ATMOSPHERIC INPUT OF CONTAMINANTS TO A SMALL MEDITERRANEAN BASIN.

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Abstract

Rainwater chemistry was monitored during a two years period (Nov. 2003 – Oct. 2005) for quantification of the atmospheric input over a small (70 km2) rural Mediterranean watershed. A refrigerated wet only precipitation collector with a nearby bulk sampler were installed at one station. Major ions contents (HCO3,Cl, NO3, SO4, Ca, Mg, Na, and K) were determined by Ion Chromatography, nutrients content (PO43-, NO3-,NO2- and NH4+) determined following APHA standard and trace element contents (Al, B, Ba, Cd, Cr, Cu, Co, Li, Mn, Mo, Ni, Pb, Rb, Sr, U, V and Zn) by ICP-MS. During the observation period, a total of 36 rain samples were collected and analysed for wet and dry deposits determination.
The dataset obtained from the field study is presented and discussed. The primary objectives are i) to evaluate the rainwater concentration, and ii) to estimate the annual atmospheric input to the basin.
Keywords. precipitations, nutrient deposition, wet and bulk deposition, deposition rate.

Introduction

The atmosphere disseminates and deposes at the ground numerous substances which have a notable geological, geochemical and ecological impact (Loye-Pilot, 1995). In the Mediterranean area, anthropogenic and natural sources are both contributing to the atmospheric deposit. Pollutions due to human activities are originating mainly from Europe (Bergametti et al. 1989; Gullu et al., 1998; and Tuncer et al., 2001) and are related to industrial / agricultural activities or traffic emissions. Natural deposition is mainly originating from pulses of mineral dust from North Africa into the Mediterranean region (Bergametti et al., 1992, Kubilay and Saydam, 1995, Migon and Sandroni, 1999 and Herut et al., 1999); and from marine aerosols delivered by the Mediterranean sea.
Mediterranean coastal waters as interfaces between the sea and the continent are receiving pollutants from various inputs: atmospheric, sea, riverine (e.g. Tournoud et al., 2006). Many studies have focused on the effects of river inputs on water quality degradation in coastal lagoons. Nutrient budget for the coastal waters have indicated that the flood loads can be critical for the equilibrium of the system. But little is known about atmospheric contribution. Observations of rainfall chemical concentration over the Mediterranean area have emphasized the importance of atmospheric deposition. Nevertheless, very few information can be gathered from the literature for a precise quantitative estimate of the atmospheric contribution to basin budget. Little is known on the magnitude and trends of atmospheric deposition to small coastal Mediterranean basins. At the basin scale many pollutant budget have been calculated on the basis of a few extrapolated literature values for the atmospheric compound. Studies conducted in the Vène basin have focused on river inputs. The

atmospheric contribution has been estimated from surrounding average rain concentration but on site data were not available.
The aim of this study is to estimate wet deposition of major elements and nutrients into the basin and the Thau pound based on data from samples collected on the basin area from October 2003 to September 2005.

Material and methods

Samples were collected at a sample station located in a small Mediterranean coastal basin, the Vène basin (Fig. 1). The basin is located in the French Mediterranean coast, north east to the Thau lagoon. The major cities around are the city of Montpellier and Sète, respectively 400 000 and 44 000 inhabitants. The area is free from major industrial activities –as is most of the region- the closest one being around Marseille, more than 100 km to the east (Luck and Ben Othman, 2002). There are a few industries southeast to the basin located close to Balaruc town that locally impact to nitrogen concentrations in groundwater (Tournoud et al., 2006). The Vène basin area is 67 km^2 that ranges from 2 to 323 m in elevation. The basin is divided in two main zones: i) the central part of the basin, which covers about 40% of the total catchment area, is a flat marly plain dedicated to agricultural activities (essentially vineyards: 21% of the total basin area); ii) on both sides, limestone massifs, highly karstified, are covered by natural garrigue used for sheep and poultry. Two highly traffic roads cross the basin on its south, the south France highway and a secondary road. Human activities are concentrated in the middle basin part. About 9 200 people overall live in three small villages. The Vène feeds the Thau lagoon. The large and brackish Thau lagoon is devoted to recreational activities and shellfish farming.

