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A contribution to the ecology and enology of platinum

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Abstract Since the introduction of the catalytic cleaning of vehicle exhaust gases, platinum is discussed as environmentally relevant element. Results of such investigations are compared with “natural” platinum background concentrations, which were determined in selected environmental and biological samples. Wine serves as an example for the following the path of platinum, beginning with the uptake during the plant growth, continuing with the fermentation process of grape juice and resulting in the final product.

1 Introduction

Discussions concerning the exposition of the environment to toxic elements (e.g. cadmium, lead, mercury) from several sources have been carried on for a long time. Platinum was included into the investigations since the widespread introduction of the catalytic cleaning of the exhaust gases from motor cars. The determination of platinum in biological and environmental materials at the “natural” level was made possible by the development of extremely sensitive analytical methods (especially adsorptive voltammetry in combination with an appropriate sample preparation) with detection limits in the ng/kg range and below. In this connection the enological topography offers a good possibility to follow the path of platinum beginning from the uptake during the growth up to the resulting food product (wine).

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2 Properties and application of platinum

The geochemical frequency of platinum is estimated to be about 5–10 ppb. Consequently it belongs to the rare elements (position 72 of the frequency table). As siderophilic element it is found in natural deposits as solid metal together with palladium, iridium and osmium. Otherwise many minerals with various Pt-content and other platinum group metals are known. An important Pt-mineral is Sperrylith (PtAs_2). Platinum shows the typical properties of a transition metal, among others occurrence in numerous oxidation states in its compounds (mainly 0, +2 and +4), the tendency to form a high diversity of complex compounds and the catalytic activity. The “precious” metal platinum shows a good resistance against chemical attacks, but it is readily soluble in aqua regia.

The catalytic activity of platinum, often in combination with palladium and rhodium, is used in the oil industry, the pharmaceutical industry, for the synthesis of ammonia and other chemical products. An important application for platinum is the use in catalytic converters for motor cars, in order to reduce the pollutants in the exhaust gases, especially hydrocarbons, carbon monoxide and the oxides of nitrogen, to about 10% of their original levels. Further use of platinum is to be found in the jewellery industry, in apparatus technology, glass and textile industry.

Platinum compounds are successfully used in tumor therapy. The best known of these is “cis-platinum” ($\text{cis-}[\text{PtCl}_2(\text{NH}_3)_2]$); recently carboplatinum (or parapl原因) has also been employed as an antitumor drug. Lyophilic cis-platinum is administered in daily doses of several milligrams of platinum [1].

The annual platinum production amounts to about 100 tonnes worldwide [2]. 42% of the production was used for catalytic converters for motor cars, 39% in the jewellery industry, 4% in the glass industry and in chemistry, respectively, 5% in electronics and 6% in the

Table 1 Platinum concentrations in biological and environmental materials

Sample	Pt-Concentration ($\mu\text{g}/\text{kg}$)
<i>Soils (uncultivated)</i>	
Mountain (1)	0.09
Mountain (2)	0.07
Mountain (3)	0.03
Coast (1)	0.26
Coast (2)	0.25
Forest (1)	0.10
Forest (2)	0.15
Mean	0.14
<i>Soils (cultivated)</i>	
Garden (1)	1.6
Garden (2)	3.9
Garden (3)	1.6
Garden (4)	0.15
Garden (5)	0.22
Agriculture	0.22
Garden mould, trade (1)	0.88
Garden mould, trade (2)	0.27
Garden mould, trade (3)	0.33
Mean	1.12
<i>Soils (motorway)</i>	
	22.7
	15.8
	20.0
	19.3
	31.7
	15.6
Mean	20.9
<i>Water</i>	
Drinking water	0.0001
Rain water (1)	0.00017
Rain water (2)	0.00008
Well water	≤ 0.000004
Fen water	0.0013
Glacier water	0.0001
Sea water (1)	0.0024
Sea water (2)	0.00028
Deionized water (Milli-Q)	0.00005
<i>Plants, vegetables, foodstuffs</i>	
Grass (1)	0.14
Grass (2)	0.29
Potatoe, peeled	0.10
Carrot	0.31
Cabbage	1.1
Lettuce	2.1
Celery	1.3
Onion, peeled	0.03
Flour	≤ 0.04
Whole meal	≤ 0.04
Honey	≤ 0.002
<i>Fertilizers</i>	
Natural	0.032
Mineral	2.3
Mineral, liquid (1)	0.43 $\mu\text{g}/\text{L}$
Mineral, liquid (2)	0.081 $\mu\text{g}/\text{L}$
<i>Fuels</i>	
Petrol	0.0002
Diesel	0.0006
<i>Air [5]</i>	
Air dust	5–130
Air	0.3–30 pg/m^3

