Supplementary information for

"Historical Nitrogen Fertilizer Use in Agricultural Ecosystem of the Continental U.S. during 1850-2015: Application, Timing, Fertilizer Type"

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1 Historical state-level farm-used N consumption

We harmonized the state-level farm-used N fertilizer consumption from Mehring et al. (1957) for period of 1930-1953, USDA (1971) for period of 1954-1969, USDA (1977) for the year 1976 and 1977, and Brakebill and Gronberg (2012) for period of 1987-2012 (Table S1).

Supplementary table S1. Data sources for state-level farm-used N consumption

Data sources	Period
Mehring et al. (1957)	1930-1953
USDA (1971)	1954-1969
USDA (1977)	1976-1977
Brakebill and Gronberg (2012)	1987-2012

2 Historical national average crop-specific N use rate

National average crop-specific N rates were derived from Mehring et al. (1957), USDA (1957), Ibach et al. (1964), Ibach and Adams (1967), USDA-ERS (2013), and USDA-NASS (2017). Crop types fertlized were reported differently between these data sources. In Mehring et al. (1957), N used in seven crop types were recorded, including corn, soybeans, wheat (sum of N used in spring wheat, winter wheat, and durum wheat), cotton, rice, barley, and cropland pasture. While in

comparison, USDA (1957), Ibach and Adams (1967), and USDA-ERS (2013) provided N use in all nine major crops during 1954 to 1964, including three wheat types and cropland pasture. For the period of 1964 to 2015, USDA reported all nine major crops without cropland pasture. In addtion, crop-specific N use documented in Mehring et al. (1957) is the sum of N, P, K, and trace nutrient fertilizers (Table S2).

Supplementary table S2. Data sources for national average crop-specific N use rate

Data sources	Period	Crop types	Data form
Mehring et al. (1957)	1927, 1938, 1942, 1946, 1950	7[a]	Total fertilizer consumption
USDA (1957)	1954	10[b]	N use rate
Ibach et al., (1964)	1959	10[b]	N use rate
Ibach and Adams. (1967)	1964	10[b]	N use rate
USDA-ERS (2013) and -NASS (2017)	1965-2015	9[c]	N use rate

The number of crop types recorded in different sources varied. [a], crop types included corn, soybeans, wheat (spring wheat, winter wheat, and durum wheat in total), cotton, rice, barley, and cropland pasture. [b], crops included all nine major crops and cropland pasture. [c], crops only included nine major crops but not cropland pasture. Total fertilizer of each crop type reported in Mehring et al. (1957) contained all N, P, K, and trace fertilizers.

3 Application timing

N fertilizer application timing information was derieved from the survey conducted by USDA-ERS (2016). The survey started from 1996, and collected the data periodically for each crop in each state (Table S3). The survey years of different crop types may vary. For example, corn plantings were surveyed in 1996-2001, 2005, and 2010, while soybean plantings were surveyed in 1996-2000, 2002, 2006, and 2012. Moreover, the survey year of a specific crop type in different states may also vary. We adopted the latest surveyed years for each of the all nine crops. For the lastest surveyed year, USDA-ERS (2016) only reported the average application timings at all survey states for sorghum (year 2011) and rice (year 2013). We therefore used the prior surveyed years to generate state-level application timings for sorghum and rice (Table S3).

Supplementary table S3. The survey years of application timing we adopted for nine crops

Crops	Survey years
Corn	1996-2001, 2005, **2010**
Soybeans	1996-2000, 2002, 2006, **2012**
Winter Wheat	1996-1998, 2000, 2004, **2009**
Spring Wheat	1996-1998, 2000, 2004, **2009**
Cotton	1996-2000, 2003, **2007**
Sorghum	**2003**, 2011 (Average timing only)
Rice	**2006**, 2013 (Average timing only)
Barley	**2011**
Durum Wheat	1996-1998, 2000, 2004, **2009**

The years with bold font were the survey years we mostly adopted for each crop in this study. But the specific year of a given state may vary due to the different survey schedule in every states. The reported application timings data for sorghum and rice in their latest surveyed years were average over all surveyed states.

