

Specificities of considering migration capability of heavy metals in assessing ecological hazard of polluting soils of urban ecosystems

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ABSTRACT

Necessity of assessing how badly are soils of urban ecosystems polluted by heavy metals has been motivated by the growth of technogenic load and awareness of the hazard level. The study aimed to identify and justify the use of an indicator to assess the actual environmental risks of soil contamination with heavy metals in urban ecosystems. This indicator eliminates the influence of soil buffering capacity on metal fixation and considers the migration potential under specific conditions of spatial pollution distribution. The studies were conducted within the urban ecosystem of Dnipro city (Ukraine) – a powerful center of metallurgy, machine-building, metal-processing industries, and rocket-space construction. In this work, the gross content and mobile forms of the following five heavy metals were studied: Cu, Zn, Pb, Cd, and Ni. The assessment of pollution intensity was carried out using the Nemerov pollution index (NPI) for the gross content and mobile forms in order to reveal the level of ecological hazard caused by the growth of heavy metals in soil. It was ascertained that by the level of ecological hazard, the pollution intensity of heavy metals in soils of the urban ecosystem of Dnipro city can be represented as the following inequality sequence: $Zn > Pb = Cu > Cd > Ni$. The migration capability of heavy metals is conditioned by an increment of their gross content, whereas the city soil is losing its buffer property. A tendency of the NPI growth is revealed within the gross content to potentially mobile forms as a result of considering the migration capability of heavy metals. The NPI application efficiency is substantiated to take into account specificities of how urban ecosystems are formed and function, including variety and spatial propagation of pollution by heavy metals, as well as their migration capability.

Keywords: heavy metals, environmental safety, estimation of man-caused load, chemical pollution, migration, urban soil, Nemerov pollution index.

INTRODUCTION

The necessity of assessing the heavy metal pollution of soils of urban ecosystems is connected not only with the ecological situation, which is complicated by the increase of technogenic loads on the components of the natural

environment (Long et al., 2021; Nyiramigisha et al., 2021; Pohrebennyk et al., 2022), but also with the awareness of the degree of hazard, caused by these toxicants (Krcmar et al., 2018; Yakovyshyna, 2023).

Peculiarities of the element-wise pollution by heavy metals within the urban ecosystem

contribute to the formation of local areas of sharp increase in their concentration, while their content distribution is pretty uniform, but above their background values. The indicators used to determine the ecological hazard of element-wise pollution of the soil by heavy metals – the concentration coefficient and the hazard coefficient – do not take those peculiarities into account. In addition, when assessing soil pollution of urban ecosystems by heavy metals, they are usually limited to using only their gross content after acid treatment of the soil. However, under the conditions of violation of the buffering capacity due to the influence of anthropogenic activity, this indicator will not be informative, because the capability of metal compounds to migrate, which is reflected by their mobility, can increase by about 10 times compared to the zonal soil. Therefore, it is quite important to define an indicator for assessing the intensity of element-wise pollution of soils of urban ecosystems by heavy metals. This indicator would firstly take into account the formed urbanized background and the presence of separate centers of intense pollution – hotspots. Secondly, it would take into account the mobility of metals in the soil, that is, be determined in terms of their potentially mobile forms.

ANALYSIS OF RECENT RESEARCH AND PUBLICATIONS

While technogenic load on soils of urban ecosystems is assessed following soil element-wise pollution by heavy metals, an advantage is given to the coefficient of their concentration. The methodological approach is based on the correlation of the results of analytical studies of the content of metal cations in a specific soil sample with the standards proposed above (Vodyanitskii, 2010; Trigub and Domuschy, 2022). This results in the fact that pollution is determined under the conditions of exceeding the natural fluctuation according to the background concentration (Hasanetal., 2020).

When heavy metal pollution is regulated, the world community mostly relies on their content in the zonal soil, that is, on the natural geochemical background (Hakanson, 1980), which is interpreted in various ways: the arithmetic mean, the geometric mean, or even the modal value of the concentration (Yakovyshyna, 2019). It is considered appropriate to accompany this indicator with an assessment of the variation of analytical data.

