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A Review on the Emission Sources of Ammonia and the Factors Affecting Its Loss

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Abstract

Among all the nitrogen species present in the atmosphere, ammonia forms a considerable portion along with the nitrogen oxides. The major sources of atmospheric ammonia are animal feedlot operations including emission from excreta of domestic animals and agricultural activities, followed by emission from synthetic fertilizers, biomass burning and to some lesser extent, fossil fuel combustion. Ammonia emission factor, expressed as the weight of ammonia per unit weight, volume, or duration of the activity emitting it, is generally used in developing emission estimates for emission inventories. The factors determining ammonia loss from soil or from manures are the temperature, pH, humidity, precipitation and the velocity of wind above it.

Key words : Ammonia emission, Ammonia source, Animal feedlot operations, Emission factor, Ammonia

1. INTRODUCTION

Anthropogenic nitrogen compounds constitute one of the major classes of significant air pollutants along with sulfur compounds, volatile and semi volatile organic compounds and toxic metals (Finlayson-Pitts and Pitts, 2000). Among the atmospheric N containing species, ammonia forms a considerable portion beside the nitrogen oxides. Being the most abundant alkaline gas in the atmosphere, ammonia plays an important role in the neutralization of atmospheric acids generated by the oxidation of sulfur dioxide and nitrogen oxides (Mosquera *et al.*, 2005).

Once emitted into the atmosphere, ammonia (NH_3) may undergo conversion to ammonium aerosol depending on the concentration of acids in the atmosphere. The following reactions are generally involved leading to aerosol formation (McCulloh *et al.*, 1998).

$$NH_3\,(g) + H_2O\,(l) \rightarrow NH_4^+ + OH^-$$

$$NH_3\,(g) + H_2SO_4\,(l) \rightarrow NH_4HSO_4\,(l)$$

$$NH_4HSO_4\,(l) + NH_3\,(g) \rightarrow (NH_4)_2SO_4\,(l, s)$$

$$NH_3\,(g) + HNO_3\,(g) \leftrightarrow NH_4NO_3\,(s)$$

$$NH_3\,(g) + HCl\,(g) \leftrightarrow NH_4Cl\,(s)$$

A little amount (approx 10%) is generally oxidized

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by the hydroxyl radical ($OH^{\cdot}$) to form an amide radical.

$$NH_3+OH^{\cdot} \rightarrow NH_2^{\cdot}+H_2O$$

Thus ammonia also influences both the composition and pH of cloud water, thereby affecting the regional air quality and acid deposition patterns (Stelt *et al.*, 2007). Ammonium nitrate and ammonium sulfate are important constituents of airborne particulate matter ($PM_{2.5}$) and can contribute significantly to visibility impairment and regional haze (Battye *et al.*, 2003).

Furthermore, ammonia contributes to the cause of eutrophication in aquatic ecosystem and nitrate leaching (Aas *et al.*, 2007; Webb *et al.*, 2006; Mosquera *et al.*, 2005). Subsequent nitrification of NH_3 leads to acidification of soils and watercourses (Webb *et al.*, 2006). As the ammonium cation transforms to nitrate anion by microbial activity, hydrogen ion gets released contributing to soil acidification (Rodhe *et al.*, 2002). The subsequent recovery of the ecosystem can be very slow (Sanderson *et al.*, 2006). These large NH_3 losses also contribute to N_2O formation (Makhabela *et al.*, 2006).

NH_3 is reported to be hazardous to human health. Exposure to the gas can cause irritation of the eyes, nose and throat of some people (Bai *et al.*, 2006). Exposure to more concentrated levels (above 25 ppm) can cause headaches, nausea, and severe burning of the nose, throat and skin. More severe exposure can lead to permanent damage of the eyes, lungs and skin (Bai *et al.*, 2006).

Hence, the quantification and assessment of ammonia volatilization have been and will continue to be of great significance for sustainable agriculture and environmental protection. The purpose of this review is to determine the different source categories for ammonia emission and to evaluate the factors affecting its loss. This review will thus help one to undertake a choice of strategies to mitigate ammonia emission.

2. SOURCES

In the total global ammonia budget, about 80% stems from anthropogenic sources. The major anthropogenic sources identified are excreta from domestic animals (poultry, dairy cattle, pigs), use of synthetic natural fertilizers, biomass burning, crops, human population, and pets (Oliver *et al.*, 1998).