4 Reconstruct historical national consumption of 11 N fertilizer types

Historical national consumption of 11 N fertilizers from 1900 to 2011 were collected from Mehring et al. (1957), USDA (1966), USDA-ERS (2013), and FAO (2017) (Table S4). We assume the fraction of NH_4^+-N and NO_3^--N remained unchanged before 1900 because the commerical N fertilizer was mainly extracted from organic products and mineral deposits (Sheridan, 1979). Diammonium Phosphate (DAP) and Monoammonium Phosphate (MAP) were not reported during 1900-1960 due to their consumption were relatively small and were incorporated in Ammonium Phosphates (APs). After 1960, however, the consumption of these two fertilizers increased to very high amount and were reported separately.

Supplementary table S4. Data sources used to reconstruct historical consumption record of 11 N fertilizers across nation

Fertilizers	AnA	AqA	AN	AS	NS	SN	Urea	CN	DAP	MAP	APs
1900-1953									\	\	
1954-1959								\	\	\	
1960-2003											
2004-2011											

Red: Mehring et al. (1957), green: USDA (1966), blue: USDA-ERS (2013), yellow: FAO (2017). AnA: Anhydrous Ammonium. AqA: Aqua Ammonium. AN: Ammonium Nitrate. AS: Ammonium Sulfate. NS: Nitrogen solution. SN: Sodium Nitrate. CN: Calcium Nitrate. DAP: Diammonium Phosphate. MAP: Monoammonium Phosphate. APs is the integration of Ammonium Phosphates, before 1960, the consumption of DP and MP were relatively small, so these two fertilizers were incorporated in APs, after 1960, however, the consumption of these two fertilizers increased to very high amount and were reported separately.

5 Data sources for state-level N consumption of 11 single N fertilizer types

Mid-decade (every sixth year of one decade) state-level N consumption of 11 single N fertilizers were derieved from mulitiple data sources, including USDA (1966) for 1946 and 1956, USDA (1971) for the year 1966, USDA (1977) for 1976, and AAPFCO (1986, 1996, 2006, and 2012) for 1986, 1996, 2006, and 2012. We gap-filled Calsium Nitrate (CN) to 1946 based on the data of the year 1986, and also gap-filled DAP and MAP to 1960 based on the data in 1986 and set to zero for years before 1960.

Supplementary table S5. Data sources used to reconstruct historical state-level consumption of 11 N fertilizers

Fertilizers	AnA	AqA	AN	AS	NS	SN	Urea	CN	DP	MP	APs
1946								\	\	\	
1956								\	\	\	
1966								\	\	\	
1976								\	\	\	
1986											

Green: USDA. (1966). Orange: USDA (1971). Purple: USDA (1977). Blue: AAPFCO (1986, 1996, 2006, and 2012). AnA: Anhydrous Ammonium. AqA: Aqua Ammonium. AN: Ammonium Nitrate. AS: Ammonium Sulfate. NS: Nitrogen solution. SN: Sodium Nitrate. CN: Calcium Nitrate. DP: Diammonium Phosphate. MP: Monoammonium Phosphate. Aps: The integration of Ammonium Phosphates

6 Estimating the ratio of application timings for 11 single N fertilizer types

Due to the physical and chemical differences between N fertilizer types, applying N fertilizers at specific timings can enhance crop production and reduce N loss. To assign consumption of each N fertilizer type into four application timings, namely, Fall (previous year), Spring (before planting), At planting, and after planting, we took the Agronomy Guide of applicatoin timings (Mengel et al., 2017) as reference (Table S6). We set the N use ratio for timing classified as Unadapted in Guide as zero. N ratio in other timings were assigned value according to the relative priority of Poor, Good, and Excellent. Take Ammonium Nitrate as an example, the ratios of Fall, Spring, At planting, and After planting were assigned as 0, 0.2, 0.4, and 0.4, respectively. Nevertheless, the practical situation may different to the Guide due to the use of nitrification inhibitors and vriations in seasonal fertilizer price. We further made adjustments for the timing ratios according to the survey from Bierman et al. (2012).