Scientists propose determining the background concentrations of metal within the subtype of soils taking into account natural differences as average content (Tomlinson et al., 1980; Ghazaryan et al., 2015). For the soils of Ukraine, the concentrations of the natural geochemical background in accordance with the specified requirements were established by scientists of the National Scientific Center “Institute of Soil Science and Agrochemistry named after O.N. Sokolovsky” (Ukraine). For urban soils, the intensity of element-wise pollution by heavy metals is determined not only by exceeding the natural geochemical background, but also by taking into account the spread of the pollution over the studied territory. In addition, under the conditions of technogenic pollution of territory of cities, one should take into account the already formed urbanized geochemical background and consider the presence of centers of intense pollution (hotspots). Recently, the Nemerov pollution index (NPI) has been increasingly used due to it taking into account the average value and the highest value of pollution by heavy metals, which, in turn, reflects all the hazards of technogenically loaded urban ecosystem (Guan et al., 2014; Kowalska et al., 2016).

The problem of the present research was to justify assessing the intensity of soil pollution of urban ecosystems by using NPI as such that takes into account the capability of hazardous metal compounds to migrate and the features of the spatial distribution of pollution, namely, the urbanized background and the presence of hotspots.

METHODOLOGY OF RESEARCH

Characteristics of the research site

The urban ecosystem of the city of Dnipro occupies an area of 390.8 km², of which the built-up part makes up 55%, landscape and recreational areas – 30%, whereas water and other surfaces – 15% (Pavlov et al., 2000). The length of the city is 22 km from north to south, and 33 km from east to west.

Among the sources of Cu compounds entering the environment of the city of Dnipro with subsequent deposition in the soil, non-ferrous metallurgy enterprises, transport, welding, galvanization, fuel burning should be noted; Zn – industrial enterprises producing non-ferrous metals, cast iron and steel, glass and cement, thermal power plants and motor vehicles; Pb – metallurgical,

metal-processing, machine-building, and chemical enterprises; Cd – ferrous and non-ferrous metallurgy enterprises, as well as the work of communal services on the processing and disposal of solid household waste; Ni – thermal power plant during hard coal combustion.

Annually, before the beginning of the war, the total emission of heavy metals into the atmospheric air from stationary sources was: Cu – 2.061 t, Zn – 4.404 t, Pb – 1.972 t, Cd – 0.508 t, Ni – 1.521 t. This is re-calculated per 1 km² of the city area as the following: Cu – 0.0015 t, Zn – 0.011 t, Pb – 0.005 t, Cd – 0.001 t, Ni – 0.004 t or Cu – 0.0026, Zn – 0.0044 kg, Pb – 0.002 kg, Cd – 0.0005 kg, Ni – 0.0015 kg per capita (the data is taken from the Main Department of Statistics in the Dnipropetrovsk region).

Site network creation and soil sampling

For the effectiveness of NPI use, a network of key soil sampling sites, totaling 65, was created by applying a 2 × 2 km grid to the city territory. The grid size was determined by the area and population of the city (Baluk et al., 2004). The key areas reflected the origin and properties of the soil, bedrock, topography, vegetation, hydrology of the territory and its use. The sampling area was about 200 m². The distribution of point sampling sites was uniform, under the conditions of avoiding atypical forms of microrelief (pits, ditches, ravines), fires, waste piles, being visually heterogeneous in composition and color spots. Soil samples were taken by the envelope method from a depth of 0–10 cm. A representative sample comprised 25 individual samples (State standard of Ukraine, 2019). The

selection of soil samples and their preparation for analysis was carried out in accordance with the requirements of current standards (State standard, 1984; 1994). The distribution of sampling sites across the districts of the city is given in Table 1.