Table 1. Global emissions of ammonia.

Source of ammonia	Emission (Tg N yr^{-1})
Anthropogenic	
Dairy cattle	5.5
Beef cattle/buffalo	8.7
Pigs	2.8
Horses	1.2
Sheep/goats	2.5
Poultry	1.3
Fertilizer	6.4
Biomass burning	2.0
Subtotal	30.4
Natural	
Wild animals	2.5
Vegetation	5.1
Ocean	7.0
Subtotal	14.6
Total	45.0

Source: Seinfeld and Pandis (1998)

A summary of the global sources of ammonia emission and deposition has been given in Table 1. The best estimate of total global emission of NH_3 is 45 Tg N/yr (Seinfeld and Pandis, 1998). Emissions from fertilized agriculture and domestic animals account for over half the total, reflecting the level of direct human impact on the global NH_3 budget of the atmosphere. Annual denitrification appears to return the largest amount of nitrogen from land to the atmosphere, followed by the transfer of ammonia flux from the biosphere to the atmosphere in the global nitrogen cycle (Schlesinger and Hartley, 1992). A further refinement of the atmospheric budget of ammonia is needed although the excess of ammonium deposition over ammonia emission noted in earlier studies have been reconciled.

The regions with highest emission rates are located in Europe, the Indian sub continent and China reflecting the animal densities, their types and intensity of synthetic fertilizer use. Synthetic fertilizers are large regional sources in Asia contributing 30%

or more with China having the highest percentage.

2.1 Agricultural sources

Unlike oxidized N and S compounds, which are predominantly emitted from industrial processes, ammonia is primarily emitted from agricultural sources. The emissions of ammonia from agricultural activities are very large (Aas *et al.*, 2007; Streets *et al.*, 2003). Livestock buildings and application of manure or slurry into the field are the most important sources of ammonia emissions from agriculture.

Most ammonia losses to the atmosphere are related to the ammonia volatilization from livestock manure (Stelt *et al.*, 2007; Battye *et al.*, 2003). In areas with high livestock-intensities more than 70% of the NH_3 emissions can be attributed to farming practices (Sommer and Hutchings, 2001). The largest part of the gas arising from animal husbandry derives from the animal excreta i.e, from freshly deposited or stored feces and urine. NH_3 is also generated in the manure store outside the house. The reaction involved is the bacterial and enzymatic decomposition of nitrogen containing compounds in the excreta, especially in the urine. A part is probably also produced directly by the animals; including in particular those compounds which make up the species specific odor.

Ammonia emissions from animal husbandry occur both during the housing and grazing periods and from storage lagoons, in case of the U.S. conventional swine farm (Rumsey, 2006). The emission from livestock generally depends on the following factors:

- The nitrogen content of the feed,
- The species, age, type of animal operation and performance (e.g. milk yield, weight gain etc) of the animal,
- Capacity of the animal operation,
- The conversion of N in feed to N in meat, milk and eggs and hence the amount of N in the animal excreta,
- The housing system of the animal operation, including storage of the manures inside the building,
- The storage system of the manure outside the building,
- Climatic conditions in the building and storage system e.g. the prevailing temperature,
- The proportion of time spent by animals indoors and outside, e.g. at pasture or in yards,

Table 2. Summary of ammonia emissions from U.S. animal husbandry operations.

Animal group	Ammonia emission in 2002 (tons/year)
Dairy including dairy cows and dairy heifers	558,094
Beef cattle, bulls and calves	656,648
Poultry including chicken and turkeys	664,238
Swine including breeding and market pigs	429,468
Sheep	24,835
Goats including milking and angora goats	14,028
Horses	71,285

Data source: National emission inventory (2004).

Ammonia emissions from animal manures during and after spreading depend on:

- Spreading techniques and surface exposed of the respective manure,
- Properties of animal wastes including viscosity, ammoniacal nitrogen content and pH,
- Soil properties such as pH, cation exchange capacity, calcium content, water content, buffer capacity and porosity,
- Meteorological conditions including precipitation, temperature, humidity, and wind-speed,
- Time of the year (seasons or month and year),
- The methods and rate of application of animal manures, including, for arable lands, the time between application and incorporation,
- The height and density of the crop or grassland.