Supplementary table S6. Agronomy Guide of application timings for 11 N fertilizer types (Mengel et al., 2017)

Fertilizer Material	Fall	Spring	At planting	After planting
Dry Solid Forms				
Ammonium Nitrate	Unadapted	Good	Excellent	Excellent
Ammonium Sulfate	Excellent	Excellent	Excellent	Good
Sodium Nitrate	Unadapted	Good	Excellent	Excellent
Urea	Excellent	Excellent	Excellent	Good
Calcium Nitrate	Unadapted	Good	Excellent	Excellent
Diammonium Phosphate	Excellent	Excellent	Excellent	Excellent
Monoammonium Phosphate	Excellent	Excellent	Excellent	Excellent
Ammonium Phosphates	Excellent	Excellent	Excellent	Excellent

Liquid Forms				
Anhydrous Ammonia	Excellent	Good	Excellent	Unadapted
Aqua Ammonia	Excellent	Good	Excellent	Unadapted
Nitrogen Solutions	Poor	Excellent	Excellent	Unadapted

Sodium Nitrate and Monoammonium Phosphate were not listed in the original guide, we assumed they were suggested the same as Calcium Nitrate and Diammonium Phosphate, respectively.

7 Comparison of national crop-specific application timing ratio between Wade et al., (2015) and this study

We estimated the application timing ratio based on the survey from USDA-ERS (2016). We further compared our results with the national application timing ratio of corn, cotton, spring wheat, and winter wheat reported by Wade et al. (2015). The differences between our study and Wade et al. (2015) are relatively small (0-6 %), with the largest difference found in after-planting cotton (6%) (Table S7).

Supplementary table S7. Comparison of national application timing percentage of corn, cotton, spring wheat, and winter wheat from Wade et al. (2015) and this study, respectively.

Application timing	Wade et al. (2015) (%)	This study (%)	Difference (%)
Fall-corn	20	18	2
Fall-cotton	7	8	1
Fall-spring wheat	21	22	1
Fall-winter wheat	36	36	0
Spring-corn	50	50	0
Spring-cotton	28	31	3
Spring-spring wheat	42	43	1
Spring-winter wheat	5	5	0
After planting-corn	22	23	1
After-cotton	59	53	6
After-spring wheat	5	6	1

