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Contribution of atmospheric deposition to heavy-metal concentrations in field crops

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Abstract

The contribution of atmospheric deposition to the concentrations of Cd and Pb in four crops was studied by growing plants under field conditions, and, as a reference, in a dust-free chamber on soil labelled with Cd-109 or with Pb-210. The contribution of soil-borne Cd and Pb was distinguished from that of the deposition by measuring the isotope dilution in the plant. The 'specific activity' of the plant material grown in the field was compared with that in the dust-free environment. Atmospheric deposition contributed considerably (73-95 %) to the Pb concentrations in the leafy material of grass, spinach and carrot, and of wheat grain and straw. The contribution of deposition to the Cd concentrations of these crops was significant only in wheat grain and straw.

Keywords: atmospheric deposition, cadmium, lead, heavy metals, carrot, grass, spinach, wheat

Introduction

In most industrialized countries the heavy-metal contents of soils tend to increase gradually (Smilde, 1989; Tjell et al., 1979). This is the result of surpluses in the balance of inputs to the topsoil from deposition, fertilizers and waste products, and the output from leachates to the subsoil, and from harvested products. Atmospheric pollution, in the form of metal-containing aerosols, is a significant factor in this imbalance. If soil properties remain unchanged, the gradual increase in soil metal levels will result in a corresponding increase in plant metal concentrations. These effects can be detected only after relatively long periods of time.

Plants are also exposed directly to metal-containing aerosols and precipitation (ter Haar, 1970; Lagerwerff, 1971; Rabinowitz, 1972; Tjell et al., 1979; Harrison, 1989). Part of the metals precipitating on the plant surface is taken up, and is involved in plant metabolism (Dollard, 1986). The contribution to plant metal levels may be considerable and may exceed that of the soil (Tjell et al., 1979; Hovmand et al., 1983; Harrison & Chirgawi, 1985; Harrison & Johnston, 1987).

An elegant technique to identify the origin of plant metal is to label the soil with a radioactive metal. Radioactivity found in the plant is specific for soil-borne metal.

In this study, radioactive cadmium (Cd) and lead (Pb) were used on plants grown at a field location and in a dust-free environment.

Materials and methods

Determination of the contribution of airborne metals to metal concentrations in field crops

The contribution of the soil to heavy-metal concentrations in plants can be distinguished from that of the atmospheric deposition by growing plants on labelled soil and determining the isotope dilution in the plants. The added nuclide is equilibrated with the indigenous soil metal, by intensive mixing of the soil with a metal solution, and keeping it at least six months under moist conditions (Tjell et al., 1979; Hovmand et al., 1983; Harrison & Chirgawi, 1985, Harrison & Johnston, 1987). Specific activities of the metal are measured in soil and in plant material. The specific activity is defined as the radioactivity per unit weight of metal, expressed in becquerel per mg metal (Bq mg $^{-1}$).

The metal concentration c_p in the plant is the sum of the metal concentrations c_a and c_s derived from uptakes from the atmosphere and the soil, respectively:

$$c_p = c_a + c_s \tag{1}$$

Dividing both sides of Equation 1 by c_p shows that the fractions c_a/c_p and c_s/c_p of metal derived from the atmosphere and the soil are related by

$$c_a/c_p = 1 - c_s/c_p \tag{2}$$

To determine the ratio c_s/c_p, the soil is labelled with a radioactive isotope of the metal. Let x_p be the specific activity of the metal in the plant, and let x_a and x_s be the specific activities of the metal derived by the plant from the atmosphere and the soil, respectively. Then the radioactive metal concentration $x_p c_p$ in the plant is given by:

$$x_p c_p = x_a c_a + x_s c_s \tag{3}$$

Let x_0 be the background specific activity of the metal. Then multiplying Equation 1 by x_0, subtracting the result from Equation 3, assumming that $x_a = x_0$, and rearranging gives:

$$c_s/c_p = (x_p - x_0)/(x_s - x_0) \tag{4}$$

Introducing Equation 4 into Equation 2 gives:

$$c_a/c_p = 1 - (x_p - x_0)/(x_s - x_0) \tag{5}$$

Equation 5 has been used regularly in studies of uptake of metals (Tjell et al., 1979; Hovmand et al., 1983; Harrison & Chirgawi, 1985, 1988). Determining the specific activity ($x_s - x_0$) of the metal taken up by the plant from the soil on the basis of soil chemical analysis turned out to be rather difficult.