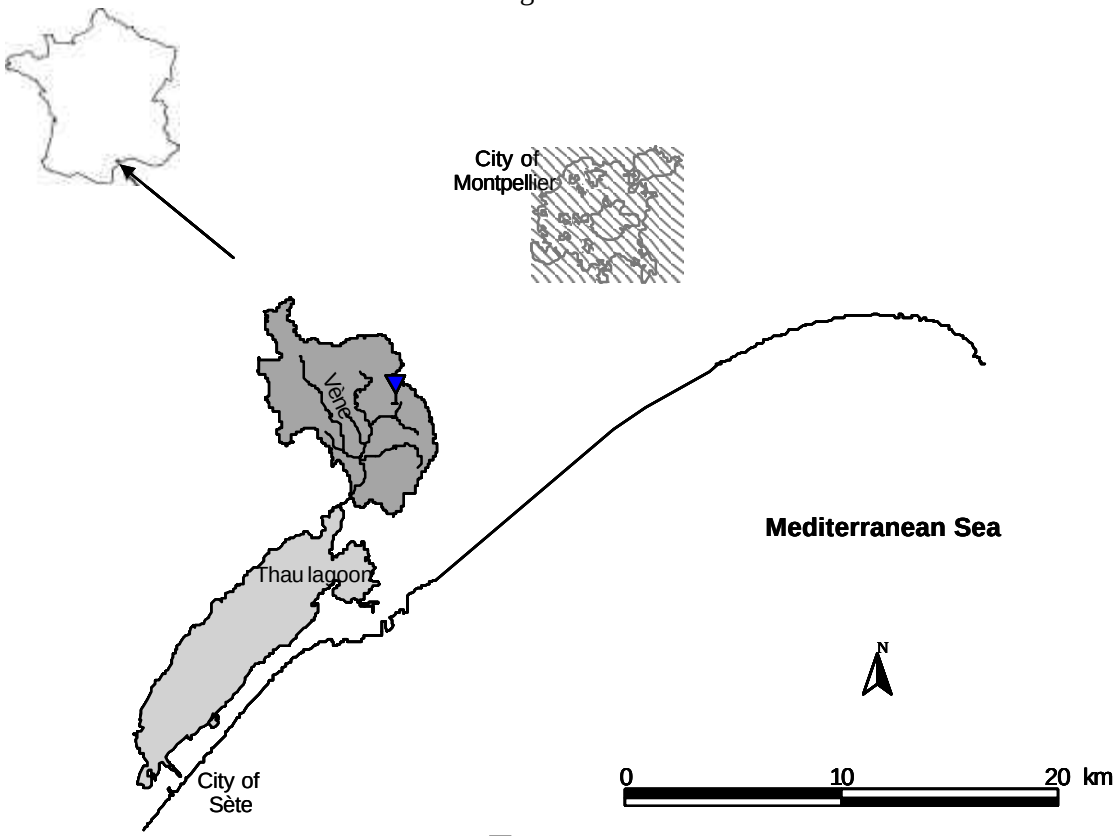

Figure 1 : Location of the study area. (▼) mark indicates the location of the bulk and wet only deposition stations.

The rain sampling station was located accordingly to the already installed pluviograph stations. At the Plagnol site located at midway of the basin two samplers were installed : a refrigerated automatic wet-only precipitation collector (Eingenbrodt NSA 181 KS collector) with a nearby bulk sampler. Plagnol site is located close to a small sewage treatment plant in a natural site. The minimal distance to the secondary road is 1 km and 4 km to the highway. Bulk precipitation was collected by a conventional tipping bucket rain gauge. It consists of a 35,6 cm diameter polyethylene funnel connected to a blinded 15 l polyethylene bucket. The polyethylene bucket was wrapped with black plastic bag to avoid light exposition of the samples. Samples were collected over two years, from November 2003 to October 2005. Wet and bulk samples were collected on rain event basis, no later than 48 h after the rain ends. Conjointly to sampling, rainfall amounts were continuously measured by the tipping buckets gauges with 0.2 mm accuracy.

The climate is characterized by a marked contrast between dry and humid conditions. Precipitation occurs mainly during autumn and spring. Rainfall is usually short, intense, heavy and irregularly spatially distributed. According to Ascencio (1984) the mean annual rainfall over the basin is close to 700 mm. Grillot (2006) analysing 1994 to 2005 data at the rain gauge station used in this study observed that annual rainfall range from 472 mm to 859 mm and over this period the mean inter annual rainfall is 668 mm marked by a high standard deviation (i.e. 121 mm).

