Results from long-term fertilizer experiments in China: The risk of groundwater pollution by nitrate

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Lessons from the long-term fertilization experiments in China and the risk of groundwater pollution by nitrate

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Abstract: NO_3-N distribution and accumulation down to 200 or 300 cm in soil profiles of different long-term fertilization systems were studied in 2002 in the China Long-term Experiments Network (CLTEN) which includes eight experimental sites where the experiments were started in 1990 or 1991. In this paper, we report on the results from five sites (Beijing, Henan, Hunan, Jilin and Xinjiang) representing a wide range of soils, climates and cropping systems, and some with irrigation. At each site, crops (wheat and/or maize) have been grown with no fertilizer or manure inputs (as control), and with various combinations of N, P or K fertilizers or with NPK *plus* different levels of manure (M) or crop straw (S) input. Agricultural fields where N input was from fertilizers, generally had higher amounts of accumulated NO_3-N in soil profiles than control or the long-term fallowed soils, which indicated that the use of fertilizer N in agricultural systems increased the risk of pollution of the environment. Long-term application of fertilizer N without P (N, NK) resulted in low crop yields and low N uptake by crops, leading to lower cumulative apparent N recovery (ANR) and higher NO_3-N concentration and accumulation in soil profiles. This increased the risk of groundwater contamination by nitrate. When fertilizer N was applied along with P (NP) or PK (NPK) the crop yields, N uptake by crops and ANR were increased markedly and the NO_3-N accumulation in soil profile was much lower than N and NK treatments. Adding manure or crop straw based on equal total N (NPK+M or NPK+S) resulted in similar concentration and accumulation of NO_3-N in soil profiles to NPK treatment in normal conditions. Increasing the N input levels whether through fertilizer or manure (NPK+1.5M or 1.5(NPK+M)) further increased the NO_3-N accumulation in soil profiles. The data shows that potentially 24 to 82 % of applied mineral fertiliser N was lost, mostly through ammonia volatilisation. It was estimated that a quarter of the N is lost through leaching beyond the root zone.

Key words: ***long-term fertilizer experiments, fertilizer use efficiency, NO_3-N distribution and accumulation, nitrate contamination of groundwater***

1. Introduction

Under aerobic conditions in soil, nitrate (NO_3^-) is the final oxidized form of inorganic nitrogen and the main chemical form in which nitrogen is taken up by plants. Nitrate ions are soluble in water and can easily be leached out of the soil to natural waters. Leaching is the main pathway of nitrate loss and this increases the risk of groundwater contamination by nitrate [1].

In order to get high yields of agricultural crops, high rates of chemical N fertilizers are usually applied. In 2000/2001, 28 % of the N and 25 % of the NPK fertilizers in the world were consumed in China [2].

The annual chemical fertilizer input per hectare of arable land in China was 2.8 times the world average level in 2000/2001. In China, N fertilizer recovery is around 35% which means that approximately 50% or more of N fertilizers (11.34 Mt) applied are lost through various pathways. Inappropriate use of fertilizers results in severe non-point sources of pollution from agriculture and applies great pressure to the environment in China. Imbalanced application of fertilizers with high rates of N relative to P and K, which are used in low quantities, makes the situation much worse. According to traditional viewpoints, greater use of organic manure (OM) results in better crop yields and higher soil fertility. But recent research findings also indicated that unreasonable use of OM can also increase the risk of fertilizer pollution [3].

Although a lot of work has been conducted on nitrate movement in the soil profile and losses through leaching [4,5], few studies have been conducted on the effect of long-term fertilization systems on nitrate distribution and accumulation in the soil profile [6,7,8].

This paper reports on the nitrate distribution and accumulation down the soil profiles (200-300 cm) on 5 of the 8 sites of the China Long-term Experimental Network (CLTEN) which covers the major soil types, climate conditions and cropping systems in China.

2. Materials and methods

2.1 Background information

The present study was carried out in the China Long-term Experiments Network (CLTEN). The CLTEN includes 8 long-term experimental sites in which experiments were started in 1989-91. The five sites reported in this paper (Beijing, Henan, Hunan, Jilin and Xinjiang) represent a wide range of soils, climates and cropping systems with three sites being irrigated. The background information and initial soil physical and chemical characteristics, as well as the cropping systems of the five sites are given in Table 1 and 2.