A summary of ammonia emissions from U.S. animal husbandry operations has been made in Table 2. In the U.S., the highest emission occurs from dairy followed by poultry and swine farming. The other major sources are from the rearing of sheep, goats (which include the milking and the angora types) and horses. Commonly applied composting strategies of forced aeration and windrow turning called the intensive composting system results in short composting times but also lead to high ammonia emissions (Szanto *et al.*, 2006).

2.2 Industrial sources

Diffuse NH_3 emission also occurs from biomass burning or industrial activities. One example is its use as a coolant in refrigerators (Heeb *et al.*, 2006). Increasing road traffic is also recognized to be an important source of NH_3 (Heeb *et al.*, 2006). Ammonia has been detected in vehicle exhaust gas (Heeb *et al.*, 2006; Huai *et al.*, 2003; Durbin *et al.*, 2002; Baum *et al.*, 2000), in air of exhaust tunnels (Heeb *et al.*, 2006; Emmenegger *et al.*, 2004; Kean *et al.*, 2000) and also in urban ambient air (Heeb *et al.*, 2006; Moya *et al.*, 2004; Perrino *et al.*, 2002). Emission measurements performed in petrol engine vehicles equipped with catalytic converters show NH_3 emission factors of 50 ± 40 mg km^{-1} (Kirchner *et al.*, 2005; Baum *et al.*, 2001; Kean *et al.*, 2000); in case of diesel cars the range of ammonia emission factors is $0.4 \sim 10.9$ mg km^{-1} (Kirchner *et al.*, 2005). In some regions the vehicular NH_3 emission may grow substantially in the future almost equaling the magnitude of the emission from the livestock when the road traffic increases and the fuel sulphur content further decreases (Kirchner *et al.*, 2005; Fraser and Cass, 1998).

2.3 Soil and fertilizer sources

Soil and plant emission account for anywhere between 11% and 28% of the global ammonia budget (Roelle and Aneja, 2002). Ammonia volatilization can occur from soil surfaces whenever free ammonia is present near it (Krupa, 2003). The amount of ammonia volatilization from soil surface occurs based on the texture of the soil and the type of nitrogen fertilizer (urea, ammonium nitrate, ammonium sulfate etc.) applied to the soil. Coarse textured soil volatilizes ammonia more than fine textured soil (Krupa, 2003). Also volatilization occur more in dry soil compared to wet soils (Krupa, 2003).

The most commonly used ammonium fertilizers can break down and release NH_3 after they are applied to soils or crops. In case of urea fertilizers microbial decomposition results in the formation of ammonium carbonate within a few days of application from which emission of NH_3 occurs (National Emission Inventory, 2004). NH_3 emission can also occur from nitrate fertilizer but the release occurs at a much lower rate because the nitrate fertilizer has to be converted to ammonium first. Soil temperatures and pH greatly influences ammonia volatilization (Roelle and Aneja, 2002). An increase in soil pH results in the conversion of ammonium to gaseous ammonia. An increase in soil temperature also increases the loss of ammonia. The loss is further guided by the amount of ammonium nitrogen present (Battye *et al.*, 1994) or the amount of urea present (Webb *et al.*, 2006). Another parameter which influences ammonia volatilization is the soil texture.

2.4 Other sources

Forests and other plants have been shown to emit ammonia to the air. In addition, studies have shown equilibrium between ammonia in the air and ammonia-compounds in plant leaves. As a result, trees, crops and other plants might release more ammonia if emissions from other sources are reduced. It is also known that ammonia emission occur from leaves of senescent plants and from grass after it has been cut (Parmar *et al.*, 2001; Whitehead and Lockyer, 1989).

3. EMISSION FACTOR

An emission factor is a tool that is used to estimate air pollutant emissions to the atmosphere. It relates the quantity of pollutants released from a source to some activity associated to those emissions. Emission factors are usually expressed as the weight of the pollutant emitted divided by unit weight, volume, distance or duration of the activity emitting the pollutant. They can be used in developing emission estimates for emission inventories. These inventories can be used in ambient dispersion modeling and analysis, control strategy development and screening of sources for compliance determinations. When source specific information is not available, the use of emission factors becomes necessary. However, emission factors are generally developed

Table 3. Comparison in literature of the ammonia emission factors from animal husbandry.