Growing plants on the same labelled soil in a dust-free environment provides an alternative direct means of determining the specific activity. Let a superscript * denote quantities associated with uptake in such a dust-free environment. Then, analogous to Equations 1 and 4:

$$c^*_p = c^*_s \tag{6}$$

$$c^*_s/c^*_p = (x^*_p - x_0)/(x^*_s - x_0) \tag{7}$$

Substituting Equation 6 in Equation 7 and assuming that the specific activity ($x^*_s - x_0$) of the metal taken up from the soil in the dust-free environment is equal to the specific activity ($x_s - x_0$) of the metal taken up from the same soil in the outdoor environment, then Equation 7 reduces to:

$$(x_s - x_0) = (x^*_p - x_0) \tag{8}$$

Introducing Equation 8 into Equation 5 gives:

$$c_a/c_p = 1 - (x_p - x_0)/(x^*_p - x_0) \tag{9}$$

According to Equation 9 the relative contribution c_a/c_p of airborne metal can be calculated on the basis of measurements of specific activities ($x_p - x_0$) and ($x^*_p - x_0$) of plants grown on the same labelled soil in the outdoor and dust-free environments, respectively. Based on these considerations experiments were performed to determine the contribution of airborne metals to metal concentrations in field crops.

Soils

Two sandy loam soils were selected, low in Cd and Pb and with a neutral reaction. The main properties are given in Table 1. The soils were passed through a 2-mm titanium sieve before subsampling. Carrier-free Cd-109 and Pb-210 were, dissolved

Table 1. Properties of the two sandy loam soils.

	General characteristics						Cadmium		Lead	
	pH	org.mat.	$CaCO_3$	<2μm	<16μm	<50μm	mg kg^{-1}	MBq mg^{-1}	mg kg^{-1}	MBq mg^{-1}
Soil 1	7.8	0.88 %	2.1 %	3.5 %	4.2 %	14.4 %	0.16	2.4	4.7	313.0
Soil 2	6.8	1.74 %	0.1 %	6.4 %	12.3 %	22.9 %	0.29	2.0	20.1	143.6

in a slightly acidified solution, added to a slurry of the soil, and thoroughly mixed in an epoxy-coated cement mixer. After drying to field capacity the soils were equilibrated under moist conditions for six months. The labelled soils were placed on top of a gravel layer in polyethylene containers. The soil surfaces (8.6 dm^2) of the pots were covered with a layer of polyethylene granules. This prevented contamination of the plants due to splashing of soil particles during rain. Excess rainwater was removed via a tube positioned in the gravel layer at the bottom of the container.

Plants

Plant species were selected differing in leaf surface and in edible organs (leaves, seeds, tubers): grass (*Lolium multiflorum* Lam., Italian ryegrass), spinach (*Spinacia oleracea* L. cv. Mazurka), spring wheat (*Triticum aestivum* L. cv. Adonis) and carrots (*Daucus carota* L. cv. Cornet RZ). Grass was grown on soil 1 in 1984 and 1985. Spinach, carrots and spring wheat were grown on soil 2 in 1986 and 1987.

Cultivation

Each experiment consisted of two sets of six containers: two untreated soils, two soils labelled with Cd-109, and two soils labelled with Pb-210. One set of containers was placed outside in a lawn with the rims level with the surrounding grass plot. The containers were exposed to wet and dry deposition. The second set was placed in a dust-free growth chamber (DGC) in a greenhouse.

Fertilization, watering, and pest control were performed according to the standards for the relevant crops. Metal contamination was avoided by using high purity reagents and metal-free equipment.

The dust-free growth chamber is similar to open-top chambers used in air pollution studies (Buckenham et al., 1981). Ambient air was supplied through a prefilter/HEPA filter system that excluded 99.999 % of particles > 0.3 μm. The growth chamber (floor surface 4.4. m^2) was made of perspex. The 'top' of the chamber was covered with a perspex lid; air could escape from the room through a 7-cm slit between cover and walls. The levels of Cd and Pb in the air of the DGC were low, but not negligible: 0.02 ng m^{-3} and 0.29 ng m^{-3}, respectively. This was probably caused by some leakage of unfiltered air through the service door and through the exit slit. Also some transport of extremely small metal-containing particles through the HEPA-filter might have occurred (Lee et al., 1968).