Determining the content of heavy metals in soil

While soil pollution by heavy metals is assessed, researchers are usually limited to the use of only their gross content, but this indicator will be quite non-informative, because it does not reflect the capability of toxicants to migration. However, urbanized soils have been violated by buffer capacity due to the development of the urban ecosystem and are unable to deposit cations of metals to the same extent as zonal non-polluted soils, which in turn affects the significant increase in their mobility and capability for migration. Therefore, the pollution assessment was carried out by gross content and mobile forms of heavy metals. The gross content of heavy metals in the soil was determined after its acid treatment on the atomic-absorption spectrophotometer, potential-mobile forms – in the extract of 1H HCl. The analysis of the samples of the heavy metal content was carried out by mathematical statistics.

Assessment of soil pollution by heavy metals

Soil pollution with lead compounds, taking into account their capability to migrate, was assessed using NPI, as presented below (Li et al., 2020):

$$NPI = \sqrt{\frac{P_{ave}^2 + P_{max}^2}{2}} \quad (1)$$

Table 1. Distribution of soil sampling sites on the territory of the Dnipro city

District	Area, hectares	Population, thousands of people	Total sampling sites	Characteristics of sampling sites			
				Industrial zone	High-rise building	Private sector	Green zone
Amur-Nizhnyodniprovsky	7162.6	154.4	13	1	-	9	3
Industrial	3267.9	132.7	5	2	2	1	-
Novokodatsky	8870.2	151.7	12	-	4	4	4
Samarsky	6683.4	77.9	8	1	-	4	3
Soborny	4409.3	169.5	8	-	3	1	4
Central	1040.3	67.2	3	-	2	-	1
Chechelivsky	3589.7	120.6	9	4	-	5	-
Shevchenkivsky	2679.4	152.0	7	1	2	2	2
Left-Bank	19300.7	438.8	26	4	2	14	6
Right-Bank	18402.1	587.2	39	5	11	12	11

where: P_{max} is the maximum value of the index of soil pollution by metal compounds; P_{ave} is the average value of the index of soil pollution by metal compounds, calculated according to the following formula:

$$P_{ave} = \frac{1}{n} \sum \frac{C_i}{S_i} \quad (2)$$

where: C_i is the content of metal cations in the studied soil (mg/kg); S_i is the background content of metal cations in the zonal soil – ordinary low-humus heavy loam chernozem for the conditions of the Northern Steppe of Ukraine, to which the urban ecosystem of the Dnipro city belongs territorially (mg/kg); n is the number of studied samples.

NPI calculations for gross content and potentially mobile forms of heavy metals were performed using Microsoft Excel. An example of the calculation is shown in Table 2.

Assessment of the intensity of pollution by hazardous lead compounds according to the obtained NPI values was performed using the scale given in Table 3.

RESULTS AND DISCUSSION

The gross content of Zn in the soils of Dnipro ranged from 15.27 to 959.11 mg/kg with an urbanized geochemical background at the level of 290.99 mg/kg or 2.9 threshold limit value (TLV). The Zn pollution of the soils of the urban ecosystem of Dnipro was characterized by increased migration capability. The content of potentially mobile forms of Zn (the extract of 1H HCl) reached 99% of the total and varied within a wide range. Thus, the maximum

concentration was 307 times higher than the minimum, while the value of the urbanized background exceeded the natural concentration by one order of magnitude.

On the territory of the city, the urbanized background for gross Pb content was 65.86 mg/kg, which is 2.94 times higher than the natural level and 2.20 times higher than the TLV. Potentially mobile forms amounted to 51.17 mg/kg, or 3.58 natural fluctuations at a potential mobility of 75% of the gross content, which indicates the loss of buffer properties by anthropogenically transformed soil.

The urbanized background, based on the average value of Cu concentrations sampled from the monitoring sites in Dnipro, was higher than the natural one in the zonal soil. Namely, for gross content, – by 2.18 times, for potentially mobile forms – by 2.47 times. Unlike the two previous pollutants, the potential mobility had a clear bond to the pollution sources and was up to 82%.