Rain water samples were analysed for major element contents (HCO3, Cl, NO3, SO4, Ca, Mg,Na, and K) by Ion Chromatography and nutrient contents : total phosphorus (TP), and total nitrogen (TN) concentration were estimated following Standard Methods procedures (APHA, 1992).

Results and discussion

Rainfall.

Annual rainfall varied markedly between 2003-04 and 2004-05. The second year annual rainfall, 397 mm is less than half the first year observation, 836 mm. Monthly precipitation at the sampling site is shown Figure 2. As the region is characterized by a Mediterranean climate, the rainy periods are mainly occurring during autumn and spring.

At the Plagnol site 13 and 23 samples from respectively wet-only and bulk deposition were analysed for major elements, nutrients and trace element content. (Trace element contents are not discussed in this study). They account for 35% (wet only) and 78% (bulk) of the two years total rain amount. That is broadly a half of the actual rain events that were sampled. Some rain events are missing due to insufficient sampled volume, sampler dysfunction, eliminated contaminated samples or lost samples.

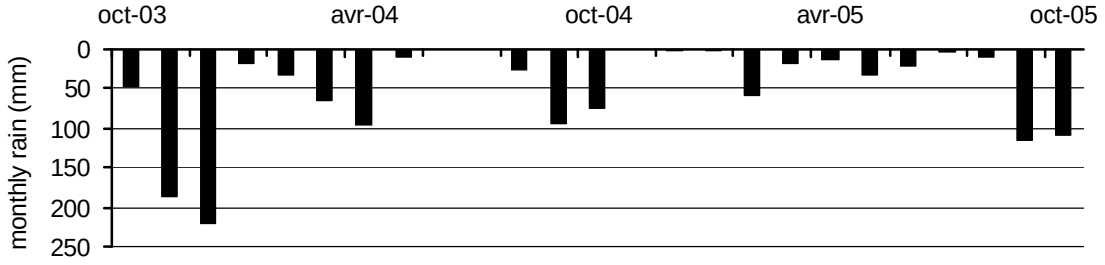

Figure 2: Monthly rainfall measured by the typing buckets rain gauge during the period of observation.

For wet-only precipitation at Plagnol site, sample concentrations, with for the whole observation period, minimum, maximum, 1st quartile, median, 3rd quartile are reported on Table 1. In order to normalise the average between light rains, having relatively high concentrations, and heavy rains in which the constituent are diluted we consider the volume weighted mean concentration (VWM). A total of 468 mm of rain were sampled. The sampled events rain depths are varying from 2.2 to 97.4 mm.

Table 1: Range, median, 1st quartile, 3rd quartile and volume-weighted-mean concentration in individual rain event during Nov. 2003 – Oct. 2005.

Parameter	Min	Max	Median	1st quartile	3rd quartile	VWM
Rain depth (mm)	2.2	97.4	28.5	-	-	-
TN (µg-N / l)	2.2	97.4	28.5	-	-	-
TP (µg-N / l)	490	3639	938	572	1148	901
HCO_3 (mg / l)	0.1	36	2	0.8	4.3	3
Cl (mg / l)	0	15.1	6.55	3.51	7,90	4.77
NO_3 (mg / l)	0.05	15.03	2.62	0.6371	5.5217	3.21
SO_4 (mg / l)	0.97	6.98	1.95	1.8	2.89	2.21
Ca (mg / l)	0.7	4.18	1.69	1.08	2.22	1.7
Mg (mg / l)	0.49	5.3	2.26	0.61	3.14	1.79
Na(mg / l)	0.02	1.01	0.17	0.06	0.22	0.16
K (mg / l)	0.23	8.13	0.82	0.4	1.57	1.6

Major ions concentrations in wet deposition.