Sampling procedure and sample treatment

Utmost care was taken to avoid sample contamination during handling of the samples. Grass was harvested fortnightly, spinach, spring wheat and carrots once, at maturity. Plant material was oven-dried and ground; it was not rinsed because it was found that the results were not affected by rinsing the leaf material with distilled water. Carrot roots were freed from soil particles and rinsed with a detergent solution (T-pol).

Sampling of air and bulk atmospheric deposition

Bulk deposition was collected throughout the experimental periods for determination of Cd and Pb, using standardized open precipitation collectors, as used at the time by the National Precipitation Network (RIVM, 1984-1987). Particulate Cd and Pb in air were collected with a dust-collection apparatus (Sartorius), fitted with a membrane filter with a maximum pore diameter of 0.3 μm.

Analytical methods

Macroconstituents in soils
Macroconstituents (particle size distribution, organic matter, calcium carbonate, pH in 1M KCL-extract) were determined by standard analytical methods.

Total and extractable Cd and Pb contents of soils
Total Cd and Pb were obtained by digesting the soil five times with concentrated HNO_3 (Balraadjsing, 1974). Four soil extractants (Andersson, 1975) were used, namely:
— 1 M NH_4-acetate, pH 7.0,
— 1 M NH_4-acetate, pH 4.8,
— 0.025 M EDTA, pH 7.0,
— 2 M HNO_3.
The dry soil sample (15 g) was shaken for 1 h with 50 ml of the extractant, and separated from the liquid fraction by centrifugation and filtration. Cd and Pb were determined by flameless atomic absorption spectrometry, after solvent-extraction at pH 3.5 by methyl isobutyl ketone (MIBK) with a mixture of 1 % ammonium pyrrolidindithiocarbamate (APDC) and sodium diethyldithiocarbamate (DDC) solution in ethanol as a complex former and subsequently back-extraction by 2 M HNO_3. The EDTA-extract was digested with HNO_3 before liquid-liquid extraction. Standard additions were made if required.

Total Cd and Pb contents of plants
Plant material was digested by boiling it with concentrated HNO_3. After addition of diluted HCI and refluxing during one hour at 100 °C the digest was filtered. Cd and Pb were determined by flameless atomic absorption spectrometry, after liquid-liquid extraction as described.

Radioactivity in soils and plants
Pb-210 and Cd-109 activities were counted by gamma-spectrometry in the solutions obtained by digestion or extraction of soils and plants. When activities were low, a higher count rate was obtained for the Pb-210 activity by using the 1.14 MeV beta from Bi-210, the daughter activity from Pb-210 (Karamanos et al., 1975). Bi-210 grows in completely after 50 days and its Cerenkov light can be counted in a liquid scintillation counter.

Results

Distribution of labelled Cd and Pb over soil fractions

The distribution of total and radioactive Cd and Pb over the soil fractions was determined by extracting the labelled and equilibrated soils with various extractants, differing in strength. If the labelled metals would be uniformly distributed over the binding forms of the soil, the specific activities of the metals in all fractions would be the same. The results of the experiments (soils 1 and 2 combined) show that the specific activities of the metals decreased when a larger proportion of the soil metal was extracted (Fig. 1). Therefore, Equation 5 cannot be used to calculate the contribution of the atmospheric deposition to plant metal concentrations.