Table 1 (continued)

Sample	Pt-Concentration ($\mu\text{g}/\text{kg}$)
<i>Human body fluids ($\mu\text{g}/\text{L}$) [4]</i>	
Urine, normal	0.0005–0.014
Urine, occupational	0.021–2.90
Blood plasma, normal	≤ 0.0008 –0.0069
Blood plasma, occupational	0.095–0.28
Whole blood, normal	≤ 0.0008 –0.0069
Whole blood, occupational	0.032–0.18
<i>Reference material (materials not certified for platinum)</i>	
BCR Human hair	0.076
BCR Pig kidney	0.040
NBS Orchard leaves, 1571	≤ 0.04
NBS Bovine liver, 1577	≤ 0.04
NIES Vehicle exhaust particulates	210

oil industry and others (1994). So far more than 50% of the produced platinum found application in environmentally relevant spheres.

3 Experimental

3.1 Sample materials

The selection of the samples was more or less accidentally. Therefore it does not claim to be complete and statistically ensured.

For the evaluation of ecological platinum concentrations (Table 1) commercial products were used (garden mould, vegetables, foodstuffs, fertilizers, fuels). Furthermore, samples were taken from locations in Dortmund or its vicinity, in particular from locations far from traffic (soils, waters).

The wines (bottled wines) generally originated from wine-grower plants in Rheinhessen (Germany), many from the village of Ober-Ingelheim. "Older" (before 1982) and "younger" (after 1985) vintages were distinguished. The vineyard locations near high traffic roads were carefully selected. Criterions were within 1 m distance from the road, position deeper than the road level and preferred direction of the wind. The grape must (Niersteiner Müller Thurgau, 1994) originated from an "unexposed" vineyard location and served as sample material for the fermentation studies, too. The soil samples from vineyards were taken from surface-near horizons (0–10 cm). Each sample was a mean of five single samplings. The plant parts of the "vitis vinifera" were separately sampled as leaf, stalk and bunches of grapes, each consisting of five single samplings. Fertilizers, pesticides and materials for wine treating were commercial products.

3.2 Determination of platinum

Solid samples (soils, plants and plant parts, fertilizers, pesticides) were submitted to digestion after drying at 104 °C and homogenization without any further preparation. Liquid samples (up to 10 mL wine and grape must, 1 L water samples) were reduced in volume by evaporating to about 1 mL and then digested. Fuels were treated with bromine in order to destroy the double bindings of organic molecules (binding partner for metal ions) [3]. Then the platinum was extracted twice into 5 mol/L hydrochloric acid. The combined aqueous phases were reduced in volume by evaporating to about 1 mL.

The samples, prepared as described above, were decomposed with a mixture of nitric, hydrochloric and perchloric acids (e.g. 200 mg solid sample, 3 mL 65% HNO₃, 0.5 mL 30% HCl and 0.1 mL 70% HClO₄), using high-pressure ashing (130 bar, 320 °C, apparatus: HPA[®], Kürner, Rosenheim, Germany) in 70-mL quartz vessels. The quantifications were performed by adsorptive stripping voltammetry (apparatus Polarecord E625, VA Stand 663 equipped with a Multi-Mode-Electrode, VA Timer E608, Metrohm, Herisau, Switzerland). Blanks were checked by using reagents of highest purity. The detection limit depended on the sample matrix. It ranged from a few pg Pt/L in water samples to about 50 ng Pt/kg in soil samples. The relative standard deviation was determined to be 5–10%. The analytical recoveries of platinum amounted to 90–100%. As no certified reference materials were available, samples, doped with known amounts of platinum, were used. In addition, the accuracy of the results confirmed by exchange of samples with other laboratories and participation in interlaboratory trials [4]. The procedure has been described in detail already in other papers [1, 5, 6].