The weighted mean concentrations of anion can be ordered in a descending order as follows: $HCO_3^- > Cl^- > NO_3^- > SO_4^{--}$. The weighted mean values of these anions were obtained as 4.77, 3.21, 2.21 and 1.70 mg / l respectively. Mean concentrations of cations ($Ca^{2+} > Na^+ > K^+ > Mg^{2+}$) were found to be 1.79, 1.57, 0.30 and 0.16 mg / l repectively. Similar dominance were observed in other studies carried out in the Mediterranean basin (e.g. Avila and Alarcon, 1999, Samara et al. 1992, Al-Momami et al., 2000 …). The ion concentration observed are in good agreement with other studies carried out in the Mediterranean basin (Hontorio et al. , 2003). The anion and cation ordering is quite stable from event to event but range of concentration vary from 1 to 3 or 4 orders of magnitude.

Nitrogen and phosphorus concentrations in precipitation.

Absolute levels of concentrations vary over one to seven orders of magnitude and from one to fifteen for TN and TP respectively.
TN concentration ranges from 0.5 to 3.6 mg-N l^{-1}. The VWM is 0.9 mg-N l^{-1} and half of the observation are in the range 0.6 to 1.1 mg -N l^{-1}. The highest concentration, i.e. 3.6 mg-N l^{-1}, was observed during a light rain event which was probably of marine and saharan origin. The concentration level of nitrate NO_3 (VWM concentration equals 366 µg-N.l-1), at the Plagnol site, is comparable and within the range of concentration observed at several locations in the Mediterranean area. Ladouche et al. (1998) at stations located a few kilometres from our basin reported average concentration from 339 to 813 µg-N.l^{-1} for NO_3. TP ranges from 3 to 49 µg-P l^{-1}. The volume weighted mean concentration is 11 µg-P l^{-1} for TP. Half of the sample concentrations are in the range 7 to 25 µg-P l^{-1}. Theses observations match values reported by other studies in the surrounding area. As example using data reported by Migon and Sandroni (1999) obtained on the Cap Ferrat (Southeastern coast of

France), we derived a VWM concentration equals to 10.8 µg-P l^{-1}. At Villefranche sur Mer (south of France), Klein et al. (1997) sampled a few rain events and determined $PO4^{3-}$ concentration. According to the classification of Loÿe-Pilot and Morelli (1990) they identified northern sector plus Saharan polluted events containing 2 µg-P $l^{-1}$and eastern sector plus Saharan polluted events revealing 140 µg-P l^{-1} concentration. Compared to other area over the world, TP is an order higher than volume-weighted P concentrations observed in Florida : 1.3 µg-P l^{-1} (Grimshaw and Dolske, 2002) but less than concentration of collected rainfall near Lake Ontario by Chan and Kuntz (1982), i.e. they found annual average rainfall TP of 20-300 µg.L^{-1} and less than average rainfall TP concentration of 44 µg.l^{-1} observed near Lake Taihu (China) by Luo et al. (2007). The phosphorus concentration is highly variable with time. Dissolved and particular phosphorus repartitions differ from event to event. For most of the rain events, the particular phase is majority but it happens that all the phosphorus if of dissolved form (e.g. 03/05/2004 and 13/10/2004 samples).
Rain total suspended solid concentration ranges from 0.1 (i.e. the detection level) to 36 mg.l^{-1}. The VWM concentration is 3 mg.l^{-1}. But the standard deviation is very high, 12 mg.l^{-1}. Actually, two ranges of values are observed, high values (more than 15 mg.l^{-1}) that could be associated to Saharan and/or anthropogenic atmospheric contributions and low level concentrations from 1 to 4 mg.l^{-1}. As a comparison, Muoghalu and Johnson (2000) observed total suspended sediment in Nigerian rain from 6 to 50 mg.l^{-1} The average equals 16.5 mg.l^{-1} and the standard deviation is 13.5 mg.l^{-1}.

Bulk precipitaion concentration.

Bulk precipitation was collected for the same period at the Plagnol site. Bulk concentrations are an order higher than wet concentration. Anion VWM concentration are 7.82, 4.70, 2.38 and 3.07 mg/l for respectively HCO_3^-, Cl^- , NO_3^- and SO_4^{--} . Cation VWM concentration are 3.67, 2.38, 0.35 and 0.28 mg/l for respectively Ca^{2+}, Na^+, K^+ and Mg^{2+}. For nutrients, TP content is in the range 8 to 274 µg-P l^{-1} and TN content ranges from 620 to 4860 µg-N l^{-1}. These values are only indicatives. Actually bulk precipitation concentration is highly dependent on the exposition duration versus the rain depth, duration which are relevant to the dry and wet contribution. Therefore, deposition rate is more pertinent for intercomparison.

