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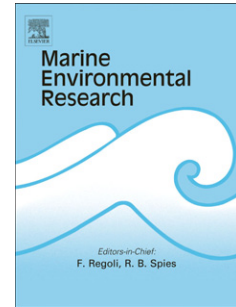
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1 IMPACTS OF MARINE AQUACULTURE AT LARGE SPATIAL SCALES:
2 EVIDENCES FROM N AND P CATCHMENT LOADING AND
3 PHYTOPLANKTON BIOMASS
4

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24 **Abstract**

25 While several studies point at off-shore aquaculture as a possible source of impacts on
26 the local marine environment, very few have analysed its effects at large scales such as
27 at the bay, gulf or basin levels. Similar analyses are hampered by the multiple sources of
28 disturbance that may concomitantly affect a given area. The present paper addresses
29 these issues taking the Gulf of Castellammare (Southern Tyrrhenian Sea) as an example.
30 Nitrogen (N) and phosphorous (P) loads were calculated for the period 1970-2007, and
31 compared to chlorophyll-a concentration as measured inside and outside the Gulf over
32 the same period. Results indicate that N and P catchment loading has constantly
33 decreased because of improved environmental management. Nevertheless, nutrient
34 concentration in the Gulf has steadily increased since the establishment of aquaculture
35 facilities in 1999. Chlorophyll-a concentration followed this trend, showing a marked
36 increase from 2001 onwards. In the same period, chlorophyll-a concentrations measured
37 inside and outside the Gulf have significantly diverged. As all the other possible causes
38 can be ruled out, aquaculture remains the sole explanation for the observed situation.
39 This paper demonstrates for the first time ever that off-shore aquaculture may affect the
40 marine ecosystem well beyond the local scale and provides an additional element of
41 concern to be kept into consideration when allocating oceans' space for new fish
42 farming activities.

43
44 **Key-words:** Aquaculture; Nitrogen; Phosphorus; Phytoplankton; Chlorophyll-a;
45 Mediterranean Sea

46

47

48 1. Introduction

49 Since the 1990s, coastal aquaculture has become a consolidated economic activity as a
50 global alternative to land-based aquaculture (Beveridge, 1996), and has been the major
51 cause of a rapid domestication of marine species (Duarte et al., 2007). Over the last two
52 decades, along with the almost exponential increase in economic gains provided
53 worldwide, coastal aquaculture has attracted a large interest either from marine
54 scientists and policy makers for the potential detrimental effects of this industry on the
55 surrounding marine environment (e.g., Munday et al., 1994; Pillay 2004; Pusceddu et
56 al., 2007; Holmer et al., 2008a-b; Borja et al., 2009). Most of the research carried out so
57 far has, indeed, documented significant changes in the physical, chemical and biological
58 attributes of sediments and the water column around off-shore aquaculture plants, and
59 these include, among the others: i) increasing organic matter contents and compositional
60 changes of the sediment below fish cages (e.g., Pusceddu et al., 2007); ii) altered
61 inorganic and organic chemistry of farm water and sediments (e.g., Sarà, 2007a-b-c;
62 Sanguling et al., 2011); iii) altered abundance, biomass and biodiversity of micro- (La
63 Rosa et al., 2004), meio- (Mirto et al., 2010) and macro-benthic (Kalantzi and
64 Karakassis, 2006) communities; iv) modified distributional patterns of phyto- and
65 microplankton abundance and production (e.g., Pitta et al., 1998). The extension of the
66 abovementioned changes either in water and benthic variables has been observed
67 generally at spatial scales ranging from meters to a few hundred meters from the edge of
68 the pens, generally not exceeding 1-2 kilometres from the rearing site (e.g., Gowen and
69 Bradbury, 1987; Sarà et al., 2006; Pusceddu et al., 2007; Holmer et al., 2008a-b; Borja

70 et al., 2009). Machias et al. (2005) showed that the presence of fish farms can increase
71 the biomass of wild fish populations at a spatial scale larger than the immediate vicinity
72 of fish cage, though with no clear impact on species diversity. However, to our best
73 knowledge, information about aquaculture impacts on water and sediment variables at
74 large spatial scale (e.g., at a basin scale) are practically not existent. Furthermore, three
75 recent and independent meta-analyses of more than 1000 study cases have pinpointed
76 the unevenly spatially resolved design of most of the reviewed experiments aimed at
77 assessing the effects of aquaculture on a wide array of abiotic (i.e., temperature, salinity,
78 pH, water turbidity, dissolved oxygen, inorganic nutrient) and biotic (e.g., chlorophyll-
79 a, organic carbon and nitrogen, prokaryotic and phytoplankton abundance) variables in
80 the water column (Sarà, 2007a-b-c).

81 The persistence of these gaps of knowledge is, on the one hand, partially due to
82 the difficulty in disentangling, at large spatial scales, the ecological effects induced by
83 coastal aquaculture from those induced by other natural (e.g., currents, circulation) and
84 anthropogenic sources of environmental variability (e.g., urban and/or industrial
85 agglomerates, terrestrial zootechny and agriculture). On the other hand, the sampling
86 bias affecting many of the studies carried out on the impacts of aquaculture on the
87 neighbouring environment (with a few exceptions, e.g., Pusceddu et al., 2007; Holmer
88 et al., 2008a; Sanz-Lázaro et al., 2011) makes even more difficult the task of identifying
89 large-scale ecological effects of aquaculture because of the presence of possibly
90 confounding explanatory variables and uncontrolled sources of variability.

91 In spite of these premises, it could be hypothesised that waste nutrients released
92 by fish farming activities might co-act with N and P released in near-shore waters
93 through continental run-off, rivers and seasonal streams, altogether promoting

94 eutrophication of coastal waters (i.e., either in terms of increased nutrient contents,
95 enhanced primary production and accumulation of algal biomass; Cloern, 2001).
96 However, we still do not know yet whether additive loads released by aquaculture may
97 trigger water eutrophication at a very large distance (>10 km) from the local area of
98 waste release, or how the duration of exposure to aquaculture wastes may affect the
99 ecological functioning of the receiving basin. As a consequence, large-scale mitigation
100 of fish-farming impacts has been *de facto* neglected in the present management
101 practices of coastal areas worldwide, thus posing a concern also in the light of the
102 predicted tendency of fish farming industries to move in off-shore waters, as the space
103 in coastal waters is increasingly becoming a limiting factor (Holmer, 2010).