2.2 Fertilizer treatments

At each site, control crops were grown with no fertilizer or manure inputs. Fertilizer treatments consist of various combinations of N, P or K fertilizers (N, NP, NK, PK, NPK) or NPK plus different levels of manure (M) or straw (S). The amounts of N, P and K applied varied from 90 to 242 kg ha^{-1}, 16 to 60 kg ha^{-1} and 30 to 70 kg ha^{-1} for each crop, respectively. Manure was applied once a year (to the first crop where more than one crop was grown) except at Hunan, where manure was applied to both wheat and maize (30% used in wheat and 70% used in maize). At all sites the rate of fertilizer N applied to the crop with manure was reduced by an amount equivalent or similar to the amount of total *N* in the manure, except at the Beijing site where the added N was not reduced. The detailed information of fertilizer N applied in every treatment at each site is shown in Table 3.

2.3 Soil sampling and NO_3-N measurement method

Composite soil samples were collected annually from the top 20 cm of soil, dried, stored and analysed for total N using the semi-micro Kjeldahl method [9].

Soil samples were taken in 2002 (after 12 years at Beijing, Henan and Hunan site; 13 years at Jilin and Xinjiang site) at the same time as or just after the crop harvest (at double cropping sites, i.e. in

Beijing, Henan and Hunan site, soil samples were taken at the same time or just after the second crop harvest) with a post hole auger at 20 cm intervals to a depth of 200 cm (at Beijing, Henan and Jilin) or 300 cm (at Hunan and Xinjiang). Soil samples were immediately brought to the laboratory for the measurement of NO_3-N. Nitrate-N was extracted from the soils using 2 M KCl (10g of fresh soil:50 ml 2 M KCl). The NO_3-N content was measured at a wavelength of 210nm on an UV-spectrophotometer. The NO_3-N concentration of the soil sample was calculated according to the reference curve of standard NO_3-N solutions made and measured at the same time.

The cumulative Apparent N Recovery (ANR) from 1990-91 to 2002 was calculated as the difference between the Cumulative N Removed (CNR) by the crop in each treatment less the control treatment, divided by the total Cumulative N Added (CNA) for the treatment, expressed as a percentage. This parameter provided an estimate of the efficiency of the applied fertiliser.

In this project Total N was only measured (using the semi micro Kjeldahl method) for the surface soil (0-20 cm) but not for the profile. Total N did not change with time for the mineral fertiliser treatments, but they increased steadily with time for the manure treatments(data not presented). It is reasonable to assume that similarly Total N should not change with depth for the fertiliser treatments; and therefore an approximate root zone N balance calculations can be done for the fertiliser treatments. The Total N Available (TNA) in the root zone (0-200 cm) from 1991 to 2002 was estimated as the Cumulative sum of N Added as fertilizer (CAN) and mineralized N (estimated to be similar to Cumulative N Removal (CNR) by the control crop). The total N lost from the system is estimated as the difference between Total N Available (TNA) in the root zone less N uptake by the crop (CNR) and the mineral N remaining in the root zone (NR). This can be expressed as a percentage of added N (CNA); this assumes that all N released by mineralisation of soil organic matter is consumed by the crop similar to that in the control treatment.

The Plant Utilisation of Water (PUW) was calculated using the total water applied as :

Yield /(mm rainfall + mm irrigation) (kg/ha mm)

PUW is an estimate of the water use efficiency of the system, it is not the true crop water use efficiency which should be calculated based on the actual amount of water used by the crop.

3. Results

3.1 Crop yields and productivity per unit water input (PUW)

At Beijing, Henan and Hunan, double cropping of wheat and maize were practiced, and at Jilin and Xinjiang one crop is grown each year (continuous maize at Jilin, and a wheat and maize rotation at Xinjiang). Table 4 shows the mean annual crop grain yields from establishment to year 2002. Unfortunately, when these experiments were established no field plot replications were included, except for Hunan. We estimated that differences of at least 1 t/ha is required before any effect can be considered significant.

Table 4 shows that yields from the control, N, NK or PK treatments were usually poor. Yields on these plots would have been limited by the lack of both available N and P. Where both N and P were applied simultaneously, yields were increased by more than 100% (NP, NPK and NPK + manure or S).