Animal type	Emission factor (Kg NH_3/animal/year)	Reference
Cattle	18	Buijsman *et al.* (1987)
	15.9	Kruse *et al.* (1989)
	18.2	Moller and Scheiferdecker (1989)
	12.6	Warn *et al.* (1990)
	23.04	Asman (1992)
	23	Battye *et al.* (1994)
	25 (dairy)	Bowman and Van der Hoek (1997)
	9 (other)	Bowman and Van der Hoek (1997)
	10.9 (dairy)	Steenvoorden *et al.* (1999)
Average value	17.29	
Swine	2.8	Buijsman *et al.* (1987)
	2.11	Kruse *et al.* (1989)
	4.28	Moller and Scheiferdecker (1989)
	3.35	Warn *et al.* (1990)
	5.357	Asman (1992)
	5.4	Battye *et al.* (1994)
	5	Bowman and Van der Hoek (1997)
	5.4	Asman *et al.* (2001)
	7.1 (Breeding pigs)	Steenvoorden *et al.* (1999)
	3.1 (Finishing pigs)	Steenvoorden *et al.* (1999)
Average value	4.93	
Poultry	0.25	Buijsman *et al.* (1987)
	0.18	Moller and Scheiferdecker (1989)
	0.07	Warn *et al.* (1990)
	0.249	Asman (1992)
	0.25	Battye *et al.* (1994)
	0.22	Bowman and Van der Hoek (1997)
	0.08 (Broilers)	Steenvoorden *et al.* (1999)
Average value	0.19	
Horses	12.2	Buijsman *et al.* (1987)
	12.2	Asman (1992)
Average value	12.2	
Sheep	3.1	Buijsman *et al.* (1987)
	2.21	Kruse *et al.* (1989)
	2.47	Moller and Scheiferdecker (1989)
	1.85	Warn *et al.* (1990)
	1.69	Asman (1992)
Average value	2.264	

to represent long term average emissions and they should not be used to estimate short-term or atypical emission. Use of emission factor in such case will result in greater uncertainty. Emission factors are often rated based on the overall assessment of how good a factor is, depending on both the quality of the test or information that is the source of the factor. Emission factors are also judged on the basis of how well the factor represents the emission source.

A comparison of emission factors obtained by different researchers from animal husbandry operations has been made in Table 3. For cattle and calves, the emission factors of Bowman and Van der Hoek (1997) are the highest followed by Asman (1992), Moller and Scheiferdecker (1989), and Buijsman *et al.* (1987). The emission factors of the National Acid Precipitation Assessment Program (Warn *et al.*, 1990) are the lowest. In the comparison of the emission factors for hogs and pigs kept for breeding, the values found by Steenvoorden *et al.* (1999), is the highest followed by Asman (1992), and Bowman and Van der Hoek (1997). Emission factors of Kruse *et al.* (1989) are the lowest. The emission factors of Asman (1992) and Buijsman *et al.* (1987) for poultry are the highest succeeded by Bowman and Van der Hoek (1997). Poultry includes chickens, ducks, and turkeys although chickens represent the largest source of NH_3 emission. However for horses, the emission factors presented by Asman (1992) and Buijsman *et al.* (1987) are the same. In case of emission from sheep, Buijsman *et al.* (1987) records the highest emission factor followed by Moller and Scheiferdecker (1989). The Asman (1992) emission factors are the lowest in this case. A look on the average emission factors reveal that the highest emission occurs from cattle, followed by horses, swine and sheep rearing. Least emission occurs from poultry farming.

The ammonia emission factors from animal husbandry operation from different regions vary with the geographical location of that area. A comparison of emission factors from different locations has been made in Table 4. The EU EPA emission factors are the highest for every animal type. The emission factors compiled and published by CAPSS are

Table 4. Comparison of ammonia emission factors from animal husbandry operations in different reports.