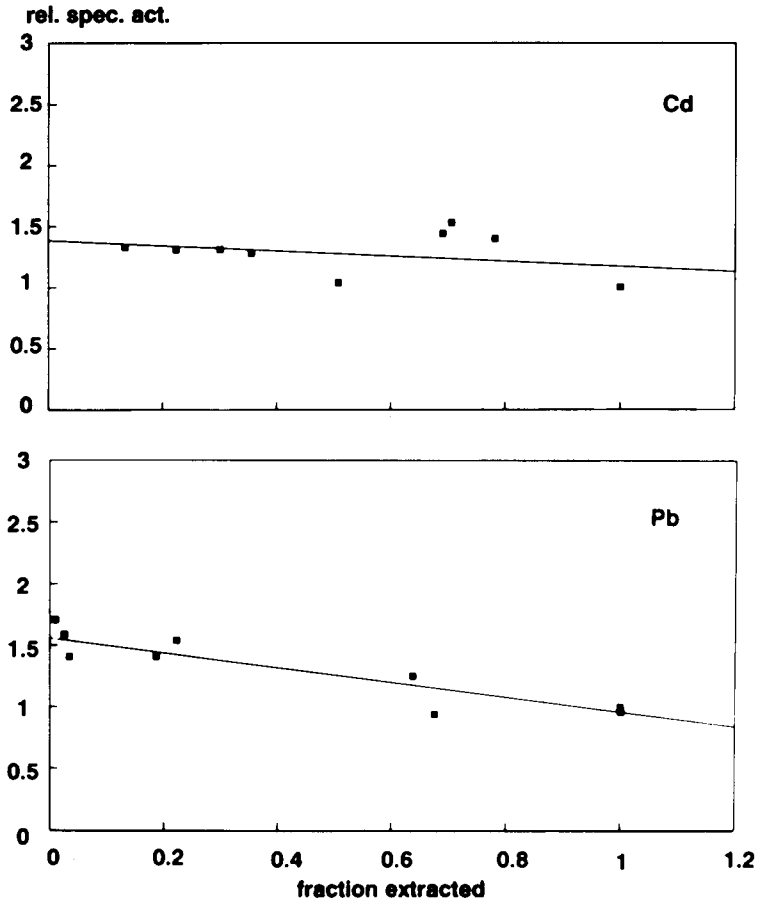

Fig. 1. Relative specific activity of Cd-109 (top) and Pb-210 (bottom) in soil extracts. (Relative specific activity of total soil Cd or Pb = 1.)

Contribution of airborne metal to plant metal concentrations

The quantities of Cd and Pb supplied to the experimental plot were collected in open bulk-deposition collectors and measured. The results were compared with the average quantities in the Netherlands (Table 2). In the last experimental year (1987), both Cd and Pb deposition levels at the experimental plot were higher than the average levels in the country, possibly because of building activities in the neighbourhood. The annual supply of Cd to the soil was of the same order of magnitude as the net annual removal by agronomic crops on arable land, estimated to be 1.4 g ha^{-1} (Smilde, 1989). The supply of Pb to the soil was much higher than the removal of 1.5 g Pb ha^{-1} by crops. The airborne Cd supplied during the growing period was lower in all investigated crops than the Cd content of these crops. This is in contrast to Pb, where the airborne Pb supply exceeded the contents of the crops. The quantities of Cd and Pb that were actually intercepted by the experimental plants were estimated by means of the isotope dilution technique.

Cd experiments

The results of the plant experiments in the field and under dust-free growing conditions are summarized in Table 3.

The (total) Cd concentrations of the plants grown in the dust-free chamber were equal to (spring wheat) or higher than (other species) those of the field-grown plants, whereas the dry-matter yields in the DGC were equal (spring wheat) or lower (other species). The higher plant Cd concentrations in the DGC might be due to a reversed dilution effect, and environmental conditions could also be involved. Soil type, treatment, planting/sowing time and period of cultivation were exactly the same, but in the DGC both air and soil temperature were higher than those under field conditions. The higher wind velocity in the DGC led to a higher rate of plant transpiration, which may have caused a greater convective transport to the soil/root interface. The higher plant Cd concentrations illustrate the strong impact of growing conditions on Cd uptake.

Table 2. Cd and Pb in wet deposition and in air.

	Cd and Pb in wet deposition ($g\ ha^{-1}\ yr^{-1}$)				Cd and Pb in air ($ng\ m^{-3}$)					
	Netherlands[1]		Haren		Netherlands[1]		Haren		DGC[2]	
	Cd	Pb	Cd	Pb	Cd	Pb	Cd	Pb	Cd	Pb
1984	1.9	128	1.7	74	2	177	1.0	107	0.09	0.63
1985	2.0	122	1.9	82	-	168	4.0	26	0.02	1.27
1986	1.5	93	1.6	50	7	126	0.3	32	0.003	0.00
1987	1.6	85	2.1	127	7	100	0.5	25	0.00	0.02

[1] Average in the Netherlands (RIVM, 1984-1987).
[2] DGC = dust-free growth chamber (incidental measurements).