Apart from the Beijing site, total rates of N application for the NPK and the NPK+M treatments were equivalent. At all sites, there was virtually no difference in average grain yields between these treatments. Ant difference equal to or less than 1 t/ha is not considered significant because of the lack of replications. Increasing N application from (NPK+M) to (NPK + 1.5 M) or 1.5(NPK+M) did not increased crop yields except at the Hunan site. Adding N as straw in addition to NPK did not increase yields relative to the NPK treatment.

It is interesting to note that despite the high rates of nutrient application and higher rainfall in Hunan, yields were only half of that in Henan. It is possible that some micro-nutrient deficiency is limiting yield at the Hunan site.

The grain yields produced per unit water (rain +irrigation) input (PUW in kg mm^{-1} water) are shown in Table 4. The PUW is an estimate of the water use efficiency of the cropping system. It is not a measure of the actual water use efficiency of the crop (grain yields produced per unit of water uptake by the crop). As PUW respond to fertiliser treatments in the same way as grain yield, these results indicate that optimising water use efficiency will be closely linked to optimising crop nutrient supply.

3.2 Plant N uptake and fertilizer N use efficiency

Cumulative fertiliser N Added (CNA), Total N added (TNA), cumulative N removed (CNR) by the crop, the apparent N recovery (ANR) by the crop and total N lost from the root zone from establishment to 2002 for three sites with wheat-maize rotations are reported in Table 5. ANR is directly affected by the fertilization systems. When N was applied with P and/or K, ANR was higher than when only N was applied. An average recovery of 14%, 4.7% and 10% were obtained with an annual application of 300, 353 and 300 kg N ha^{-1} alone at Beijing, Henan and Hunan, respectively. The application of P with N increased apparent N recovery to 42%, 31 % and 27 % respectively, for the same sites. When N was applied with K, the recoveries for these sites were 22%, 18% and 14 % respectively. When N, P and K were applied simultaneously, ANR significantly increased to 49%, 42% and 34% respectively. This means that P and K are also limiting crop performance at these three experimental sites besides N. Table 2 shows that P is deficient at all sites while K is deficient in the Beijing and Henan sites. These deficiencies would be consistent with the long history of cultivation on these soils.

Where mineral N application is partly replaced by an organic N such as manure (Henan and Hunan) or straw (Henan), crop removal of N tends to increase, indicating the effectiveness of organic matter as a slow release N fertiliser, and this is reflected in a higher ANR. Where N application is significantly increased with the use of manure ((NPK +M) or (NPK +1.5 M) at Beijing: or 1.5(NPK+M) at Henan and Hunan) crop uptake of N is increased, however this increase is generally smaller than the increase in N application. Therefore, this is reflected in a reduction in ANR.

These results show that the efficiency of N uptake from added mineral or organic fertilisers under the conditions of these trials is at best around 50 % with more than half of applied N either lost through leaching, denitrification or volatilization, or retained within the soil as mineral or organic N. The results also suggest that the application of N fertilizer alone without P and K will decrease the fertilizer use efficiency and increase the potential losses of nitrate. The addition of organic N sources along with mineral N fertilizer can also improve N use efficiency.

3.3 Nitrate balance and losses from the root zone.

The risk of groundwater pollution is better shown through changes in the NO_3-N profile in the soil at harvest and the associated N balance of the root zone (0-200 cm depth). Figures 1 and 2 show the NO_3-N distribution in soil profiles under different long-term fertilization systems, including an unfertilized long term fallow plot with no cropping. Figure 3 shows the accumulated amounts of NO_3-N in the soil profile while Figure 1 and Figure 3 show that at harvest, the application of fertilizer N resulted in higher soil NO_3-N contents than the control and the long-term fallowed soils at almost all the sampling depths at all sites except on the dry Xinjiang site. In the dry Xinjiang site, the long-term fallow tends to release and accumulate a large amount of N through mineralization which is not lost through denitrification or leaching.