104 In this study, we aimed to evaluate, in an unprecedented manner: 1) whether and
105 how N and P catchment loading may affect quasi-decadal variations in the pelagic
106 phytoplankton biomass of a Mediterranean Sea Gulf (Gulf of Castellammare, Southern
107 Tyrrhenian Sea), and 2) whether and how much additional N and P inputs released from
108 off-shore fish farming in the same area have contributed to the observed variations. To
109 achieve these objectives we: i) quantified the amount of N and P loads in the Gulf
110 (using empirical and statistical data) coming from point (population, industry, terrestrial
111 and marine farming) and non-point (uncultivated and cultivated soils) sources; ii)
112 analysed the quasi-decadal fluctuations in phytoplankton biomass (expressed as
113 chlorophyll-a concentrations in the seawater), and iii) analysed the difference in
114 chlorophyll-a concentrations within the Gulf vs. those in open-sea waters, to understand
115 if chlorophyll-a fluctuations occurred at the Gulf scale were due to local or basin-wide
116 long-term changes of the whole Southern Tyrrhenian Sea.

117

118 2. Material and Methods

119 2.1 The study area

120 The Gulf of Castellammare (Tyrrhenian Sea, surface $\sim 370 \text{ km}^2$) is the widest inlet along
121 the northern coast of Sicily (Fig. 1a). Bordered by rocky cliffs, it opens into shallow
122 sandy shores with patches of rocky bottoms and interspersed seagrass beds. The area is
123 influenced by limited continental inputs ($\sim 88,500 \times 10^3 \text{ m}^3 \text{ y}^{-1}$; about 600 times less
124 than the Po River input in the northern Adriatic Sea and about 25 times less than the
125 Simeto river inputs in the South Ionian Sea along the SE coasts of Sicily; Sarà et al.,
126 1998; Sarà et al., 2004; Modica et al., 2006; Sarà et al., 2007). Those inputs are
127 discharged by two of the four seasonal streams located in the central sector of the Gulf,
128 namely the Nocella (13 km long, with a surface of about 15,000 ha) and the S.
129 Bartolomeo (50 km long, 40,000 ha) streams (Fig. 1a). These two small rivers show
130 only narrow fluctuations of discharge rates (on average $0.32 \text{ m}^3 \text{ sec}^{-1}$ and $0.24 \text{ m}^3 \text{ sec}^{-1}$
131 for the Nocella and S. Bartolomeo river, respectively; PTA, 2007) generally interrupted
132 by seasonal rainfall episodes which cause short (i.e., a few days) flushing events (never
133 exceeding $1.2 \text{ m}^3 \text{ sec}^{-1}$; PTA, 2007). The whole drainage basin of the Castellammare
134 Gulf includes 16 small municipalities (a total of about 175,000 inhabitants), of which
135 just six are located on the coast. The local population density is about 100 people per
136 Km^2 (ISTAT, 2001).

137 The economy of the Gulf catchment is based on the tertiary sector, including
138 tourism and small industries (fishery processing), for about 60-70% of the local gross
139 domestic product. Agriculture, pastoral, fish farming and fisheries account altogether
140 for about 30-40% of the local gross domestic product (ISTAT, 2005; PTA, 2007). In the
141 Castellammare Gulf, fisheries have played a major role in the local economy for many

142 decades but only till 1990 (Badalamenti et al., 2000), when bottom trawling was banned
143 from the area (Pipitone et al., 2000). One of the biggest Mediterranean aquaculture
144 companies has been operating in the eastern area of the Gulf since 1999, producing
145 about 500 t y⁻¹ of sea bass (*Dicentrarchus labrax*) and sea bream (*Sparus aurata*) ever
146 since (Mazzola et al., 2000; Sarà et al., 2007). Since 2000, a bluefin tuna farming
147 industry (only fattening) located in the central sector of the Gulf (off the small harbour
148 in Castellammare del Golfo; Fig. 1a; Messina et al., 2001) produces about 1,000-1,100
149 tons of farmed tuna per year (from July to December).

150

151 *2.2 Long-term variations in chlorophyll-a, total N and P concentrations in the* 152 *Castellammare Gulf and the adjacent open waters*

153 We inventoried records of chlorophyll-a ($\mu\text{g L}^{-1}$), total nitrogen (N, μM) and
154 phosphorus (P, μM) concentrations in the upper (within 1 m depth) surface layer of the
155 Gulf of Castellammare waters in the period 1970-2007. Data were obtained from
156 research and oceanographic cruises carried out by several Italian Academic Institutions
157 and Governmental Agencies as well as from our own data (Fig. 1a). Those data,
158 consistently obtained using spectrophotometric techniques (chlorophyll-a: Lorenzen,
159 1965; total N and P: Valderrama, 1981), were averaged to obtain mean annual values
160 and, though unevenly dispersed in space and time, used to assess inter-annual variations
161 in the Gulf of Castellammare. Moreover, within the period 1998-2007, we inventoried
162 also data of chlorophyll-a concentrations from five areas spread in the Southern
163 Tyrrhenian Sea along the western coast of Sicily (Fig. 1b). These data, extracted from
164 the Environmental Marine Information System (EMIS) published at
165 http://emis.jrc.ec.europa.eu/4_1_gismap.php and maintained by the Joint Research

166 Centre of the European Commission (2008), were used to compare inter-annual trends
167 (1998-2007) in phytoplankton standing stocks inside and far from the Gulf of
168 Castellammare (Fig. 1a-b).

169

170 *2.3 Estimates of N and P loads entering the Gulf*

171 Wasted loads (tons per year) of total N and P coming from the most important point
172 (i.e., identifiable localized sources, such as resident and touristic cities/villages;
173 industries; zootechny plants; fish farms; cattle, swine, sheep and goat farms) and non-
174 point (i.e., diffuse sources, such as agriculture and uncultivated soils) sources were
175 estimated according to the transformation coefficients recently proposed by Pagnotta
176 and Barbiero (2003; Table 1) and adopted by the Italian Ministry of Public Health to
177 assess the N and P loads along the Italian coasts. Since these coefficients were assumed
178 to be constant across the year and since data about inputs coming from tourism are not
179 available for the region, we are aware that our estimates of the N and P wastes entering
180 annually the Castellammare Gulf from inland are affected by some uncertainty.
181 Nevertheless, statistical data released by the Italian Statistical Institute (ISTAT),
182 similarly to those released by Statistical Institutes of most Western European countries,
183 are only available as annual means, so that we were not able to carry out our estimates
184 on shorter (e.g., monthly or seasonal) temporal scales.

185 Data on resident population, industry, terrestrial farming and cultivated and non-
186 cultivated soils in the study area were extracted from censuses carried out by the Italian
187 Statistical Institute (ISTAT) in 1962, 1972, 1982, 1990, 2000 and 2007 (only
188 population).