4 Ecology of platinum

Investigations concerning platinum group metals (platinum, palladium and rhodium) in the environment have been undertaken more intensively since vehicle exhaust catalysts were introduced (in USA 1975, Germany 1984, UK 1993). Using this technique, no addition of lead to fuels was required. Soon speculations were uttered, that with the emission of platinum metals from the catalysts a new environmental problem will arise, what means that one toxic metal (lead) in motor car exhausts would be replaced by another (especially platinum).

In principle, a certain amount of platinum metal emission must be expected from catalytic converters. Metals in a highly dispersed form together with aluminium oxide as a carrier are released into the air as suspended particles and are subsequently deposited. The amount of platinum emitted from catalytic converters is up to now discussed controversially. According to measurements under laboratory conditions with a computer controlled dynamometer, the emission concentrations ranged from 3 to 40 ng/m³ (particulate) [7], respectively from 80 to 110 ng/m³ (total emission) [8], which is equivalent to approximately 3–80 ng Pt per kilometre driven. Several speed simulations were applied. These results from three-way catalysts were at least two to three orders of magnitude lower than those from the older, pelleted catalysts (as used in the USA) [9]. The main part of particles, separated in an impactor, was greater than 10 µm. In this fraction the highest platinum concentrations were measured [10]. The emitted particles consist not only of platinum. The platinum forms clusters (< 10 nm), which are positioned on the surface of particles of the carrier material (Al₂O₃) with a maximum size of 50 µm [11].

In contrast to the above mentioned values of Pt-emissions, the results of the platinum determinations in samples from the vicinity of roads in Germany and the

UK are to be discussed. Concentrations and distributions of platinum metals in near surface soil samples from along the motorway Frankfurt-Wiesbaden (Germany) were measured. The results revealed average abundances of 10 ng/g platinum and 3 ng/g ruthenium. Palladium and rhodium were below the detection limit of 2 ng/g. With increasing distance from the motorway the values gradually decreased until, after 20 m no platinum could be detected. Only the upmost soil layer, down to 20 cm contained traceable Pt concentrations [12]. In another work, concentrations of platinum and lead were measured in common grass sampled near a motorway (Stuttgart, Germany). Additionally, platinum was determined in spot checks of ash from sewage sludge incinerators. The increase in Pt levels since 1987 was traced back to releases from automobile catalytic converters. The emission rate of a car, equipped with a catalytic converter has been estimated to 4.7–9.7 µg/km [13]. This value is at least two orders of magnitude higher than that measured on a computerized motor stand [7, 8]. In the south of London (UK), platinum in roadside soils and dusts were measured. The Pt concentration ranged in soils from < 0.3 to 8 ng/g and in dusts from 0.42 to 29.8 ng/g [9]. There is a good agreement with values measured for soils in Germany (average 10 ng/g) [12] and for dusts in Germany (from 0.6 to 130 ng/g) [6]. In another German biomonitoring study [14], the accumulation of platinum was investigated in a standardized culture of grass in dependence on the distance from roads. The highest average platinum concentrations were measured at locations close to highly frequented urban roads (range 0.8 to 2.9 ng/g). The platinum concentrations at locations far from emissions was found in the range of 0.17 to 0.51 ng/g. Airborne platinum concentrations were determined in interior air of Munich city buses. The route with the highest traffic showed 33.0 pg/m³ compared to 3.0 pg/m³ at a suburban bus route. In 96 samples a mean concentration of 7.3 pg/m³ was found [15]. In a Swedish study concentrations of platinum in the < 63 µm fraction of some road sediments have been shown to increase from 3.0 ng/g in 1984 to 8.9 ng/g in 1991. Road sediments contained 39–88% more Pt than gullypot sediments and sequential extraction showed a distinct shift from predominantly inorganic Pt on the road surface to totally organic bound Pt in the gullypot. Dissolved Pt concentrations in disturbed gullypot liquor were within the range 1.7 to 3.8 ng/L and are explained by bacterial action in the gullypot sediment, mobilizing organically bound dissolved Pt species [16]. Already in 1975 Brubaker [17] investigated the biomethylation of dissolved Pt compounds by bacteria.

Engine test stand experiments for assessing platinum uptake by plants were performed by Rosner et al. [18]. Grass cultures were exposed to diluted (1:10/20) exhaust gas from an engine equipped with a three-way

catalyst for 4 weeks (average 17 ng/m^3 , 8 h/d, 5d/week). No platinum could be detected in the shoots, possibly due to the bad detection limit (2 ng/g) of the AAS determination method and the low Pt concentration in the exhaust gas.