The concentration of gross Cd content in the soils of Dnipro was within TLV, but higher than the natural geochemical background. The value of the urbanized background was at the level of 0.59, with a minimum and maximum range of 0.133 and 1.647 mg/kg, respectively. An increase in the content of potentially mobile forms of Cd

Table 3. Scale of intensity of soil pollution by heavy metals according to NPI (Li et al., 2020)

Value of NPI	Intensity of soil pollution
$NPI \leq 0.7$	Clean soil
$0.7 < NPI \leq 1.0$	The boundaries of the hazard of soil pollution
$1.0 < NPI \leq 2.0$	Slight soil pollution
$2.0 < NPI \leq 3.0$	Moderate soil pollution
$NPI > 3.0$	Heavy soil pollution

Table 2. An example of NPI calculation based on the gross Pb content

Calculation of NPI (Pb, gross content)										
1	Coordinate	District	Gross content	Ci/Si	8-Amur-Nizhnyodniprovsky District					
2	A3	8	57.7	2.58	Pave	Pave^2	P_{max}	P_{max}^2	$(Pave^2 + P_{max}^2)/2$	NPI
3	A4	8	38.39	1.71	2.85	8.1002	17.10	292.334	150.2169332	12.26
4	A5	8	59.89	2.67	7-Industrial District					
5	B4	8	14.16	0.63	Pave	Pave^2	P_{max}	P_{max}^2	$(Pave^2 + P_{max}^2)/2$	NPI
6	B5	8	6.74	0.30	1.54	2.37931	3.58	12.8126	7.595935411	2.76
7	B6	8	23.42	1.05	6-Samarsky District					
8	B7	7	28.06	1.25						
9	B8	7	11.2	0.50						

was observed, which was 2.5 times higher compared to natural conditions.

In the soils of Dnipro, the gross content of Ni is quite low – 3.2–26.78 mg/kg against an urbanized background of 10.93 mg/kg, which is within TLV. This indicates the absence of degradation and correlates with the natural geochemical background of zonal soils – ordinary chernozems. The lower boundary of the gross Ni content is explained by the processes of deconcentration during soil disturbance due to construction accompanied by dilution by construction debris and underlying rocks, whereas aerogenic input with emissions from industrial enterprises is absent. The content of potentially mobile forms was almost half of the total.

According to high reliability of the approximation of the regression equations and the values of the correlation coefficients between the gross content and potential mobile form of heavy metal (Cu, Zn, Pb, and Cd) in the soils, (Table 4), it was determined that the pollution process affected the increment of their capability to migrate. The lack of a close relationship between the Ni²⁺ concentrations obtained by different extracts was caused by deconcentration during the development of the urban ecosystem and the absence of powerful sources of pollution.

The distribution of heavy metal pollution according to gross content and mobile forms, across the territory of the Dnipro urban ecosystem was quite scattered for Zn. As for Cu and Pb, 3–4 hotspots could be identified with reference to industrial zones and a gradual decrease in concentration. Cd and Ni, relative to the gross content, were characterized by a slow monotonous increase in concentration within the bounds of TLV from the periphery (private sector) to industrial zones.

Under the conditions of intensive pollution by heavy metals and the loss of buffering capacity of

the soil, the value of NPI for potentially mobile forms was higher than for the gross content of Zn, Pb, and Cu (Figure 1, a-c). For Zn, the Nemerov pollution index corresponded to severe pollution in both cases, Pb and Cu – from severe to insignificant. In relation to Cd, the gross content of pollution was determined mostly as insignificant, and in terms of mobile forms – severe (Figure 1d). The conditions of the NPI value of potentially mobile forms lower than the NPI value of the gross content were preserved for peripheral regions and the region with the absence of powerful industrial zones for metals the gross content of which did not exceed TLV, such as Ni (Figure 1e). According to the degree of ecological hazard of soil pollution of Dnipro, the studied heavy metals can be arranged in the form of the following series: Zn > Pb = Cu > Cd > Ni.

The use of the NPI indicator to assess the intensity of soil pollution of urban ecosystems with metal compounds made it possible to assess the hazard taking into account the variety and spatial distribution. This was quite clearly tracked for Zn, Pb, and Cu, especially for the Amur-Nizhnyodniprovisky and Novokodatsky districts, which include powerful industrial zones and private sectors on the periphery, where the presence of hotspots marked a significant increase in the NPI value. In contrast to assessing the soil pollution using the concentration coefficient and the hazard coefficient, the information provided by NPI is more compact and complete, because it includes the existing urbanized background by average value and the maximum content of metal compounds recorded in the studied area (in the considered case – by districts and cities), rather than singly at separate key sampling sites.