189 Estimates of nitrogen and phosphorus wastes released from seabream, seabass
190 and bluefin tuna farms and entering the Castellammare Gulf were based on individual
191 gross metabolic waste outputs and coefficients of particulate and dissolved N and P
192 released on a daily and biomass basis, as modelled in Piedecausa et al. (2010) and
193 Aguado-Giménez et al. (2006). These models allow simulations of real rearing and
194 husbandry practices. Gross metabolic waste output estimates for three cultivated species
195 (i.e., seabream, seabass and tuna) in the Gulf were made on a nutritional basis, using the
196 following bioenergetic model (Piedecausa et al., 2010):

$$197 \quad C = G + E + F$$

198 where C is the nutrient (nitrogen, N and phosphorus, P) content in the food intake, G
199 represents the retained nutrient as growth, E and F represent the excretory (dissolved)
200 and faecal (particulate) losses, respectively. To calculate waste output coefficients of
201 particulate and dissolved nitrogen and phosphorus on a daily and per biomass basis (mg
202 N or mg P kg⁻¹ d⁻¹), the nitrogen and phosphorus content in the diet and faeces, the
203 feeding conversion rate and nitrogen and phosphorus digestibility were considered
204 (Piedecausa et al., 2010).

205

206 *2.4 Statistical analyses*

207 Since we had gathered data from very different sources, we were forced to analyse our
208 hypotheses using a combination of different statistical techniques. Therefore, we used
209 the t-Student (Sokal and Rohlf, 1981) to test differences in the measured variables
210 among cruises and years, while the Permutational Multivariate Analysis of Variance
211 (PERMANOVA; Anderson, 2001) was used to test the (null) hypothesis of no
212 significant differences in chlorophyll-a concentrations between data collected during the

213 cruises (spectrophotometrically determined) and the data extracted from the EMIS GIS
214 database (obtained by means of satellite sensors). The latter comparison was done
215 separately for each year as well as for the data recorded before and after the launch of
216 aquaculture activities in the Gulf. We applied the linear correlation approach to relate
217 chlorophyll-a and nutrients concentrations (Sokal and Rohlf, 1981) The role of rainfall,
218 water temperature and salinity on long-term (from 1980 to 2006) variations in
219 chlorophyll-a annual mean annual concentrations was assessed by means of a distance-
220 based multivariate multiple regression analysis with forward selection of explanatory
221 variables, using the DISTLM Fortran-written routine (McArdle and Anderson, 2001).

222

223 **3. Results**

224 *3.1 Long-term changes in N and P loading from different sources*

225 N wastes produced separately by all sources, with exception of agriculture, increased
226 from 1970 to 1990 (by on average 15% for population, industry and zootechny), than all
227 consistently decreased till 2007 (by about 30% on average; Table 2). P wastes derived
228 from all sources increased in the period 1970-1990 (by on average 16% for population,
229 industry and zootechny); P released by resident population and tourists decreased in
230 2000 (by 4% when compared with the preceding ten years average) while increased
231 again in 2007. P released by industry, terrestrial farming and non-cultivated landscapes
232 decreased from 1990 to 2007 (~32%; Table 3). N and P wastes derived from agriculture
233 decreased almost constantly in the investigated period, with values in 2007 about 15%
234 lower than in 1970. Generally, N and P wastes derived from diffuse sources (e.g.,
235 agriculture and non cultivated landscapes) decreased almost continuously from 1970 to
236 2007 (by about 34% for both N and P), whereas wastes derived from point sources

237 showed a biphasic temporal trend, increasing from 1970 to 1990 than decreasing again
238 till 2007 (Fig. 2a-b).

239 Over the study period (1970-2007), the main sources of nitrogen and phosphorus
240 were from cultivation (on average 1,058 tons of N and 39.6 tons of P per year,
241 representing ~67.2% and 36.7% of total N and P inputs, respectively) and human
242 population (378.5 tons of N and 56.9 tons of P per year, representing 24.4% and 52.4%
243 of total N and P inputs, respectively).

244 In 1999, a fish farm producing about 500 tons per year of sea bass
245 (*Dicentrarchus labrax*) and sea bream (*Sparus aurata*) was installed in the Gulf. While
246 no data are available for the 1999, the total amount of N and P discharged every year
247 from 2000 and 2007 in the Gulf by sea bass, sea bream and tuna fish farming activities
248 accounted up to 274.9 tons, representing about 17.4% and 34.2.5% of the relative N and
249 P total inputs per year. These, in turn, represent about 51.5% and 122.0% of the N and P
250 inputs derived from coastal discharge, respectively (Table 2-3).

251

252 3.2 Temporal changes in chlorophyll-a in the Gulf

253 Dissolved nutrient concentrations in the Gulf, as extrapolated from field campaigns
254 data, showed a steady increase in mean annual values from 1980 to date (Fig. 3). On the
255 contrary, in the upper 10-m layer of the water column annual mean values of
256 chlorophyll-a concentration, used here as a proxy of phytoplankton biomass, decreased
257 between 1980 and 1990 (by 30%), and another significant (t-Student $p < 0.05$) drop was
258 observed between 1991 ($0.04 \mu\text{g L}^{-1}$) and 1995 ($0.01 \mu\text{g L}^{-1}$). Chlorophyll-a
259 concentrations have significantly risen again in the Gulf after 1998 (t-Student test
260 between before and after 1998 values: $p < 0.05$), setting on constant values until 2001,

261 when another sudden positive shift led to the maximum value over the entire study
262 period ($0.36 \mu\text{g L}^{-1}$ in 2006-2007) (Fig. 4). Such increase in phytoplankton biomass
263 observed in the Gulf of Castellammare was not observed in any of the five areas outside
264 the Gulf spread in the Southern Tyrrhenian Sea. Indeed, while in 1998 and 1999
265 chlorophyll-a concentrations increased consistently either in the Gulf and the open sea
266 area, showing not significant differences (PERMANOVA $p>0.01$; Fig. 4), from 2000
267 (about two years after the beginning of aquaculture operations), chlorophyll-a
268 concentrations in the open-sea and the Gulf started to be significantly different
269 (PERMANOVA, $p<0.05$; Fig. 4). Accordingly, since 2000, chlorophyll-a
270 concentrations followed the increase in N and P loading (Table 2-3), and were
271 significantly correlated to phosphate concentrations (Fig. 5).