Table 3. Concentrations and specific activities of Cd in soil and in plants, grown in the field and in a dust-free growth chamber (DGC).

	Field conditions				Dust-free conditions			
	yield[1] (kg m^{-2})	conc. (mg kg^{-1})	spec. act. (MBq mg^{-1})	SD[2] (MBq mg^{-1})	yield[1] (kg m^{-2})	conc. (mg kg^{-1})	spec. act. (MBq mg^{-1})	SD[2] (MBq mg^{-1})
Soil 1		0.158	2.44	0.48		0.158	2.44	0.48
Grass 1984	3.35	0.170	3.47	0.05	1.86	0.406	3.74	0.05
Grass 1985	1.78	0.162	3.42	0.05	1.57	0.226	3.32	0.05
Soil 2		0.287	1.98	0.05		0.287	1.98	0.05
Spinach 1986	0.37	1.150	3.27	0.18	0.22	1.48	3.18	0.23
Carrot roots 1986	2.52	0.206	2.60	0.22	2.14	0.238	2.29	0.31
Carrot leaves 1986	0.95	0.344	2.52	0.11	0.85	0.428	2.48	0.16
Wheat grain 1987	1.62	0.106	1.91	0.16	1.78	0.089	2.43	0.20
Wheat straw 1987	1.60	1.163	1.21	0.08	1.72	0.094	2.34	0.11

[1] On dry-matter basis.
[2] Standard deviation.

Comparison of the specific activities (Table 3) of plant Cd shows that for grass, carrots and spinach no significant contribution of airborne Cd could be demonstrated (Table 4). This contribution was negative and resulted from higher specific activities observed in some field plants as compared with the dust-free plants. The supply of Cd via atmospheric deposition was small compared with the amount taken up from the soil. There was a significant contribution only in spring wheat, both for straw and grain. This means that not only metal aerosols adhering to the leaf surface

Table 4. Soil- and air-borne Cd and Pb in crops.

	Cd					Pb				
	n[1]	soil-borne (μg m^{-2})	air-borne (μg m^{-2})	SD[2] (μg m^{-2})	air-borne (%)	n[1]	soil-borne (μg m^{-2})	air-borne (μg m^{-2})	SD[2] (μg m^{-2})	air-borne (%)
Grass 1984	66	282	22	6	7.3	66	124	2521	3	95
Grass 1985	36	155	−5	4	−2.9	36	144	1532	2	91
Spinach 1986	12	209	−6	20	−2.8	6	77	212	29	73
Carrot roots 1986	8	249	−33	45	−13.2	8	127	8	19	5.7
Carrot leaves 1986	12	168	−3	14	−1.7	12	32	1254	7	98
Wheat grain 1987	4	64	18	8	21.4	4	0	337	53	100
Wheat straw 1987	4	67	63	5	48.4	4	3	1032	2	99.7

[1] Number of observations.
[2] Standard deviation.

contributed to plant Cd but also metal absorbed by the leaves and translocated to the seed. The spring wheat experiments demonstrate that a much longer exposure to atmospheric deposition can lead to a significant contribution of aerosol Cd to plant Cd.

Pb experiments

The results are summarized in Table 5. The (total) Pb content of the plant material grown in the field was markedly higher than that of the plants grown under dust-free conditions. In the case of Cd, the differences in growing conditions caused higher Cd concentrations in plants in dust-free conditions; in the case of Pb they were masked by the strong effects of airborne Pb and by the low uptake of soil Pb. The differences in specific activities of Pb in plants grown under field conditions and in the chamber demonstrate the dominant effect of airborne Pb. The contribution of airborne Pb, calculated with Equation 9, amounts to over 90 % for grass, carrot leaves, and wheat grain and straw (Table 4). Lower contributions were calculated for spinach (73 %), and carrot roots (5.7 %) (Table 4). The lower contribution for spinach can be attributed to the short growing period, those for carrot roots to the indirect supply of airborne Pb to the organs. Table 5 also shows that in most plants grown under dust-free conditions the specific activities were equal or lower than those of total soil-Pb. This suggests that the residual Pb contamination of the air of the DGC had a significant effect on plant Pb concentrations, leading to a small underestimation of the contribution of airborne Pb.