Taking into account the capability of heavy metals to migrate led to a change in the pollution category to a stronger one. Factors contributing to the reduction of the migration capability of heavy

Table 4. The relationship between potentially mobile forms and the gross content of heavy metals in the soil

Heavy metal	Regression equation	Coefficient of approximation	Coefficient of correlation
Cu	$Cp.-m.f._{Cu} = -0,0007Cg.c._{Cu}^2 + 0,5608Cg.c._{Cu} - 0,2958$	0.786	0.854
Zn	$Cp.-m.f._{Zn} = 0,000009Cg.c._{Zn}^3 - 0,0011Cg.c._{Zn}^2 + 1,0695Cg.c._{Zn} - 25,399$	0.900	0.945
Pb	$Cp.-m.f._{Pb} = -0,000006Cg.c._{Pb}^3 + 0,0009Cg.c._{Pb}^2 + 0,5663Cg.c._{Pb} + 4,9997$	0.930	0.962
Cd	$Cp.-m.f._{Cd} = -0,3845Cg.c._{Cd}^3 + 0,8125Cg.c._{Cd}^2 + 0,3493Cg.c._{Cd} + 0,0522$	0.841	0.910
Ni	$Cp.-m.f._{Ni} = 0,3632Cg.c._{Ni}^{1.0889}$	0.637	0.729

Note: Cg.c – is the concentration of metal cations after acid treatment (gross content), Cp.-m.f. – is the concentration of metal cations in the 1N HCl extract (potentially mobile forms).