272

273 **4. Discussion**

274 The most recent literature has provided consistent and repeated evidence that
275 aquaculture can have remarkable effects on the surrounding environment with clearly
276 detectable effects on the ecology of the benthos (Holmer et al., 2008a-b), but
277 idiosyncratic effects on the ecology of the water column (Sarà, 2007a,b,c). Many of
278 those investigations have proven that effects are detectable at a relatively short distance
279 (i.e. hundreds of meters) from the farms, mainly because of the dilution effect promoted
280 by the local currents (Sarà et al., 2006; Pusceddu et al., 2007; Mirto et al., 2010).
281 However, given the well documented consistency of nutrient release from fish farms
282 into the marine environment (Piedicausa et al., 2010) and the crucial importance of
283 water currents in the dispersion of wastes (Pusceddu et al., 2007; Sanz-Lázaro et al.,

284 2011), effects at large spatial scales may be reasonably expected in protected gulfs, bays
285 and inlets, such as the area under scrutiny in this study.

286 We show here that the surface waters of the Castellammare Gulf have
287 experienced large-scale and long-term variations in the nutrient and chlorophyll-a
288 concentrations, mostly as the result of the chronic release of nutrient waste produced by
289 local aquaculture. Between the 1970s and the 1990s, N and P loads from the Gulf of
290 Castellammare catchment have constantly increased, though diminishing around 2000
291 (Tab. 2-3). This is consistent with the socio-economic evolution of several Western
292 Mediterranean and European countries over the last decades (Gurluk, 2009). The
293 economic development of many Mediterranean coastal areas, led by tourism and
294 resulting (among other effects) in the increased discharge of N and P into near shore
295 waters (Garau-Taberner and Manera, 2006), was likely counteracted around 2000 by the
296 effects of the European Union's (EU) Urban Wastewater Treatment Directive
297 (91/271/EEC - 21 May 1991). The implementation and enforcement of the EU Directive
298 has resulted in a significant amelioration of wastewater treatment before discharge into
299 European rivers, and in measurable improvements in the water quality. This has in turn
300 led to a decrease in point sources of N and P reaching near-shore waters in 2000.
301 Contextually, agricultural run-off has significantly declined, reducing the discharge of
302 N and P by at least 35% from the seventies to date (Tables 2-3). This has been probably
303 due to the progressive shift of the local economy from agriculture to more profitable
304 activities like tourism (Tyrakowski, 2004). Similar trends have been quite common in
305 all North Mediterranean countries (Milesi et al., 2003), though in the North African
306 countries the tendency appears to be inverse (e.g., agriculture, population and tourism
307 are expected to increase till 2025, with effects still to be determined; Turley, 1999).

308 Data for the Gulf of Castellammare do not strongly deviate from the common European
309 scheme of the last decade. Indeed, suspended chlorophyll-a concentration was rather
310 high in the 1980s, probably due to the almost total deregulation of N and P catchment
311 loading, to the absence of wastewater treatment and to the intense bottom trawling
312 activities carried out in the study area, which could have contributed to an enhanced
313 mobilisation of benthic nutrients resuspended by fishing gears (Pusceddu et al., 2005).
314 At the end of the eighties, important steps towards the correct management of natural
315 resources in the catchment area included the regulation and depuration of urban wastes
316 released at sea and the ban on bottom trawling (since 1989; Pipitone et al., 2000). As a
317 plausible immediate consequence, the Gulf waters dropped to an ultra-oligotrophic
318 status (i.e., $<0.1 \mu\text{g chlorophyll-a L}^{-1}$, according to the OECD scheme based on
319 concentrations of chlorophyll-a; Zurlini, 1996; Sarà and Mazzola, 1997; Sarà et al.,
320 1998), which lasted till 1999, when chlorophyll-a concentration in the surface waters of
321 the Gulf steadily and significantly increased compared to open-sea baseline data sets
322 (i.e., EMIS data). Between 2001 and 2007, the concentrations of chlorophyll-a in the
323 surface waters of the Gulf were from 3 ($0.15 \mu\text{g L}^{-1}$, in 2001) to over 10 times (ca. 0.50
324 $\mu\text{g L}^{-1}$, as averaged for 2006-2007) higher than in the reference open-sea values. In the
325 period 2004-2005 the concentration of chlorophyll-a in the eastern part of the Gulf was
326 on average about $1.1 \mu\text{g L}^{-1}$, but exceeded $1.5 \mu\text{g L}^{-1}$ (with frequent peaks over $3 \mu\text{g L}^{-1}$)
327 close to the aquaculture facilities (Sarà et al., 2009).

328 The divergent temporal patterns in chlorophyll-a concentrations in the two
329 investigated systems might be attributed to a wide and complex array of explanatory
330 variables and processes. Many reasons, indeed, could be invoked for explaining the rise
331 in chlorophyll-a concentration in the surface waters of the Gulf which, among the

332 others, include: i) increased terrestrial runoff due to rainfall; ii) increased release of
333 nutrients from the sea bottom as affected by sediment resuspension induced by bottom
334 trawling, iii) variations in global climate factors (e.g., temperature, storminess, etc.) and
335 iv) increased nutrient loadings from local point and/or non-point sources. Over the last
336 decades rainfall in the Gulf had a swinging trend (Fig. 6; data from SEDEMED II,
337 2009) and was accompanied by a general decrease of the number of wet days in
338 Southern Italy (Brunetti et al., 2004). Nor rainfall or water temperature and salinity did
339 explain significant proportions of chlorophyll-a variations in the Gulf (Table 4).
340 Therefore, we can conclude that variations in the thermohaline conditions of the water
341 column and in rainfall did not have crucial roles in modifying the trophic status of the
342 Gulf waters. Bottom trawling was the main economic activity in the Castellammare
343 Gulf until the ban in 1989 (Badalamenti et al., 2000; Pipitone et al., 2000). Disturbance
344 of the seabed resulting from bottom trawling affects different ecosystem processes such
345 as the rate and magnitude of nutrient regeneration (Pusceddu et al., 2005; Dounas et al.,
346 2007). By cascade, plankton communities are expected to be influenced, which adds a
347 pulsing component to local primary production (Dounas et al., 2007). The ban on
348 bottom trawling in the Gulf (1989) could have led to a reduction of sedimentary nutrient
349 contribution to the water column, so that we can exclude that the rise in nutrient and
350 chlorophyll-a concentrations observed in the Gulf since 1999 was stimulated by bottom
351 fisheries.