For the investigation of Pt uptake and speciation in plants, two series of grass cultures were grown in parallel: a "native" one and one treated with platinum (tetraammineplatinum(II) nitrate, uptake exclusively by roots). In the "native" grass only one Pt-species (MW 160–200 kDa) could be isolated. The separation of the treated grass yielded in up to seven fractions Pt species in the molecular weight range from 19 kDa to 1000 kDa and one fraction below 10 kDa (containing 90% of the platinum taken up by the grass) [19, 20].

At present little is known about the metabolism of platinum and its compounds in man. Apparently soluble platinum compounds are easily absorbed and rapidly excreted through the kidneys. This is particularly true for "cis-platinum", which is used for therapeutic purposes [2]. In contrast, this does not apply when elementary platinum or surface-oxidized platinum particles are inhaled. Results of studies carried out on occupationally exposed persons indicate that these substances have very long biological half lives [5, 21]. Apart from the antitumor agent "cis-platinum" and its analogues, the toxicological relevance of platinum is confined mainly to some of its complex halide salts, which are some of the most potent sensitizers known. Symptoms of platinum-related allergy, such as rhinitis, conjunctivitis, asthma, urticaria and, occasionally, contact dermatitis, have been reported almost exclusively from occupational environments [22]. The MAK value for platinum and its compounds is $2 \mu\text{g/m}^3$ (as platinum). It is primarily intended as a protection against inhalation and skin allergies [1].

An ecological evaluation of the above reviewed investigations is only possible with the knowledge of the "natural" background concentration of platinum. The results of our work on this field is presented in Table 1. The selection of the samples was more or less accidental. Therefore, these values do not claim to be complete and statistically ensured. Nevertheless, they offer the possibility to recognize certain trends. Interesting, for example, is the series of platinum contents in uncultivated, cultivated soils and soils, sampled near motorways. The high values in the last group can be explained by immissions from motor car exhaust gases, particular as investigations of other working groups came to the same conclusion (see above). The differences between cultivated and uncultivated soils may be explained by the use of fertilizers and other technical products, like e.g. pesticides, in agriculture. These products contain (caused by their production process) platinum concentrations up to the $\mu\text{g/kg}$ range. Evidently, this platinum exists in a well bioavailable form. Thus, the astonishing high platinum content in some vegetables may be explained.

5 Enology of platinum

Because of its favourable properties, secondary metal contents in vine (*vitis vinifera*) range within narrow limits. Expositions to metal emissions may be observed by industry, traffic or mining. About 25 of vine-relevant environmental cases are reported in the literature, whereby about half of them are concerned with enological contaminations by motor cars exhaust gases. Lead is in this connection the best investigated metal, but also other metal contents are reported. For platinum no investigations are known up to now.

Various measurements have been carried out on lead contaminations of vineyard soils, grape-vine, vine leaves, bunches of grapes and wine caused by motor car exhaust gases close to high-traffic roads near Würzburg (1972), Stuttgart (1972), Freiburg (1973), Triest (1973–1990), Neapel (1974), Eger (1979) and in Macedonia. An asymptotic decrease of the lead concentration within a distance of 100 m, depending on the micro-climate and the topography, was typically observed for vineyards and vineyard soils. Wines from such sites contained excessive lead concentrations, but generally they did not reach 300 mg/l (maximum value according to wine decree) and were thus less than expected [23].

During an experiment for 1 h exhaust gases from a motor car (Alfa Romeo, petrol consumption 5 L with 1 g/L lead) were conducted into a vineyard on grape-vine and bunches of grapes. After the treatment, the lead content increased in the wine from 120 to $700 \mu\text{g/L}$, in the skins of pressed berries from 1.7 to $47 \mu\text{g/g}$ and in the yeast from 2.9 to $57 \mu\text{g/g}$ referred to dry weight. Under the same experimental conditions a significant increase of the cadmium content was observed: in wine from 1 to $20 \mu\text{g/L}$ and in yeast from 0.04 to $0.20 \mu\text{g/g}$ referred to dry weight [24].