The application of N alone or in combination with K resulted in higher NO_3-N contents in the soil profile at most of the experimental sites except at the Xinjiang site. The application of N with P had improved the apparent N recovery compared to N treatments (Table 5) and resulted in generally lower NO_3-N contents in the soil profile compared to N treatments (Figure 3). The application of P and K with N further improved the apparent N recovery compared to the NP treatment (Table 5) and resulted in even lower NO_3-N content in soil profile compared to NP treatments (Figure 3), except in the Xinjiang and Hunan sites. On the latter sites, NO_3-N contents in the soil profile remains high under NPK associated with smaller yield responses to these fertilisers and possibly less leaching from the root zone.

Manure application was done based on the equivalent total N input principle except at the Beijing site, i.e. when manure was applied the amount of fertilizer N was reduced by an amount equivalent to the total N added in the manure. In most cases, replacing fertilizer N with manure N or straw N did not significantly affect the NO_3-N content in the soil except at the Xinjiang site where equivalent amounts of N supplied as NPK with manure or straw, significantly reduced the NO_3-N accumulation in the soil profile.

The risk of nitrate leaching is illustrated by the root zone N balance for the fertiliser treatments as shown in Table 5 for the Beijing, Henan and Hunan sites where the cropping systems consist of winter wheat followed by summer maize. The root zone is assumed to be 200 cm deep. The calculations shown in table 5indicate a large pool of N that cannot be readily accounted for. While some of this may be retained in the soil as organic N, particularly where N is applied as manure, there still appears to be a significant potential for N losses from these systems when mineral N fertilizer was applied on its own. The most probable loss pathways for this N would be volatilisation of ammonia, denitrification and deep leaching. In a 4 year experiments by Yu et all [10] on the irrigated winter wheat-summer maize rotation on the North China plains, they measured N losses of about 22 % and 40% under wheat and maize respectively, for similar rates of mineral N application. Using N^{15} tracers, they found that ammonium volatilisation is the major mechanism of loss under this system (19 % under wheat and 25 % under maize) with a small proportion due to leaching (2.7 % under wheat and 12 % under maize). Averaging wheat and maize, a quarter of N losses is due to leaching while denitrification losses are very small (0.1 to 3.3 %). A recent review by Dalal et al [11] also showed that under Australian dryland arable grain farming, reports of losses of N from denitrification ranged from <0.01 % to 9.9% of fertiliser N applied. Table 5 shows that over the 11 years of monitoring, the average fertiliser N lost ranged from 24 to 56 % where N, P and K are applied, and up to 82 % where N was applied without P and K. Using the results from Yu et all, we can estimate that leaching losses of N beyond the root zone can be substantial with the largest quantities being from N fertilizer alone, ranging from 6 to 20 %. The significance of leaching as a mechanism of N loss is likely to be more pronounced in the higher rainfall areas as shown in Fig 3 where the Nitrate N concentrations are presented for 2 and 3 m depth on the Hunan sites. The amount of Nitrates between 2 and 3 m are substantial and may well reached beyond 3 m depth.

It is interesting to note that despite the high rates of nutrient application and higher rainfall in Hunan, yields were only half of that in Henan. Perhaps some micro-nutrient deficiency maybe limiting yield on the Hunan site.

4. Conclusions

In conclusion, we have shown that P and K as well as N, are limiting crop yields on all sites. This is most likely associated with the long history of cultivation on most Chinese soils which has depleted the soils' P and K status (see Table 2). Other micro-nutrients may be limiting yields on some sites and should be investigated in future studies. We have also found that in general, whether N is applied as mineral fertilizer or in the form of composted animal manure or crop straw, its effects on crop yield are similar. We have also demonstrated significant potential for N leaching below the root zone when N is applied at high rates, regardless of the source of applied N.