352 Along with a general temperature increase of sea surface waters, other climate
353 related factors like ocean acidification could be invoked as additive macro-causes of the
354 deviation of ecosystem functions (including primary production) from their common
355 natural patterns (IPCC, 2007). Although it has been shown that the Mediterranean Sea

356 has experienced a significant increase in CO₂ water concentrations in the last 25 years
357 (Touratier and Goyet, 2011), it must be noticed that, whatever the (physical or
358 biological) mechanism involved, it can be reliably hypothesised that climate change in
359 the study area couldn't have determined a general increase in primary production. On
360 the one hand, this hypothesis is supported by the fact that an increase in sea surface
361 temperature, as the one observed in the whole study area, could have led – under
362 constant CO₂ concentration in the atmosphere, that is not the case (Touratier and Goyet,
363 2011) – to a decreasing CO₂ availability for primary producers (Harrison et al., 2005).
364 On the other hand, phytoplankton possess C-concentrating mechanisms
365 (counterbalancing CO₂ deficiency under warm temperature conditions; Giordano et al.,
366 2005), and the eventual increase in CO₂ contents in the seawater may have had a
367 mixture of relevant impacts on the availability of nutrients (Raven et al., 2005).
368 Moreover, the temperature increase occurred in the whole Mediterranean Sea basin has
369 considerably altered the water column stability and likely modified even the ecological
370 response of deep fauna (Coma et al., 2009). Altogether these data suggest that if a
371 change in primary production would have occurred in the study area this have led most
372 likely to a decrease or invariance rather than to an increase in chlorophyll-a
373 concentrations in the Gulf. It must be noticed also that the expected diminished
374 availability of nutrients under warmer conditions (Coma et al., 2009) might have
375 favoured smaller phytoplankton taxa, with a consequently lower total primary
376 production and C export to the sea bottom (Danovaro et al., 2000; Danovaro et al.,
377 2001). Therefore, we conclude that changes in the nutrient loads from inland, rainfall,
378 bottom trawling activities and other climate-related factors have had a minor role in
379 explaining the increase in chlorophyll-a concentrations in the sea surface waters of the

380 Gulf, as observed from 1999 onwards. Moreover, we would stress here that the huge
381 and continuous *in situ* nutrient enrichment due to aquaculture in the Gulf could have
382 reasonably been a major factor determining the observed divergence from the open sea
383 area. However, we also acknowledge that other local forcing factors (including
384 circulation and altered food web characteristics), acting individually or synergistically,
385 might have contributed to promote and maintain the observed shift in the productivity
386 of Castellammare Gulf when compared with the adjacent open Tyrrhenian Sea. We also
387 foster further research addressing these causes in future investigations. Nevertheless, the
388 results of our study provide the first evidence that a long-term exposure of a certain
389 coastal embayment to aquaculture wastes might result in a general modification of its
390 trophic status (i.e. nutrient concentrations and phytoplankton biomass) at a spatial scale
391 never considered before.

392 Our conclusions, on the one hand, give us the opportunity to stress the need of:
393 i) improved bio-monitoring protocols to detect possible impacts of aquaculture at spatial
394 scales larger than usual (i.e., > 1-10 km from the farming structures) and ii) more
395 appropriate temporal scales of investigation. On the other hand, our results provide an
396 additional element of concern to be kept into consideration when, likely in the very next
397 future, fish farming activities will expand in off-shore waters, with a potentially
398 expectable impact on the trophic status of vaster regions of the coastal oceans (Holmer,
399 2010).

400

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410

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ACCEPTED MANUSCRIPT

The manuscript provides much-needed empirical data addressing the contentious but current question of whether aquaculture has an effect on local marine environment at a spatial scales larger than 10-km. We addressed this issues using long series data of the Gulf of Castellammare (Southern Tyrrhenian Sea) as an example. Nitrogen (N) and phosphorous (P) loads were calculated for the period 1970-2007, and compared to chlorophyll-a concentration as measured inside and outside the Gulf over the same period. Our study provides results about that although N and P catchment loading constantly decreased because of improved environmental management, the nutrient concentration in the Gulf increased since the establishment of aquaculture facilities in 1999. In addition, we showed that chlorophyll-a concentration in the Gulf markedly increased from 2001 onwards, while outside the Gulf was significantly lower. As all the other possible causes can be ruled out, aquaculture remains the sole explanation for the observed situation.

1 Table 1. Transformation coefficients used to calculate gross wastes from point and non-
 2 point sources into dissolved N and P wastes entering annually the Gulf. Modified from
 3 Pagnotta and Barbiero (2003) and Piedecausa et al. (2010).
 4
 5

Point sources	Waste coefficient		Unit
	Nitrogen	Phosphorus	
Resident and touristic population	6.16	0.92	g per person day ⁻¹
Industry	27.4	0.09	g per employed day ⁻¹
Cattle farming	7.51	1.01	g per individual day ⁻¹
Swine farming	1.56	0.52	g per individual day ⁻¹
Sheep and goat farming	0.68	0.11	per individual day ⁻¹
Seabream farming *	298.9 (92.5)	12.2 (53.8)	mg Kg ⁻¹ fish ⁻¹ day ⁻¹ (particulate)
Seabass farming *	454.0 (21.5)	21.7 (50.7)	mg Kg ⁻¹ fish ⁻¹ day ⁻¹ (particulate)
Tuna farming *	852.0 (61.5)	65.5 (93.0)	mg Kg ⁻¹ fish ⁻¹ day ⁻¹ (particulate)
Non-point sources			
Uncultivated Soil	5.48	0.27	g per hectare day ⁻¹
Cultivated Soil	43.8	1.64	g per day h ⁻¹

6
 7 * Mean values obtained considering different values for small (60 Kg final weight) and
 8 big (140 Kg final weight) tunas (from Piedecausa et al., 2010)
 9

10 Table 2. Mean annual N loading (tons) entering the Gulf of Castellammare and percentages of each source to the total catchment. Data
 11 refer to the period 1970-2007.
 12