The cadmium content of unwashed vine leaves and bunches of grapes (Traminer), grown on the side of a much frequented road near Triest (Cormons, Gorizia), showed an asymptotic decrease in relationship to the distance of the road: in leaves from 0.146 to $0.044 \mu\text{g/g}$ and in berries from 0.020 to $0.008 \mu\text{g/g}$ referred to dry weight. The ratio of "outer" (berry membrane) to "inner" (seed) Cd-contents likewise decreased as a function of the distance from the road. Similar results were obtained from the anthropogenic lead of motor car exhaust gases [25, 26]. In contrast to this, no significant effects were recognizable for the distribution of other trace elements (Al, Co, Cr, Cu, Fe, Mn, Ni and Zn) between the "outer" and "inner" parts of the berries [27].

Current results of speciation investigations are very interesting in this context. Wines from vineyards near highways contained besides "inorganic" lead also "organic" lead: trimethyl lead ($\text{Pb}(\text{CH}_3)_3$) up to $0.5 \mu\text{g/L}$ and triethyl lead ($\text{Pb}(\text{C}_2\text{H}_5)_3$) up to $0.055 \mu\text{g/L}$. The

concentrations of organic lead compounds in wines from 1950 to 1991 followed the consumption of leaded fuel. A biomethylation of inorganic lead during the vegetation cycle or during the fermentation process could not be continued. On the other hand, a triethyl lead content (0.025 µg/L) in a wine from 1950, for the present cannot be explained because in this year the wide-spread use of leaded fuel was still not customary [28, 29].

For the eco-enological judgement of platinum 14 "older" wines (vintages 1935–1982) were chosen, pressed before the wide-spread introduction of catalysts for motor cars. In comparison, 17 "younger" wines, vintages 1985–1993 (during increasing numbers of cat-vehicles), were investigated. A difference in the platinum concentrations in both groups is not recognizable. In most wines (Table 2) the Pt-content was below the detection limit (0.0005 µg/L), in 7 wines from 0.0005 to 0.0024 µg/L (mean 0.001 µg/L). So far, platinum in wine is an ultra-trace element in the lower ppt-range (ng/L).

Fermentation experiments with grape must (Nierstein, Müller Thurgau, 1994) with a "natural" Pt content of 0.0004 µg/L proved that added and co-fermented platinum (up to 200 µg/L Pt in must) was absorbed to 70 to 90% at the "Trubstoff" and was thereby almost completely concentrated in the yeast. Only a small part

Table 2 Platinum concentrations in grape must and wine

Vintage	<i>n</i>	Pt-concentration (µg/L)
<i>Grape must</i>		
1994 Nierstein Müller-Thurgau	1	0.0004
depletion by fermentation process	3	70–90%
<i>Wine</i>		
1976–1982 Rheinhessen	14	≤ 0.0005
1935 Malaga Südwein	1	0.0014
1974 Ober-Ingelheimer Portugieser	1	0.0010
1978 Niersteiner Silvaner	1	0.0011
1979 Ober-Ingelheimer Silvaner	1	0.0006
1982 Ober-Ingelheimer Portugieser	1	0.0005
1985–1993 Rheinhessen	17	≤ 0.0005
1990 Ober-Ingelheimer Spätburgunder	1	0.0024
1990 Ober-Ingelheimer Riesling	1	0.0008

of platinum traces in berries and must pass into the wine (see also Table 2).

Vineyard locations near traffic-free field paths in Ober-Ingelheim (Rheinhessen, Germany) showed in soils, vine leaves, berry stalks and berries definite lower Pt contents in comparison to vineyard locations near roads with high traffic. Even though the locations under investigation were chosen more by chance and were not sufficient for statistical evaluation, the results point

Table 3 Eco-enology of platinum (year of vegetation 1991)

Vineyard location	Soil	Leaf	Stalk	Berry	Membrane	Seed
"Natural" contents, traffic free field-paths (µg/kg)						
Ober-Ingelheim						
	(1)	0.14	0.032	0.0015		
	(2)	0.075	0.025	0.0020		
	(3)	0.21	0.037	0.0030		
	(4)		0.073	0.0010		
	(5)		0.082	0.0010		
	(6)		0.061	0.0030		
Kreis	(7)	0.26	0.045			
Kaut	(8)	0.58	0.050			
Wasserros	(9)	0.21	0.11	0.018	0.0010	
Garden W		0.25				
Garden A		0.23				
Garden St		0.29				
Mean		0.30	0.10	0.047	0.0018	
"Contaminated" contents, roads with high traffic (µg/kg)						
Nierstein						
	(1)	0.49	0.53			
	(2)	1.04	0.24			
	(3)	1.00	0.44			
Ockenheim						
	(1)		0.22	0.16	0.004	
	(2)			0.21	0.006	
	(3)			0.081	0.017	
Geisenheim						
	(1)		0.19	0.062	0.014	0.022
	(2)		0.38	0.20	0.007	
Sion, after rain						
	(1)		0.18	0.094	0.007	
	(2)		0.49	0.13	0.007	
Mean		0.84	0.33	0.13	0.009	