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Table
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Table 1. Background information on the five sites from the China Long-term Experiments Network used in the N leaching study

Site	Beijing	Henan	Hunan	Jilin	Xinjiang
Location	Changping	Zhengzhou	Qiyang	Gongzhuling	Wulumuqi
Longitude	116^{o}12’ 08’’ E	113^{o}39’ 25’’ E	111^{o}52’ 32’’ E	124^{o}48’ 34’’ E	87^{o}25’ 58’’ E
Latitude	40^{o}12’ 34’’ N	34^{o}47’ 02’’ N	26^{o}45’ 12’’ N	43^{o}30’ 23’’ N	43^{o}58’ 23’’ N
Altitude, m	44	91	120	220	553
Climate zone	Cool temperate, semi-humid	Cool temperate, semi-humid	Sub-tropical, humid	Cool temperate, highly continental	Cool temperate, semi-desert
Meanannual temperature, ^{o}C	11.8	14.2	18.3	6.6	7.4
Annualrainfall, mm	577	644	1276	559	247
Cropping,per year	Wheat-maize	Wheat-maize	Wheat-maize	Maize	Wheat or maize
Plot size, m^{2}	200	400	196(x 2 reps)	400	468
Meanannual irrigation, mm	300	225	None	None	450

Table 2. Initial physical and chemical characteristics of the surface (0-20 cm) soil from the China Long-term Experiments Network used in the N leaching study

Site	Beijing	Henan	Hunan	Jilin	Xinjiang
Soil name	Drab Fluvo-aquic	Fluvo-aquic	Red	Black	Grey Desert
Texture (USDA)	Silt loam	Silt loam	Clay	Clay loam	Silt loam
Sand, %	20.3	26.5	3.7	38.3	18.5
Silt, %	65.0	60.7	34.9	29.9	53.2
Clay, %	14.7	12.8	61.4	31.8	28.3
Bulk density, g cm^{-3}	1.50	1.55	1.3	1.19	1.25
pH[z]	8.2	8.3	5.7	7.6	8.1
Organic C[y], g kg^{-1}	7.1	6.6	8.1	13.5	8.1
Total N[x], g kg^{-1}	0.79	0.65	1.07	1.40	0.87
Avail P, g kg^{-1}	4.2	6.6	10.8	11.8	4.5
Avail K, g kg^{-1}	65.0	70.3	122.0	158.3	288.0

[z] The soil pH was measured in 1.2.5 soil:water ratio.

[y] Organic C was measured by the Walkley wet combustion method [12].

[x] Total N by the Kjeldahl method [3], available P with 0.5 *M* $NaHCO_3$ [13], exchangeable K with *N* ammonium acetate [14].

Table 3. Total N applied as inorganic fertilizer, manure (M) or straw (S) in an N leaching study

	Beijing[z]		Henan[y]		Hunan		Jilin	Xinjiang[x]
	Wheat	Maize	Wheat	Maize	Wheat	Maize	Maize	Rotation
CK	0	0	0	0	0	0	0	0
N	150	150	165	188	90	210	165	242
NP	150	150	165	188	90	210	165	242
NK	150	150	165	188	90	210	165	242
PK	0	0	0	0	0	0	0	0
NPK	150	150	165	188	90	210	165	242
NPK+M	150+146	150	50+115	188	27+63	63+147	50+115	85+240
NPK+1.5M	150+218	150	na	na	na	na	na	Na
1.5(NPK+M)	na	na	74+173	281	41+94	95+221	74+173	152+360
NPK+S	150+18	150	123+42	188	90+9	210+9	112+53	217+29

[z] At the Beijing Site in 1997 no fertilizer was applied.

[y] At the Henan site: the first year (1991) of the N fertilizer is 165 kg/ha for maize, after that it is 188 kg/ha. In 2003, in the NPK+S treatment the N from chemical fertilizer is 87 kg/ha and N from crop straw (S) is 78kg/ha.

[x] At the Xinjiang site, the rates of fertilizers are the average of rotation crops.

na = not application.

Table 4. Crop grain yields (annual average from 1990-91 to 2002: t/ha), Plant Utilisation of Water (PUW) (kg/mm) of different fertilization systems

Treatments	Beijing		Henan		Hunan		Jilin		Xinjiang	
	Yield	PUW	Yield	PUW	Yield	PUW	Yield	PUW	Yield	PUW
CK	2.6	3.0	5.2	6.4	0.8	0.6	3.7	6.6	2.2	2.9
N	3.2	3.7	6.3	7.8	1.5	1.2	7.7	13.8	3.7	4.8
NP	6.8	7.8	11.9	14.7	4.0	3.1	8.6	15.4	4.8	6.2
NK	3.8	4.3	7.5	9.3	2.0	1.6	8.4	15.0	3.5	4.6
PK	3.7	4.2	4.7	5.8	1.3	1.0	4.1	7.3	3.2	4.2
NPK	7.9	9.0	12.1	14.9	5.5	4.3	8.9	15.9	4.7	6.1
NPK+M	8.9	10.2	12.0	14.8	6.7	5.2	8.6	15.4	5.3	6.9
NPK+1.5M	8.2	9.4	na	na	na	na	na	na	na	na
1.5(NPK+M)	na	na	12.8	15.8	7.9	6.2	9.5	17.0	5.5	7.2
NPK+S	7.9	9.0	12.9	15.9	6.3	4.9	8.8	15.7	4.6	6.0