Deposition rates
The deposition rate represents the mass of the constituant that reach the ground per unit surface and per unit time. We will consider wet, bulk and dry deposition. At the rain event time scale, wet deposition rate is derived from the product of rain constituent concentration and rain depth divided by the exposition duration:

$$D_w = C\ h\ /\ d_e \qquad (1)$$

where D_w (mg m^{-2} d^{-1}) if the concentration C (mg l^{-1}), the rain depth h (mm) and the exposition duration d_e (days). The bulk deposition rate, D_b, is estimated in a similar way.

$$D_b = C_b\ h\ /\ d_e \qquad (2)$$

D_b units are (mg m^{-2} d^{-1}), if the concentration of the bulk sample C_b (mg l^{-1}), the rain depth h (mm).
The dry deposition rate, D_d can be estimated from bulk and wet deposition using the following formula :

$$D_d = (D_b - D_w)\ /\ (d_e - d) \qquad (3)$$

where d (days) is duration of the rain events that occurred during the exposition period.

At the rain event time scale, wet and bulk depositions are highly fluctuant. Maximum rate deposition can be two to three orders higher than minimum deposition. For a better intercomparison, of D_w and D_b and an average estimation of D_d we consider the two largest continuous exposition periods, i.e. from 24th of February 2004 to 3rd May 2004 (62 days) and from 13th September 2004 to 13 December 2004 (53 days). For each of these two periods five rain events occurred and were sampled.

The measured deposition rates for major ions, phosphorus and nitrogen are reported table 2. The deposition observed during this two periods can be extrapolated at the year time scale. Annual wet deposition rate can be estimated assuming that period 1 and period 2 are representative of the year. From these two periods, the rain specific wet deposition rates, Ds_w, (i.e. total deposition divided by the rain depth) are estimated. The annual wet deposition is then derived from the mean annual rainfall multiplied by Ds_w. Another way to estimate, the annual rain specific wet deposition rate is by considering the VWM concentration of the wet samples. This second approach gives values in the same order of magnitude than the first (see table 2). Considering the mean inter annual rainfall is 668 mm (Grillot, 2006), the annual wet deposition rates could be estimated and are reported table 2. The annual dry deposition rate estimations are extrapolated dry deposition from period #1 and from period #2. Therefore, the bulk deposition rate can be estimated as the sum of dry and wet. At the annual scale, wet and dry contribution to TN deposition are comparable but for TP the dry deposition is four to six more than the wet deposition.

The TP wet deposition rate, 6.6 +/- 1.0 mg-P m^{-2} y^{-1} is slightly larger than that of Migon and Sandroni (1999) in a site eastern of our location : Cap Ferrat, 5.11 mg-P m^{-2} y^{-1} and in the same order of the southeastern Mediterranean observations reported by Herut and Kron (1996) : 6.7 mg-P m^{-2} y^{-1}. But the range of deposition is half of the mean wet deposition estimated over USA, i.e. 20 mg-P m^{-2} y^{-1}. The bulk deposition rate, 39+/- 15 mg-P m^{-2} y^{-1} agrees with 40 mg-P m^{-2} y^{-1} estimated by Bergametti et al. (1992). SRP wet deposition, 3.0 +/- 0.5 mg-PO_4^{3-}-P m^{-2} y^{-1}, is lower than Sicilian basin deposition flux, which was estimated equals to 10 mg-PO_4^{3-}-P m^{-2} y^{-1}. (Öszoy, 2003).Migon et al. (1989), at Cap Ferrat and Col de Bavella (Corsica) reported wet atmospheric input of total nitrogen slightly higher than our study, from 610 to 766 mg-N m^{-2} y^{-1}. Arsene et al. (2007) reported wet deposition fluxes of NO_3 for various sites in Europe. The NO_3 wet depositions range from 28 mg-N m^{-2} y^{-1} (Romania ; Bytnerowicz et al. (2005)) to 644 mg-N m^{-2} y^{-1} (Poland; Polkowska et al., 2005)). It demonstrates a high variability from location to location. Nevertheless, NO_3 depositions observed in site close to the Mediterranean : Turkey (153 mg-N m^{-2} y^{-1}) , Greece (94 mg-N m^{-2} y^{-1}) and Spain (273 mg-N m^{-2} y^{-1}) are in the same magnitude as ours observations : 200 to 246 mg-N m^{-2} y^{-1}