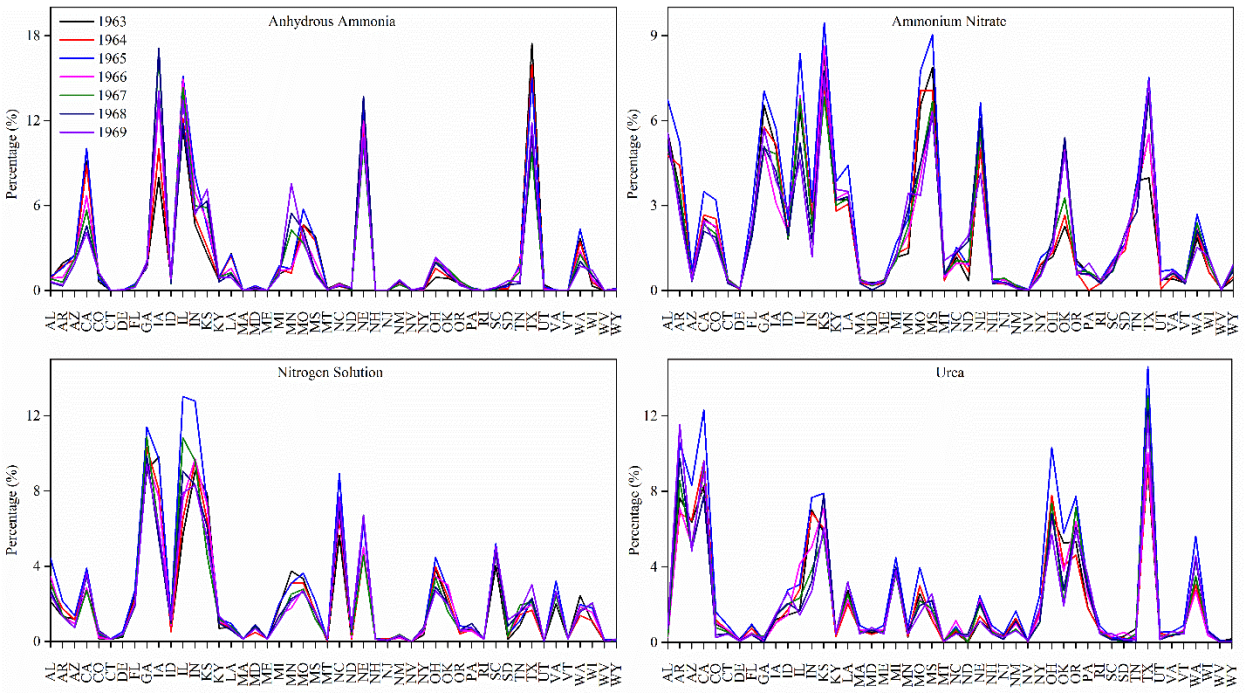
After-winter wheat	45	44	1

Fall refers to the fall in previous year. Spring refers to the time before planting. Difference is the absolute difference between Wade et al. (2015) and this study.

8 Reconstructing historical state-level N consumption of 11 fertilizer types

Due to the lack of annual state-level N use of 11 major N fertilizer types from 1850 to 2015, we utilized the state-level N use of the mid-decade year (every sixth year in a decade) as reference to infer the state-level N use of 11 N fertilizers in the decade. We calculated the percentage of each N fertilizer type consumed by a given state to all states and analyzied the interannual