Point sources	Tons per year					Percentage of total catchment				
	1970	1980	1990	2000	2007	1970	1980	1990	2000	2007
Population	365.1	378.1	392.6	378.5	378.5	21.0	23.4	24.5	29.0	24.0
Industry	37.0	38.8	48.2	40.0	40.0	2.1	2.4	3.0	3.1	2.5
Seabass and seabream farm	0.0	0.0	0.0	0.0	84.9	0.0	0.0	0.0	0.0	5.4
Tuna farm	0.0	0.0	0.0	0.0	190.0	0.0	0.0	0.0	0.0	9.5
Cattle	15.4	21.7	13.4	4.9	4.9	0.9	1.3	0.8	0.4	0.3
Swine	0.0	0.8	4.1	3.1	3.1	0.0	0.0	0.3	0.2	0.2
Sheep and goat	0.0	7.8	16.1	7.6	7.6	0.0	0.5	1.0	0.6	0.5
Non-point sources										
Cultivated	1306.5	1151.7	1115.9	858.0	858.0	75.2	71.3	69.5	65.8	54.3
Not cultivated	14.6	15.5	15.3	12.1	12.1	0.8	1.0	1.0	0.9	0.8
Total Catchment	1738.5	1614.3	1605.5	1304.1	1579.0					
Total Coastal towns	671.9	560.6	543.8	445.9	445.9					
% Coastal vs. Catchment	38.7	34.7	33.9	34.2	28.2					
% Aquaculture vs. Catchment	0	0	0	0	17.41					
% Aquaculture vs. Coastal towns	0	0	0	0	61.65					

13

14

15 Table 3. Mean annual P loading (tons) entering the Gulf of Castellammare and percentages of each source to the total catchment. Data refer
 16 to the period 1970-2007.

17

Point sources	Tons per year					Percentage of total catchment				
	1970	1980	1990	2000	2007	1970	1980	1990	2000	2007
Population	54.5	56.5	58.6	56.5	58.6	51.3	53.8	54.8	61.3	40.9
Industry	0.1	0.1	0.2	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Seabass and seabream farm	0.0	0.0	0.0	0.0	16.1	0.0	0.0	0.0	0.0	11.2
Tuna farm	0.0	0.0	0.0	0.0	33.0	0.0	0.0	0.0	0.0	23.0
Cattle	2.1	2.9	1.8	0.7	0.7	1.9	2.8	1.7	0.7	0.5
Swine	0.0	0.3	1.4	1.0	1.0	0.0	0.2	1.3	1.1	0.7
Sheep and goat	0.0	1.3	2.6	1.2	1.2	0.0	1.2	2.4	1.3	0.9
Non-point sources										
Cultivated	48.9	43.1	41.8	32.1	32.1	46.0	41.1	39.0	34.8	22.4
Not cultivated	0.7	0.8	0.8	0.6	0.6	0.7	0.7	0.7	0.6	0.4
Total Catchment	106.3	104.9	107.1	92.3	143.4					
Total Coastal towns	44.1	41.0	41.7	37.2	37.2					
% Coastal vs. Catchment	41.5	39.1	39.0	40.3	25.9					
% Aquaculture vs. Catchment	0.0	0.0	0.0	0.0	34.2					
% Aquaculture vs. Coastal towns	0.0	0.0	0.0	0.0	131.9					

18

19 Table 4. Results of the DISTLM analysis with permutations to test the role of rainfall,
20 water temperature and salinity in explaining variations in chlorophyll-a concentrations
21 in the period 1980-2006 in the Gulf of Castellammare. SS = sum of squares; pseudo-F =
22 F statistic; P = probability level. Reported are the individual and cumulative percentage
23 of variance explained by each variable.
24

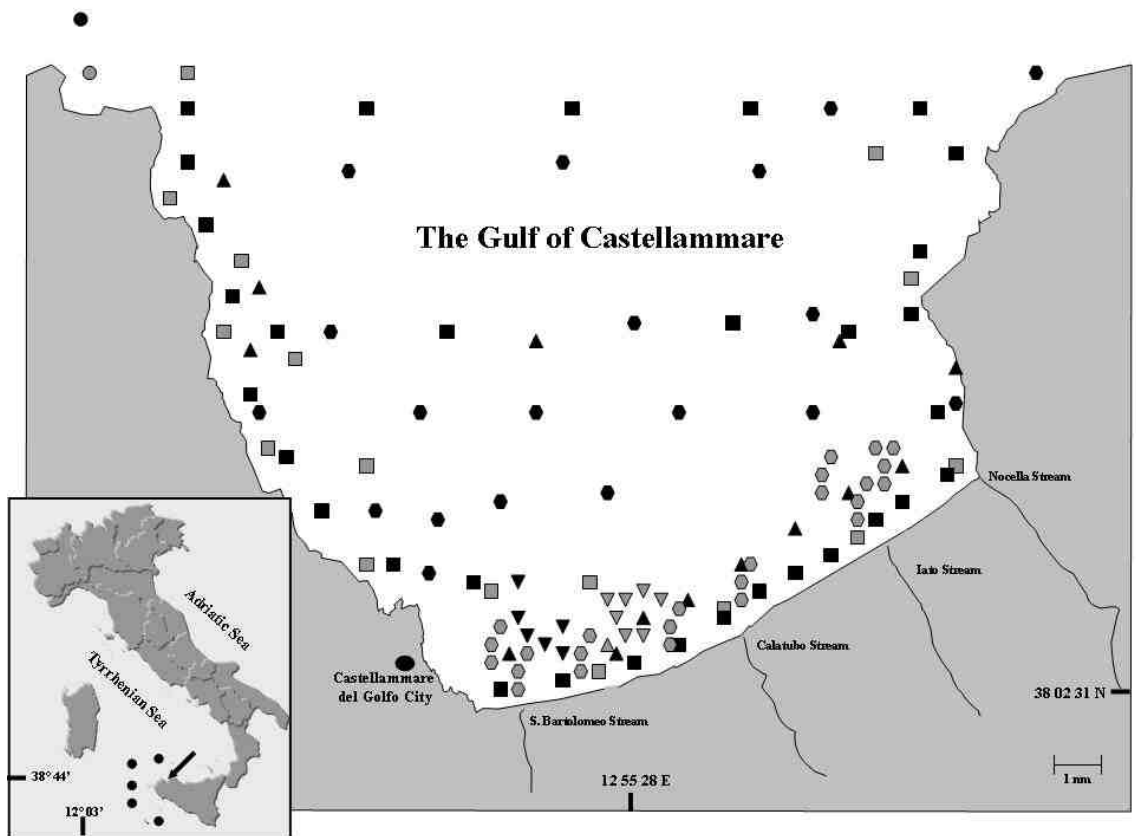
Variable	SS	pseudo-F	P	Explained variance	
				%	Cumulative
Rainfall	0.010	0.708	0.402	4.81	4.81
Water Temperature	0.002	0.158	0.738	1.15	5.96
Salinity	0.004	0.224	0.704	1.72	7.68

25

1 Figures Captions

2 Figure 1. Map of the Gulf of Castellammare with symbols referring to the source of data
3 as follows: grey circle = University of Messina, CNR Messina (1981); black circle =
4 University of Messina, CNR Messina (1982); grey square= Sicilian Region/Dept. Of
5 Territory and Environment (1984-1985); black square = CNR Castellammare (1990);
6 grey triangle = Sarà G. PhD Thesis (1991-1993); black triangle = Dept. of Animal
7 Biology and Marine Ecology, University of Palermo (1994-1995); grey reversed
8 triangle = Mediterranean Oceanological Centre (2000-2001); black reversed triangle =
9 Sarà G., unpublished data (2003-2005); grey hexagon = Sarà G., unpublished data
10 (2006-2007); black hexagon = Environmental Marine Information System (EMIS;
11 1998-2007).