out increased secondary platinum contents, which may mainly be due to vehicle exhaust gases. Contributions from other sources, geological or anthropogenic, can only be supposed. Reliable results for the enology of platinum are listed in Table 3 (topography in Fig. 1). The contribution of agents for wine treatment may be assessed on the basis of their Pt content (see Table 4). Striking is the relatively high content in the "natural" product Bentonit ($0.24 \mu\text{g}/\text{kg}$) and in one active charcoal ($0.14 \mu\text{g}/\text{kg}$). The low Pt contents in pesticides and fertilizers have obviously no influence on the enology of platinum. Other fertilizers, usual on the market, had much higher Pt contents (up to $30 \mu\text{g}/\text{kg}$, see Table 1).

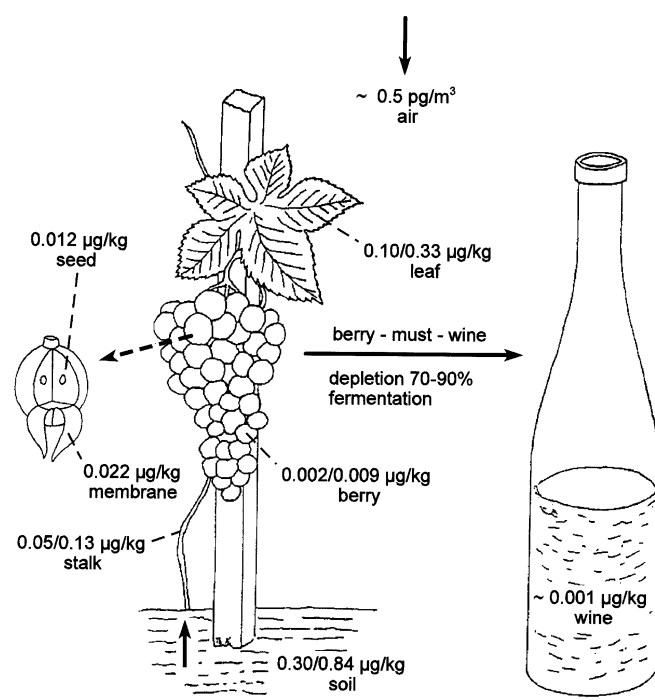


Fig. 1 Enological topography of platinum

Table 4 Platinum concentrations in agents for wine treatment

Agent	Pt-concentration ($\mu\text{g}/\text{kg}$)
Active charcoal	0.14
Chlorocarbon	0.017
Active charcoal "Peska"	0.035
Wine-eponit	0.016
Bentonit E	0.24
Bentonit M	0.24
Siliceous earth	≤ 0.02
Gelatine	≤ 0.005
Tannin I	0.014
Tannin II	≤ 0.004

6 Conclusion

As could be shown above platinum is more widespread in our environment, than to be expected from its natural abundance in the earth crust. Anthropogenic pollutions, namely the use of platinum containing products (e.g. fertilizers and therapeutics) and the immission by exhaust gases from motor cars equipped with catalytic converters, may be the sources. Model experiments proved that platinum is readily taken up by plants, and is in particular accumulated in the roots [30]. To a lower degree it is transported into the upper parts of the plants. Increased platinum concentrations, compared to control groups (which were not treated with platinum during their cultivation), could be measured in nearly all compartments (e.g. leaves, stalks, fruits). Thus platinum may enter the food chain for man. Today the load is still low, but in the longer term it will rise. As there exists no knowledge about the metabolism of platinum in biological systems, investigations in this field should be carried out. Platinum offers here a good chance for species analysis in biological materials, because of the extremely sensitive analytical methods available. Possibly, methods for platinum speciation may be developed exemplarily, which can then be transferred to the investigation of other metals.

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