Table
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Table 5. Total (N) input and output balance and the apparent N recovery (ANR,%) of different fertilization systems for the period from 1990-91 to 2002. Note: ANA-annual N application (kg ha^{-1}), CNA-cumulative fertilizer N added (kg ha^{-1}), CNR-cumulative N removed by crop (kg ha^{-1}), ANR- apparent N recovery (%)

	Beijing						
Treatments	**ANA** Annual N Application (kg ha^{-1})	**CNA** Cunulative Fert N Added (kg ha^{-1})	**TNA** Total N available (kg/ha)	**CNR** Cumulative crop N Removal (kg ha^{-1})	**ANR** Apparent fert N Recovery (%)	**2002 NR** Nitrate N in root zone (0-200cm) (kg ha^{-1})	**% N Loss** % Fertiliser N lost from root zone (TNA – CNR – NR)/CNA
CK	0	0	741	741	--	21	-
N	300	3300	4041	1205	14	364	52.5
NP	300	3300	4041	2116	42	157	31.1
NK	300	3300	4041	1455	22	215	49.4
PK	0	741	741	838	--	37	-
NPK	300	3300	4041	2352	49	144	24.4
NPK+M	446	4906	5647	2683	40	229	-
NPK+1.5M	518	5698	6439	2648	33	320	-
1.5(NPK+M)	na	na	na	na	na	na	-
NPK+S	318	3498	4239	2425	48	187	-
	Henan						
Treatments	**ANA**	**CNA**	**TNA**	**CNR**	**ANR**	**2002 NR**	**% N loss**
CK	0	0	1091	1091	--	27	-
N	353	4213	5304	1296	4.7	538	82.4
NP	353	4213	5304	2394	31	88	67
NK	353	4213	5304	1838	18	437	71.9
PK	0	1091	1091	1698	--	14	-
NPK	353	4213	5304	2871	42	47	56.5
NPK+M	353	4213	5304	2778	40	65	-
NPK+1.5M	na	na	na	na	na	-	-
1.5(NPK+M)	528	6303	7394	3269	35	263	-
NPK+S	353	4213	5304	3385	54	41	-
	Hunan						
Treatments	**ANA**	**CNA**	**TNA**	**CNR**	**ANR**	**2002 NR**	**% N loss**
CK	0	0	253	253	--	263	-
N	300	3600	3853	597	10	1256	55.6
NP	300	3600	3853	1222	27	942	46.9
NK	300	3600	3853	754	14	795	64
PK	0	0	253	446	--	490	-
NPK	300	3600	3853	1478	34	504	52
NPK+M	300	3600	3853	1944	47	492	-
NPK+1.5M	na	na	Na	na!	na	na	-
1.5(NPK+M)	450	5400	5653	2592	43	265	-
NPK+S	318	3816	4069	1700	38	356	-

Figure
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Fig. 1. Effect of N, P and K application on NO_3-N distribution in the soil profile

Fig. 2. NO_3-N distribution in soil profile as affected by application of manure and crop straw

Fig. 3. NO_3-N accumulation in the soil profile of different fertilization systems