variation of this percentage during 1963 to 1969 to verify the representative of 1966 for other years in the 1960s. Here only the percentage of Anhydrous Ammonia (AnA), Ammonium Nitrate (AN), Nitrogen Solution (NS), and Urea (Figure S1) were showed, accounting for 83% of the total 11 N fertilizers consumption.

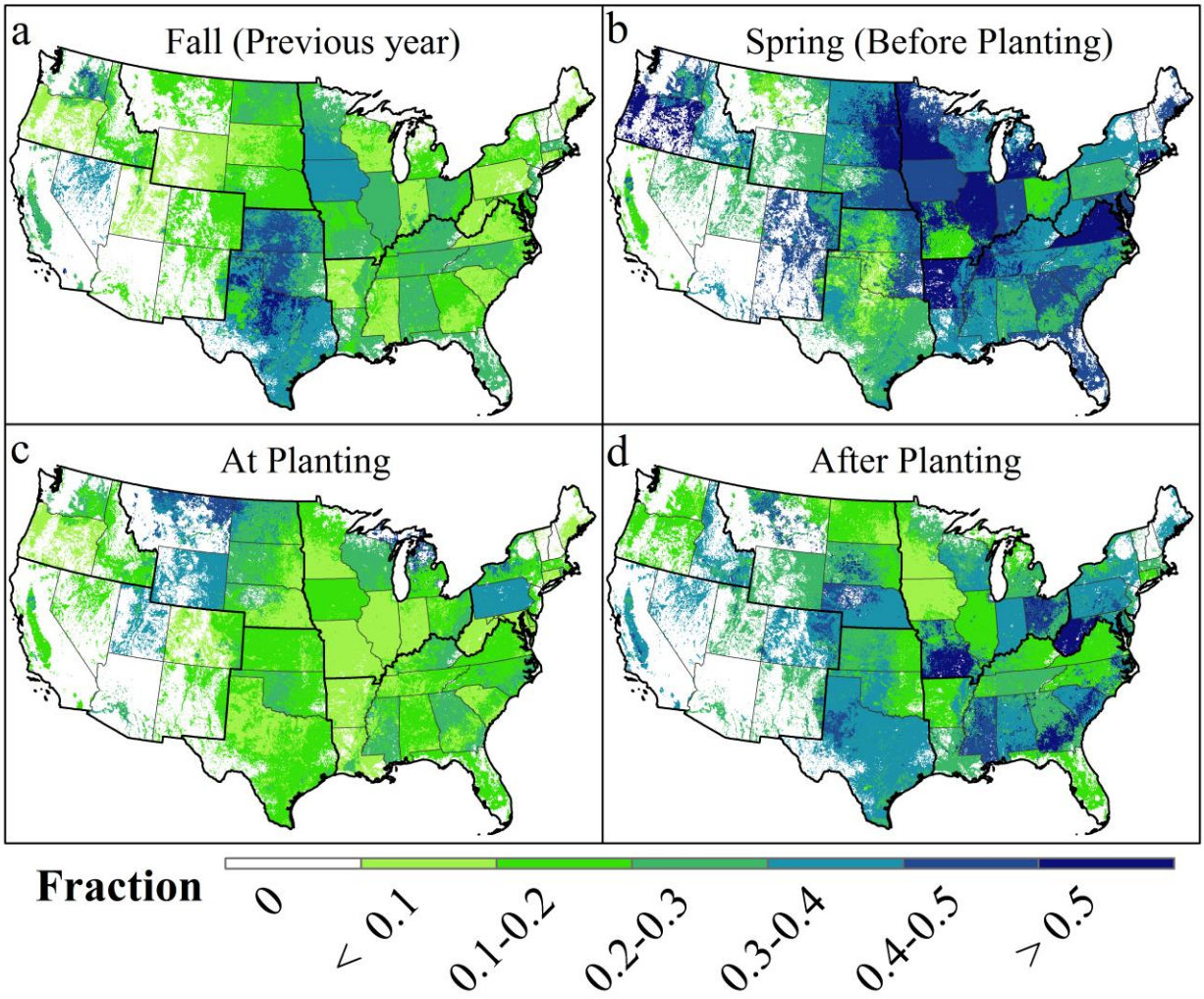
The interannual variation of percentage for most states is relatively small within 7 years (Figure S1). The percentages of some states showed larger flutuations but in reasonable ranges. The few largest interannual changes were found in Iowa for AnA (13.3±3.1%), Missouri for AN (5.4±1.6%), Illinois for NS (8.6±2.4%), and Indiana for Urea (5.2±1.9%).