Table 5. Concentrations and specific activities of Pb in soil and in plants grown in the field and in a dust-free growth chamber (DGC).

	Field conditions				Dust-free conditions			
	yield[1] (kg m^{-2})	conc. (mg kg^{-1})	spec. act. (KBq mg^{-1})	SD[2] (KBq mg^{-1})	yield[1] (kg m^{-2})	conc. (mg kg^{-1})	spec. act. (KBq mg^{-1})	SD[2] (KBq mg^{-1})
Soil 1		4.69	313.02	89.91		4.69	313.02	89.91
Grass 1984	3.78	1.48	1.48	0.01	2.28	0.120	31.38	0.22
Grass 1985	2.07	1.73	1.30	0.01	1.90	0.093	15.13	0.11
Soil 2		20.10	143.56	5.74		20.1	143.56	5.74
Spinach 1986	0.33	1.59	3.48	0.62	0.13	0.12	13.10	4.26
Carrot roots 1986	2.35	0.11	134.31	9.40	2.33	0.115	142.45	19.37
Carrot leaves 1986	1.13	2.60	3.00	0.49	1.13	0.135	119.88	10.91
Wheat grain 1987	1.51	0.43	d.l.[3]	-	2.08	0.013	d.l.[3]	-
Wheat straw 1987	1.62	1.30	0.15	0.02	1.85	0.037	44.40	29.30

[1] On dry-matter basis.
[2] SD = standard deviation.
[3] d.l. = below detection limit.

Discussion

Various techniques have been used to estimate the contribution of soil and atmosphere to the metal content in field crops. Tjell et al. (1979), Hovmand et al. (1983) and Harrison & Johnston (1987) used the radioactive tracer technique mentioned before, based on Equation 5. Our study demonstrates that the relationship between the specific activities of soil and plant cannot always be used to estimate the content of soil-borne metals in plants. Harrison & Johnston (1987) used two other techniques to determine the relative contributions of atmosphere and soils to the Pb content of crops:

— The dual-compartment chamber technique, in which plants are cultivated in filtered air or in unfiltered, ambient air. This technique provides specific information on the contribution of dry deposition.

— A technique in which plants are grown in a common soil at field sites differing in atmospheric Pb deposition levels. The local atmospheric deposition is monitored with moss-bag collectors. Soilborne Pb is estimated by extrapolation of plant Pb levels to zero deposition.

The moss-bag collector probably better monitors plant-related Pb deposition than bulk deposition, but the differences between plant species in intercepting atmospheric Pb are not accounted for. Moreover, because of inevitable differences in growing conditions an error is introduced leading to a large bias in the conclusions. The authors conclude that the contribution of particulate Pb in air (dry deposition) is much larger than that of wet deposition.

The technique used in our study, in which specific activities of plants grown in the field and under dust-free conditions are compared, does not require estimation of a plant-available fraction in soil, and is not sensitive to differences in environmental growing conditions.

The results show a significant contribution of atmospheric deposition to crop Pb concentrations, and, for spring wheat, also to crop Cd concentrations. For Pb, these conclusions agree with the results of other studies (Tjell et al., 1979; Hovmand et al., 1983). For a proper comparison, soil conditions (metal levels and availability) as well as deposition rates have to be considered. Hovmand et al. (1983) and Tjell et al. (1979) used sandy loam and sandy soils with a moderate pH ($CaCl_2$) of 6.3 and 4.6, and relatively low Cd (0.08 and 0.09 mg kg^{-1}) and Pb (14.4 and 11.1 mg kg^{-1}) levels, combined with relatively low Cd and Pb deposition levels. Under these conditions the contributions of soilborne Cd and Pb are low, and the relative effect of airborne metals is high. In our experiments, soil Cd and Pb levels and pH were higher and the deposition is of the same order of magnitude, also leading to low Cd and Pb levels in the plants. In our studies, corresponding Cd levels were found for wheat grain and straw, but no significant contribution of airborne Cd could be detected in grass and carrot roots and leaves as were found by Hovmand et al. (1983), who observed contributions of airborne Cd of up to 52 %

The results of Harrison et al. (1987) on Pb cannot be compared with our results, as no information is given on the bulk deposition rate.

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