Figure
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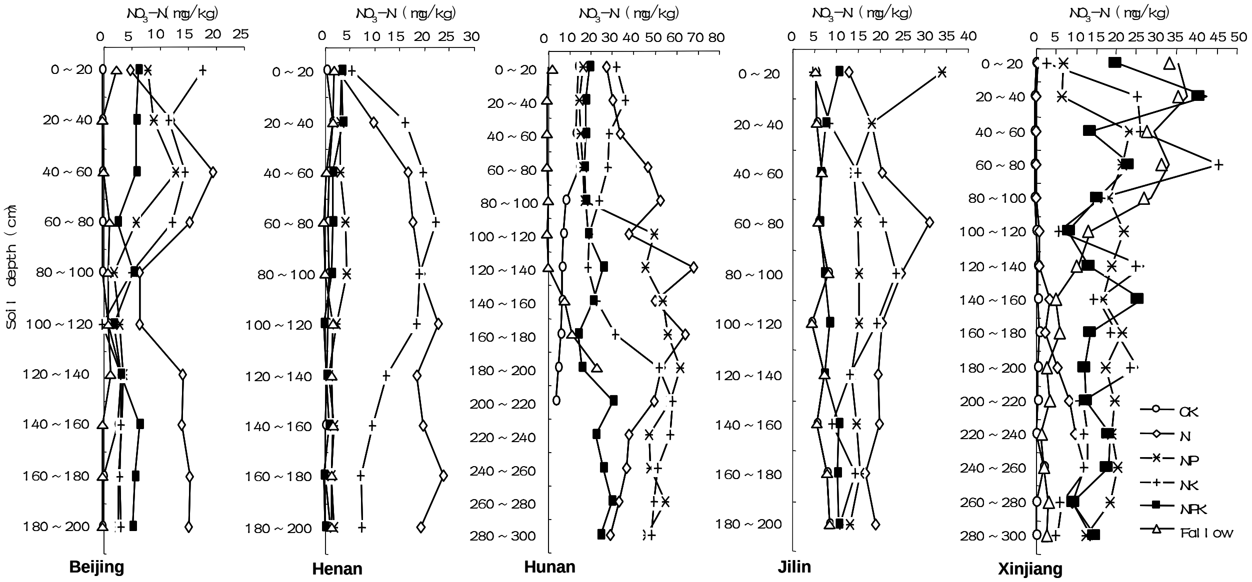

Figure

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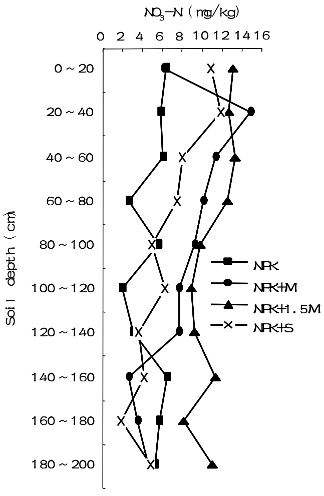

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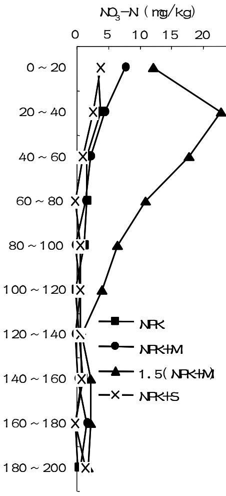

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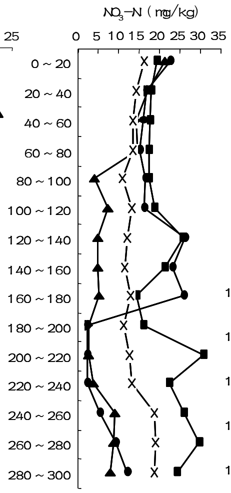

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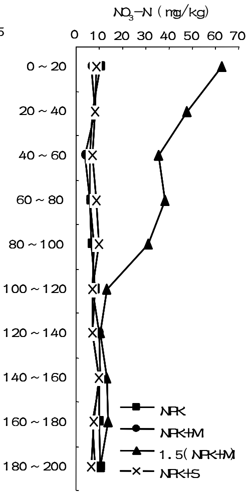

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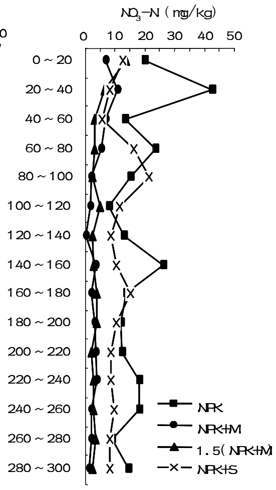

Xinjiang

Figure

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