12 In the small rectangle reported below on the left, the geographic area is showed with the
13 black arrow indicating the location of the Gulf of Castellammare in the northern Sicily
14 and five black circles indicating open-sea areas from which EMIS chlorophyll-a data
15 were retrieved.



16
17
18
19
20

Figure 1

1 Figures Captions

2 Figure 1. The study area; a) Map of the Gulf of Castellammare with symbols referring
3 to the source of data as follows: grey circle = University of Messina, CNR Messina
4 (1981); black circle = University of Messina, CNR Messina (1982); grey square=
5 Sicilian Region/Dept. Of Territory and Environment (1984-1985); black square = CNR
6 Castellammare (1990); grey triangle = Sarà G. PhD Thesis (1991-1993); black triangle
7 = Dept. of Animal Biology and Marine Ecology, University of Palermo (1994-1995);
8 grey reversed triangle = Mediterranean Oceanological Centre (2000-2001); black
9 reversed triangle = Sarà G., unpublished data (2003-2005); grey hexagon = Sarà G.,
10 unpublished data (2006-2007); black hexagon = Environmental Marine Information
11 System (EMIS; 1998-2007); b) open-sea areas from which EMIS chlorophyll-a data
12 were retrieved. The small arrow indicates the location of the Gulf of Castellammare; c)
13 monthly discharges coming from the two main rivers (Nocella and S. Bartolomeo) of
14 the area [tot. surface, ha = total surface of river basin; in. = total inhabitants insisting in
15 the basin of the river].

16 Figure 2. Nitrogen (a) and phosphorus (b) loads released annually from point (resident
17 and touristic population, industry and terrestrial farming) and non-point (agriculture and
18 non-cultivated landscapes) sources in the Gulf of Castellammare.

19 Figure 3. Annual means dissolved nitrogen (N tot, μM) and phosphorus (P- PO_4 , μM)
20 concentrations in the surface waters of the Gulf of Castellammare from 1982 to 2007.

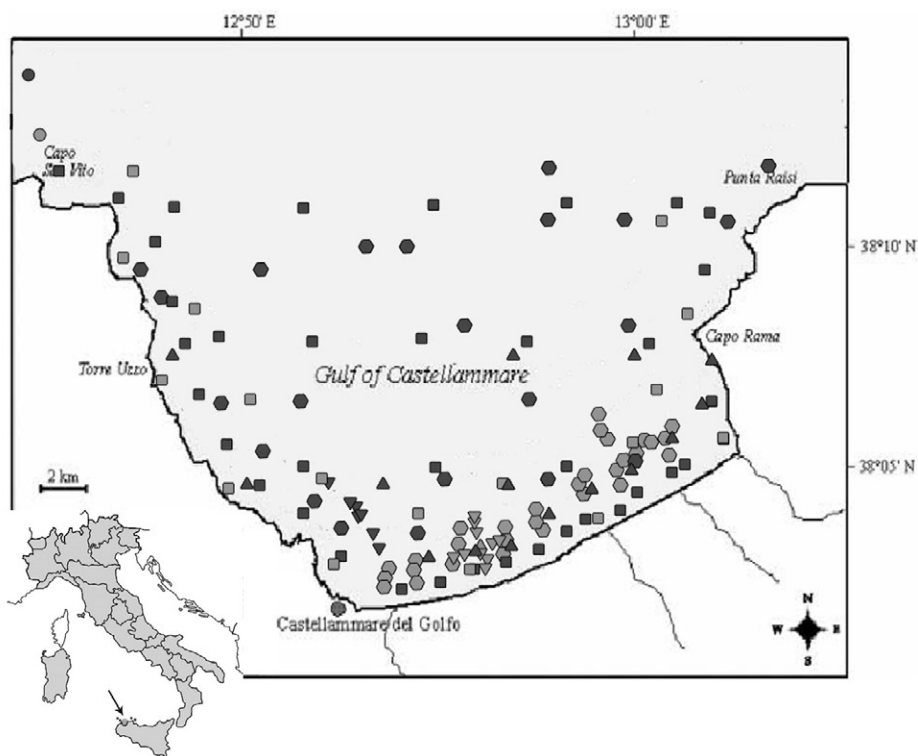
21 Figure 4. Chlorophyll-a ($\mu\text{g L}^{-1}$) inside and outside the Gulf of Castellammare between
22 1982 and 2007. Data have been obtained from cruises (squares) and satellite data (filled
23 circles, inside the Gulf; triangles, outside the Gulf). Open circles represent the average
24 values between cruise and satellite data inside the Gulf. No chlorophyll-a from satellite
25 data before 1998 are available.

26 Figure 5. Relationship between chlorophyll-a and phosphates in the Gulf of
27 Castellammare between 1980 and 2007.

28 Figure 6. Temporal trend of rainfall in the Castellammare Gulf (PTA, 2007)

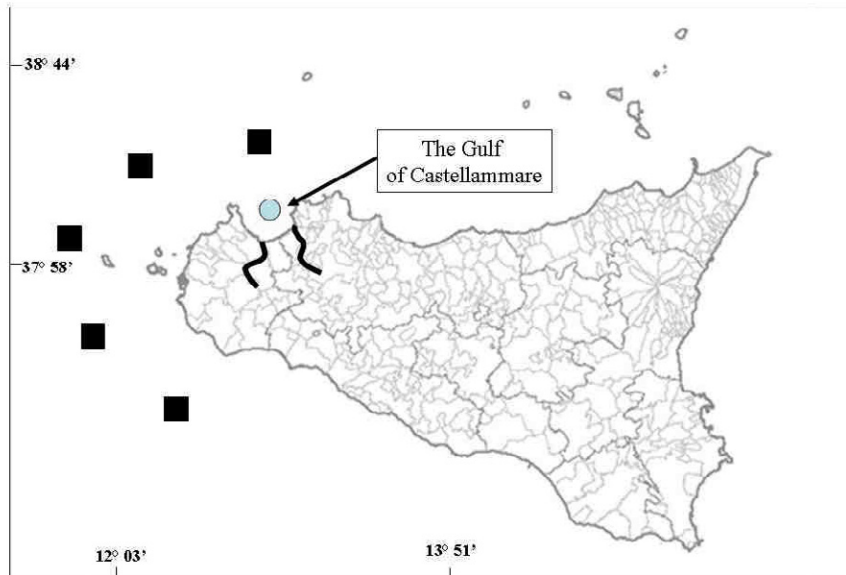
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Figure 1a