Animal type	Ammonia emission factors		
	Reference No*		
	[1]	[2]	[3]
Dairy cows	28.5	25.0	24.6
Other cattle	14.3	9.0	12.3
Fattening pigs	6.39	7.0	6.39
Sows	16.43	–	16.43
Sheep	1.34	–	–
Horses	8.0	–	–
Laying hens	0.37		–
Broilers	0.28	0.22	
Other Poultry	0.92		

*Source of data [1] EU EPA (2003), [2] Coe and Reid (2003), [3] Data compiled and analyzed by CAPSS (2006)

very similar to the US EPA emission factors.

Ammonia emission factors are representative values that attempt to relate the quantity of the gas released to the ambient air with any activity associated with its release.

The general equation for emission estimation is:

$$E=A*EF*(1-ER/100)$$

Where E=Emission,
A=Activity rate,
EF=Emission Factor,
ER=Overall emission reduction efficiency, %

The ammonia emission factors for animal houses were originally expressed in kg-NH_3 per animal space per year. This led to estimates that were too high because variations in animal weights, mortality and vacancy were not taken into account. As a result, a new parameter was introduced: average animal present (AAP). NH_3 emission factors are expressed in percent volatilization of average nitrogen excretion (kg N) per AAP for each specific source (EPA R & D April 2002). Also a comparison of emission factors from animal husbandry operations for developed and developing countries have been made in Table 5. The difference in the emission factors may be due to the difference in the feeding pattern of the animals in different geographic locations. In developing countries lower feeding levels and lower N content of the feed result in lower volatilization fraction of the N in animal waste. It is assumed that this difference is counteracted by the higher temperatures in the developing countries, as compared to the developed countries.

The emission factors obtained from the use of different types of fertilizers have been summarized in Table 6. Nitrogenous fertilizers are extremely important for agriculture. Many different compounds are used to provide nitrogen in fertilizer. Fertilizers extensively used in agriculture including ammonia, urea, ammonium nitrate, mono and di-ammonium phosphate, ammonium sulphate, ammonium thiosulphate, potassium nitrate, calcium nitrate, and sodium nitrate. These and other compounds used as

Table 5. Comparison of emission factors from animal husbandry for developed and developing Countries.

Category	Developing countries (kg NH_3/head/year)	Developed countries (kg NH_3/head/year)
Dairy cattle	21	25
Beef cattle	10	9
Pigs	11	5
Poultry	0.22	0.22

Data source: EPA research and development (2002)

Table 6. Summary of emission factors for ammonia from fertilizers.

Source category	Reference No*		
	[1]	[2]	[3]
Anhydrous ammonia	12	49	40
Nitrogen solutions	30	97	80
Urea	182	194	150
Diammonium phosphate	49	61	50
Ammonium nitrate	26	24	20
Liquid ammonium polyphosphate	49	61	–
Aqueous ammonia	12	97	–
Ammonium thiosulphate	30	30	–
Calcium ammonium nitrate	–	24	20
Potassium nitrate	–	12	–
Monoammonium phosphate	49	61	20
Ammonium sulphate	97	121	80
Miscellaneous	–	85	20

All the emission factors are expressed in kg NH_3/Mg nitrogen
*Denotes source of data [1] US EPA emission factor (1994), [2] Coe and Reid (2003), [3] EU EPA (2003)

fertilizers listed in Table 6 decompose to release ammonia after being applied to croplands. The ammonia released is being quantified based on the percentage of nitrogen in the applied fertilizer that is lost to the air in the form of NH_3. As can be seen from Table 6, the emission from urea is the highest, followed by the emission from ammonium sulphate and nitrogen solutions applied to agriculture.

Recent researches from the East Asian countries have shown their respective ammonia emission patterns. Results of a detailed NH_3 emission inventory for South Korea indicate that there has been a 21% increase in NH_3 emission over the last 11 years from 143,000 t yr^{-1} in 1988 to 181,000 t yr^{-1} in 1998, the major contributors being livestock and fertilizer application (Lee and Park, 2002). Total emissions in China are estimated at 9.7 and 11.7 million tons NH_3 in 1990 and 1995 and are forecasted to increase to nearly 20 million tons NH_3 in the present scenario, the major contribution coming from N-fertilizer application and livestock (http://www.epa.gov). The overall total NH_3 emission for Japan, from livestock and the application of fertilizer was 2.0×10^5 tonnes NH_3 per year. The total NH_3 emission due to livestock in Japan was 4.6, 6.0 and 4.4×10^4 tonnes NH_3 per year from dairy cows, beef cattle, and pigs respectively (Murano *et al.*, 1995). Sakurai and Fujita (2002) studied the atmospheric budget of NH_3 for the most densely populated area of Japan (Kanto region). On the basis of monitoring results they found that a total emission of 60.8 Gg N a^{-1} occurred from this region.