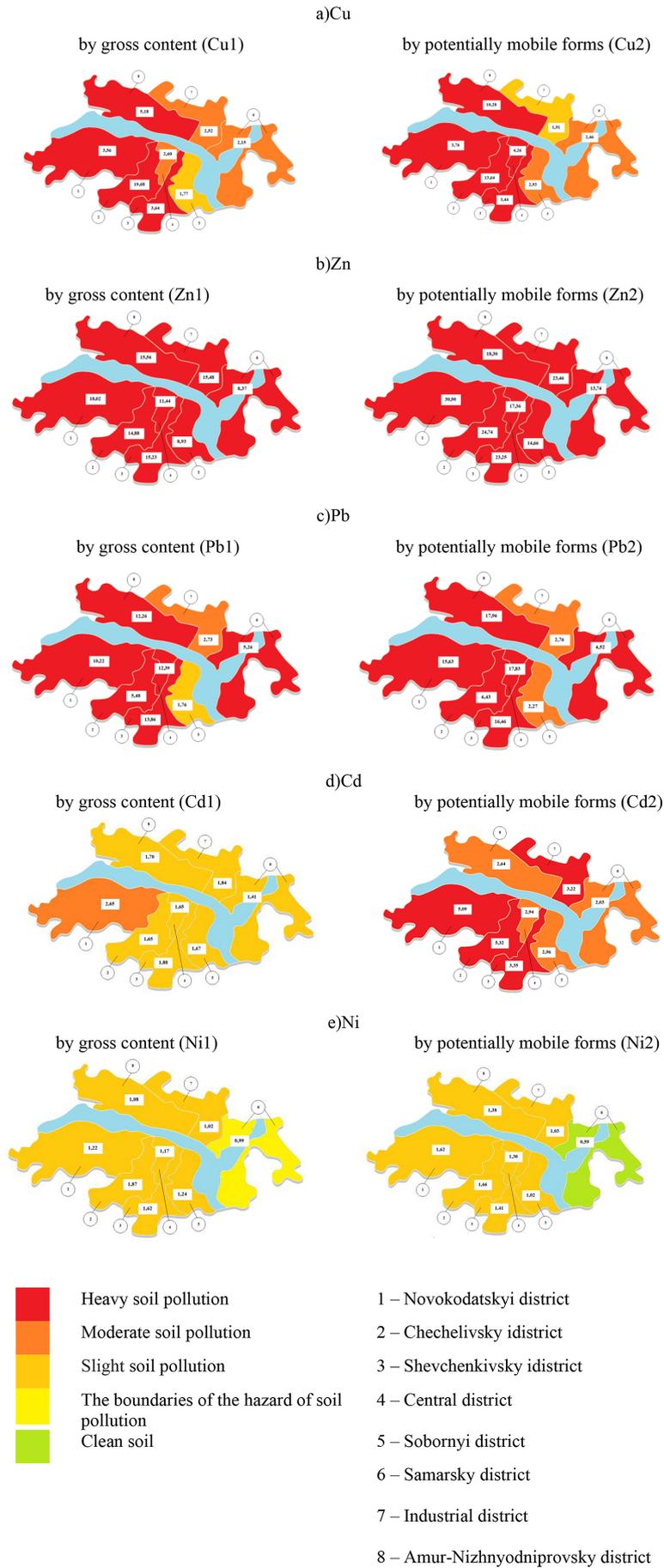


Figure 1. Intensity of soil pollution by heavy metals according to NPI

metals (Zhongetal, 2020) under the conditions of an urbanized environment do not fully work caused by soil degradation due to the intervention of human activities. Therefore, taking into account the migration capability of heavy metals when determining the intensity of soil pollution according to NPI makes it possible to assess real ecological hazard, as confirmed by the results of the conducted research.

CONCLUSIONS

The intensity of heavy metal pollution of the urban ecosystem of the city of Dnipro (Ukraine) is determined using NPI, which varied according to the degree of ecological hazard in the following order: $Zn > Pb = Cu > Cd > Ni$. The study revealed a heterogeneous distribution of soil contamination by Zn compounds within the city. The hotspots of Cu and Pb were detected near industrial zones, with concentrations gradually decreasing with distance from these areas.

Using the methods of mathematical statistics, it was determined that the migration capability of heavy metals is caused by an increase in their gross content while the urban soil loses its buffering capacity. Significant correlation coefficients (0.854–0.962) and approximation coefficients (0.786–0.930) were observed, particularly for Cu, Zn, Pb, and Cd. In contrast, during the deconcentration of Ni^{2+} in the soil, the relationship between the total content and potentially mobile forms was weak, with correlation and approximation coefficients of 0.729 and 0.637, respectively. The intensity of soil contamination in Dnipro's urban ecosystem was classified as strong for Zn compounds, ranging from weak to strong for Cu, Pb, and Cd, and weak for Ni, with some localized areas of uncontaminated soil.

A tendency of NPI growth is revealed within the gross content (Cu – 1.77–5.18; Zn – 8.37–18.02; Pb – 1.76–13.86; Cd – 1.41–2.65) to potentially mobile forms (Cu – 1.91–10.28; Zn – 13.74–30.90; Pb – 2.27–16.46; Cd – 2.03–5.32) as a result of considering the migration capability of heavy metals, except for two city districts (Chechelivskiyi and Shevchenkivskiyi) where copper contamination was observed. The NPI values for total nickel content were slightly lower than those for its mobile forms, ranging from 0.99 to 1.87 and 0.59 to 1.66, respectively. This discrepancy is due to the deconcentration of nickel

in soils during urban development and the small quantities emitted by industrial facilities.

The NPI application efficiency is thoroughly substantiated, which, as opposed to the concentration coefficient and the hazard coefficient, takes into account specificities of how urban ecosystems are formed and function, including variety and spatial propagation of pollution by heavy metals, and also their migration capability.

The results of the research can be used to justify the involvement of NPI in the assessment unit of a system for monitoring the state of urban ecosystems. This will make it possible to determine the ecological hazard taking into account the capability of metal compounds to migrate, the formed urbanized background, and the maximum level of pollution in hotspots. Assessing the level of ecological risk based on the content of potentially mobile forms will enable a more precise calculation of ameliorant doses relative to the actual pollution levels, thereby significantly enhancing the effectiveness of soil chemical detoxification efforts.

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