest soils after 20–50 years. Based on the data summarized in Central Europe and Scandinavia by Nilssen (1986) and Ulrich (1991) and data from the United States (Drohan and Sharpe 1997), the reduced pH values and the initial soil pH are plotted in Fig. 3.2a (after Sase 2015). Greater pH decline can be observed in the soils with higher initial pH. This may be because buffering systems function differently in soils with different pH values, as suggested by Ulrich (1991).

As described above, re-sampling of pre-established plots after 20−50 years helped to quantify soil acidification, as proven in the 1980s in Europe and the United
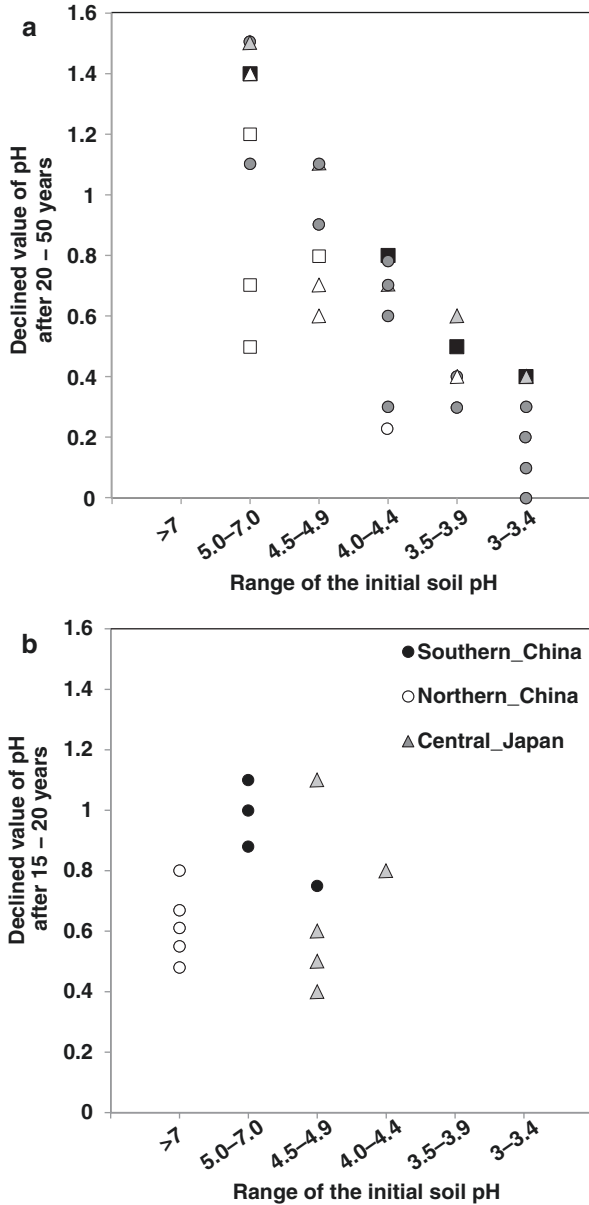
Fig. 3.2 Initial soil pH and declined pH values in (**a**) Central Europe, Scandinavia, and the United States for 20–50 years and (**b**) China and Japan for 15–20 years (After Nilssen 1986; Ulrich 1991; Drohan and Sharpe 1997; Dai et al. 1998; Liu et al. 2010; Nakahara et al. 2010; Yang et al. 2012; Sase 2015). *Closed circles* humus layers, *open circles* A-horizons, *open triangles* B-horizons, *closed triangles* silt, *open squares* C-horizons, *closed squares* sand

States. Unfortunately, in East Asia, this approach has not been conducted to a sufficient extent. Soil acidification was reported by field observation only in those areas of China and Japan that received high acid deposition. In China, this occurred in the mountainous areas of Hunan Province and the Guangxi Zhuang Autonomous Region from 1959/1960 to 1994 (Dai et al. 1998); in the Dinghushan Biosphere Reserve, Guangdong, from the 1980s to 2005 (Liu et al. 2010); and in northern China's grasslands from the 1980s to 2000s (Yang et al. 2012). In central Japan, this occurred in the Lake Ijira catchment, Gifu Prefecture, from 1990 to 2004 (Nakahara et al. 2010). The observational data from China and Japan are plotted in Fig. 3.2b (after Sase 2015). Similar to the findings noted above, in China and Japan, when initial soil pH was lower than 7.0, greater pH decline seemed to be observed in the soils with higher initial pH, although the amount of supporting data was limited. Liu et al. (2010) also showed that forest soils with originally higher pH values were acidified more rapidly than those with originally lower pH values. In soils with high pH (>7) in northern China, soil carbonates functioned as the main buffering systems (Yang et al. 2012), and therefore pH declines in these soils may be relatively insignificant. Dai et al. (1998) showed that soil acidification was not apparent in areas covered by red and yellow soils, such as acrisols, alisols, and ferralsols. These soils are highly weathered acidic soils and are rich in aluminum, and are theoretically within the aluminum buffer range. Therefore, pH values in these soils may not readily decline, even with high acid deposition.

3.4 Summary

Emission levels of S and N remain high in Asia. Accordingly, deposition levels are significantly higher in Northeast and Southeast Asia than in Europe and the United States. Wet deposition monitoring data have been accumulated owing to the efforts of the EANET, the regional network in East Asia, although total deposition and dry deposition have not yet been sufficiently evaluated. Observational data on acid deposition in forested areas are also lacking. Atmospheric deposition, including wet deposition and dry deposition, should be monitored in Asian forested areas. Soil acidification caused by acid deposition poses a potential risk for plant growth. Although the number of observations is limited, soil acidification phenomena have been observed in China and Japan. These phenomena corresponded to the soil-buffering mechanism, a finding that was similar to observations in Europe and the United States. Long-term observational data on soil chemical properties should be gathered to determine the potential risk for plant growth with greater precision.

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