The annual wet deposition rates of major ions estimation obtained from period #1 and #2 are in the same range. The range of wet deposition rate are in agreement with value given by Tuncel (2001) for the Mediterranean area. But dry deposition varies from spring to autumn. For all the considered ions dry deposition rates is less during the autumn than during the spring. Carbonate and Cl are the dominating fraction of deposited mass. Then sulfate and SO_4 are also representing animportant contribution to the mass deposited. The presence of NA and Cl in the high mass deposition is due to the proximity of the station to the Mediterranea Sea.

Wet, dry and bulk annual depositions rates are estimated over a small Mediterranean basin. These first results are giving indicative values of the atmospheric input to Mediterranean coastal area. Further analysis should be conducted on the dataset in order i)to evaluate the spatial and temporal variation in rainwater for contaminants, ii)to identify the possible origins and the main factors affecting the concentrations of the measured species at the rain event time scale iii) take into account trace element content. And therefore refine the atmospheric deposition rate and the contaminant basin and lagoon budget.

Table 2: Deposition rate measurement for two continuous exposition period and estimation of the annual deposition rate for major elements and nutrients

	Deposition	Period 1*	Period 2**	Annual extrapolation (mg m^{-2} y^{-1})		
				Period 1*	Period 2**	VWM C
bulk	TN (µg-N. m^{-2} d^{-1})	4031	3081	1131	866	-
	TP (µg-P. m^{-2} d^{-1})	95	135	32.5	48	-
	HCO_3 (mg m^{-2} d^{-1})	24	19	6243	5211	
	Cl (mg m^{-2} d^{-1})	17	13	4906	3225	
	NO_3 (mg-N. m^{-2} d^{-1})	1.4	1.0	354	272	
	SO_4(mg. m^{-2} d^{-1})	11	9	3311	2106	
	Ca (mg m^{-2} d^{-1})	11	13	2650	3487	
	Mg (mg m^{-2} d^{-1})	11.2	0.8	371	224	
	Na (mg m^{-2} d^{-1})	11	5	3126	1207	
	K (mg m^{-2} d^{-1})	1.4	1.2	391	588	
wet	TN (µg-N. m^{-2} d^{-1})	2566	2161	557	506	602
	TP (µg-P. m^{-2} d^{-1})	27	30	5.9	7.0	7.3
	HCO_3 (mg m^{-2} d^{-1})	19	14	4126	3276	3186
	Cl (mg m^{-2} d^{-1})	12	13	2606	3042	2144
	NO_3 (mg-N. m^{-2} d^{-1})	1.1	0.8	246	199	244
	SO_4(mg. m^{-2} d^{-1})	6	9	1303	2106	1135
	Ca (mg m^{-2} d^{-1})	8	11	1737	2574	1195
	Mg (mg m^{-2} d^{-1})	0.7	0.8	152	187	107
	Na (mg m^{-2} d^{-1})	7	5	1520	1170	1069
	K (mg m^{-2} d^{-1})	1.8	0.7	391	398	194
dry	TN (µg-N. m^{-2} d^{-1})	1573	988	574	360	-
	TP (µg-P. m^{-2} d^{-1})	73	113	27	41	-
	HCO_3 (mg m^{-2} d^{-1})	5.8	5.3	2117	1935	-
	Cl (mg m^{-2} d^{-1})	6.3	0.5	2300	183	-
	NO_3 (mg-N. m^{-2} d^{-1})	0.3	0.2	108	11	-
	SO_4(mg. m^{-2} d^{-1})	5.5	0.	2008	0	-
	Ca (mg m^{-2} d^{-1})	2.5	2.5	913	913	-
	Mg (mg m^{-2} d^{-1})	0.6	0.1	219	37	-
	Na (mg m^{-2} d^{-1})	4.4	0.1	1606	37	-
	K (mg m^{-2} d^{-1})	0.	0.52	0	190	-

* period 1 is continuous exposition from 02/24/2004 to 05/03/2004
* period 2 is continuous exposition from 09/13/2004 to 12/13/2004

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