4. FACTORS AFFECTING THE LOSS OF AMMONIA

In the absence of recent fertilization, NH_3 appears in the soil through a process called mineralization or ammonification whereby micro organisms satisfy their energy needs in the soil by converting amino acids in the dead organic matter to NH_3 according to the following reaction (Roelle and Aneja, 2002).

$$CH_2NH_2COOH + 11/2\ O_2 \rightarrow 2CO_2 + H_2O + NH_3$$

Since the rate of all biochemical reactions rise exponentially with temperature, it is expected that concentration of NH_3 in soil solution will rise with an increase in temperature (Roelle and Aneja, 2002). The NH_3 and NH_4^+ are in equilibrium in the soil solution. Volatilization will occur when the vapor pressure in solution is greater than the vapor pressure in the surrounding air. An increase in temperature will cause an increase in NH_3 concentration in the soil, leading to a rise in vapor pressure of the soil solution and in turn enhancing the rate of volatilization. Thus temperature influences the rate of loss by affecting the rate of biochemical reaction, the equilibrium between NH_3 and NH_4^+, and the rate of water evaporation from the soil surface.

As soil pH increases, more NH_3 is released from the soil. However in highly managed soil, the pH value of the soil column tends to remain fairly uniform and therefore no significant relationships between soil pH and NH_3 flux can be understood. In case of transfer of NH_3 in soil from soil surface to the immediate atmosphere, surface roughness and wind velocity are considered to be other parameters besides soil temperature (Singh and Nye, 1986).

Ammonia release from fertilizers is highly dependent on the amount and type of ammonium fertilizer used. Once applied, urea is rapidly hydrolyzed in soil by the urease enzyme, generating HCO_3^- and NH_3. Under alkaline condition, ammonia is lost to the atmosphere because the rise in pH shifts the $NH_4^+ : NH_3$ equilibrium to the right. Black *et al.* (1985) found that maximum daily losses of ammonia from urea coincided with periods of maximum soil pH. Rainfall close to fertilizer application time leads to its transportation to soil matrix as a dissolved component of the soil water. At this stage volatilization is much reduced because it is placed at a depth beyond possible capillary movement to the surface (Weerden and Jarvis, 1997). However the rainfall after a couple or three days of fertilizer application, does not affect the volatilization rate to a considerable extent. The effect of rainfall has been studied by Black *et al.* (1987) where a simulated 2 mm rainfall event reduced the quantity of urea and increased the quantity of ammonium in soil, which increased the soil pH which in turn also increased the rate of NH_3 loss.

The NH_3 emission from manure is guided mainly by the prevailing meteorological or climatic conditions which include ambient temperature, air velocity, humidity and precipitation. The properties of the manure, the amount that was spread, the way of application and the time between spreading and ploughing also affects the emission. The pH value of the manure is one of the most important factors related to its NH_3 release. The pH value influences the concentration of NH_3 in the aqueous phase, which increases 10 fold per unit rise in pH up to pH 9 (Ni *et al.*, 1999). Cumby *et al.* (1995) reported that the NH_3 release kinetics is related to the air velocity over the manure and the agitation of the manure. Also, air density, air velocity, air viscosity, air temperature, manure temperature are considered to be other important factors.

5. CONCLUSIONS

Agricultural activities and animal feedlot operations are two major sources of atmospheric NH_3. Ammonia emission from soil is determined by its temperature, pH, the roughness of the surface, and the velocity of the ambient air flowing above it. In case of its release from manure surface the determining factors are almost the same. In case of its release from fertilizer, the fertilizer type (i.e. whether ammonium or nitrate) is the principal determining factor. Besides this, the amount and the proximity of the rainfall are other factors affecting its release from a particular fertilizer application. The ammonia emission factor, usually expressed as the weight of the gas per unit weight, volume, distance or duration of the activity emitting NH_3, serves as a tool for estimating the concentration of the pollutant in the atmosphere.

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