Supplementary figure S1. Proportion of state-level Anhydrous Ammonia and Ammonium Nitrate fertilizer use to national total consumption in a few selected years.

9 Fraction of N fertilizer use rate in application timing to total N use rate in 2015

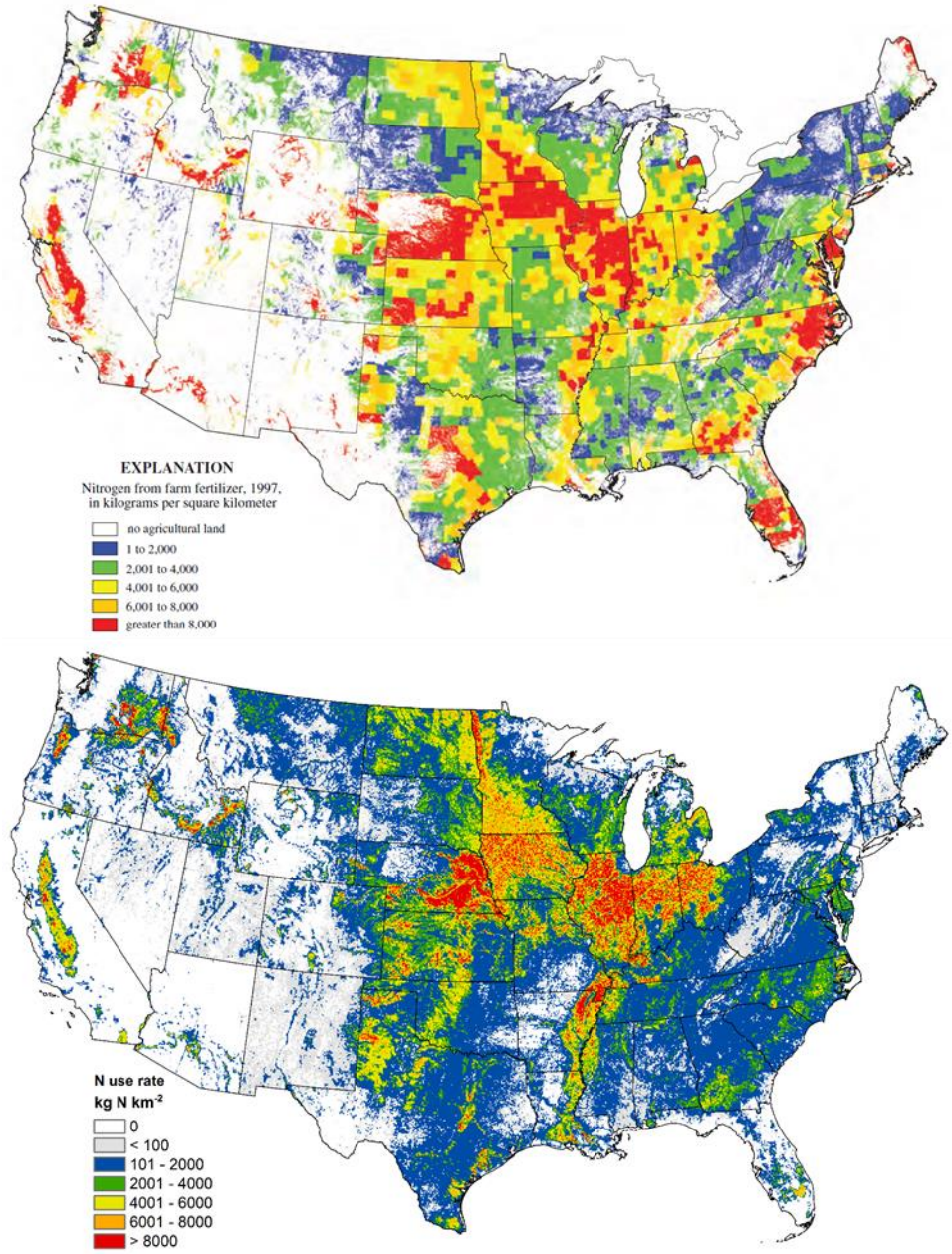
To explore the N use characteristics at different application timings across the continental U.S., we divided the total N use rate by N use rate of each application timing and generated the fraction of N use at four timings in 2015 (Figure S2). Most regions, such as the Northwest, the east of the Northern Great Plains, the Midwest, the Southeast, and the Northeast applied the majority of N fertilizer in spring and after planting (>40%) (Figure S2b&d). Meanwhile, farmers in some regions preferred applying N fertilizer in fall or at planting due to the late growth season of some specific crop types or the habits of fertilizer use. For example, the winter wheat in the Southern Great Plains and the north of the Northwest received most of fall-applied N fertilizer (Figure S2a), which is equivalent to fertilizer use before planting for other crops, and contributed to the rise of winter wheat yield (Mahler et al., 1994). Moreover, corn and soybeans in Minnesota, Iowa, and Michigan in the Midwest received over 30% of annual N in fall, which may be related to the lower fertilizer price and favorable weather and soil conditions in the season (Dinnes et al., 2002). In comparison, N fertilizer uses for spring wheat and barley were only found at-planting in Montana and Wyoming (Figure S2c).



Supplementary figure S2. The fraction of N use rate at four timings to total N use rate in 2015. a, Fall (previous year); b, Spring (before planting); c, At planting; d, After planting.

10 Spatial distribution of N fertilizer use rate in 1997

We compared the spatial distribution of N use rate in 1997 between Ruddy et al. (2006) and our map (Figure S3). Ruddy et al. (2006) used top-down approach to obtain the N use rate by dividing the total county-level N consumption by agricultural area within the county. While we adopted the bottom-up approach to generate the N use rate based on the state-level crop-specific N use rate and cropland distribution map developed by Yu and Lu (2017).



Supplementary figure S3. N fertilizer application rate across the contiguous U.S in 1997. (a) Ruddy et al. (2006); (b) this study

11 Temporal ratio of the consumption of 11 single N fertilizers to national total

We calculated the temporal change of the ratio of 11 N fertilizers to the national N consumption (Figure S4). Generally, the ratios were below 50% before 1950, and increased rapidly to above 85% after 1980. The rest of percentage represents small fraction of other single N fertilizers and large

fraction of mixed N fertilizers used. Due to a lack of data and the difficulty in accounting N content in mixed N fertilizers, we adopted the 11 major single N fertilizers to estimate the NH_4^+-N and NO^{3-}-N fraction of historical N fertilizer use in each state.

However, according to Mehring et al. (1946), ammonium N accounted for only 2% of mixed fertilizer N in 1900 but gradually rose to 72% of mixed N fertilizer from 1925 to 1944. Therefore, we might overestimate the NH_4^+-N fraction before 1925 while underestimate the fraction after 1925.

Supplementary figure S4. Temporal percentage of 11 N fertilizers to total N consumption.

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