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Figure 1b

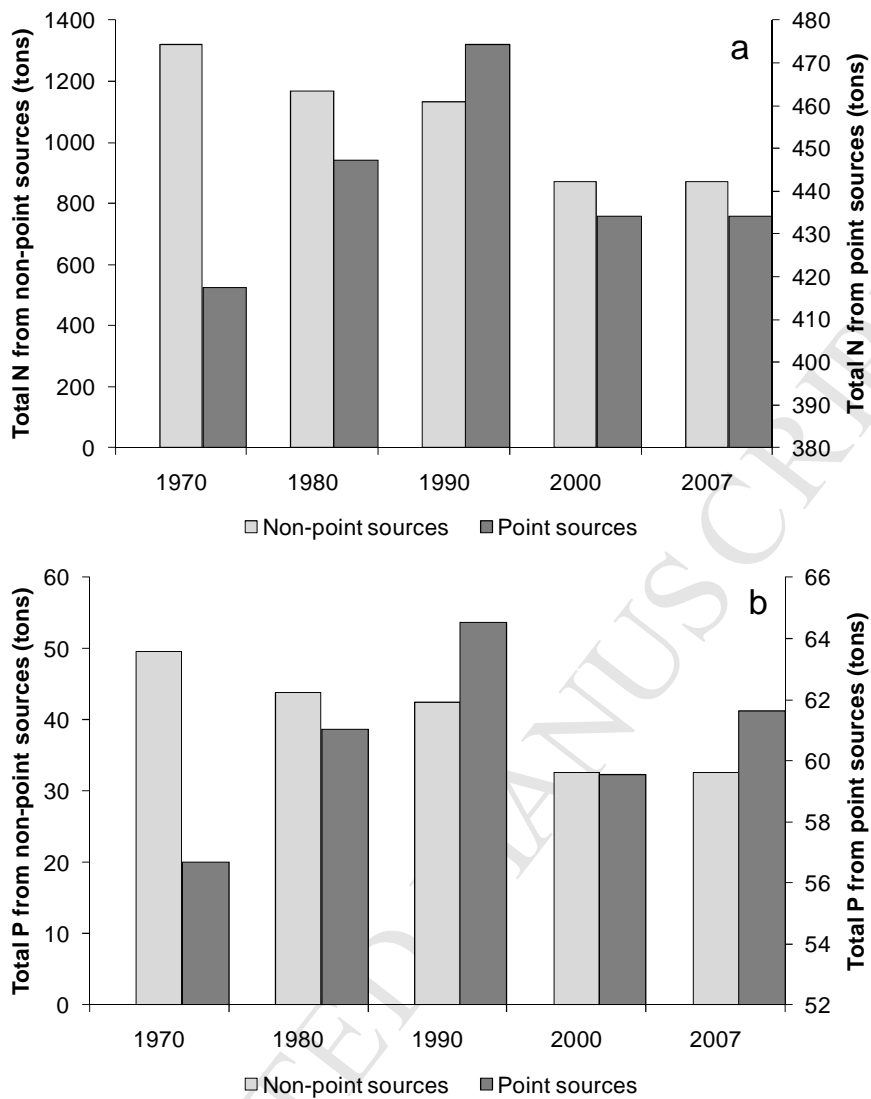
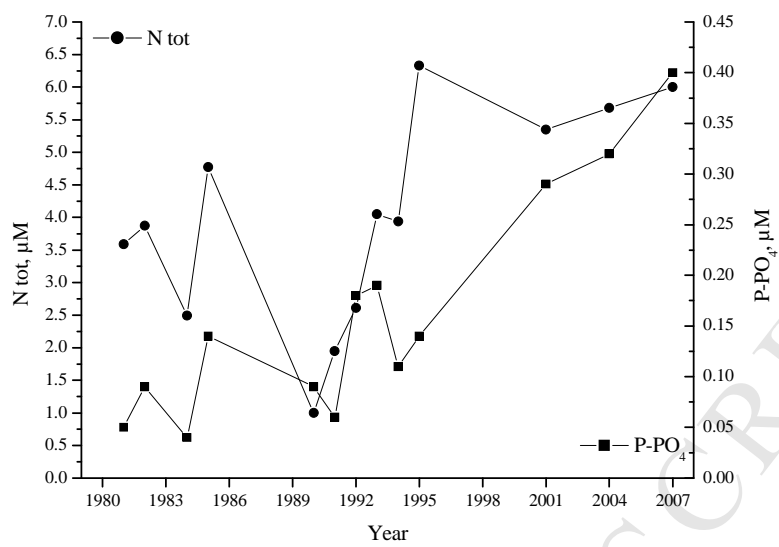


Figure 2

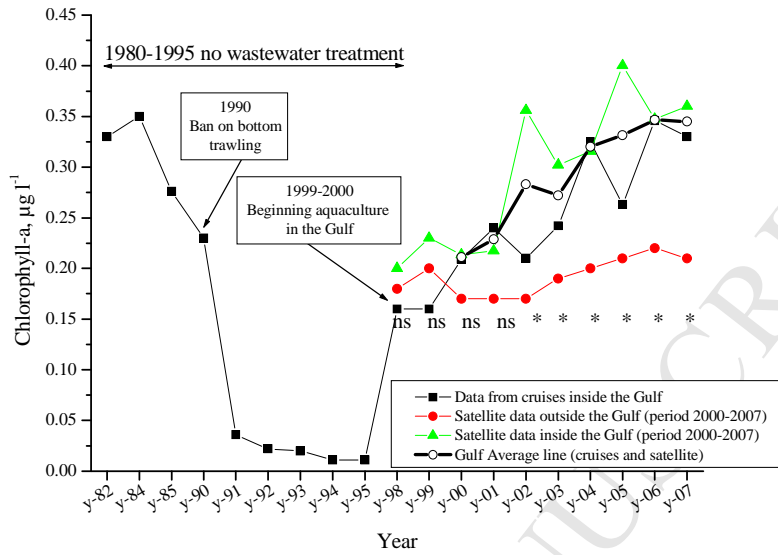
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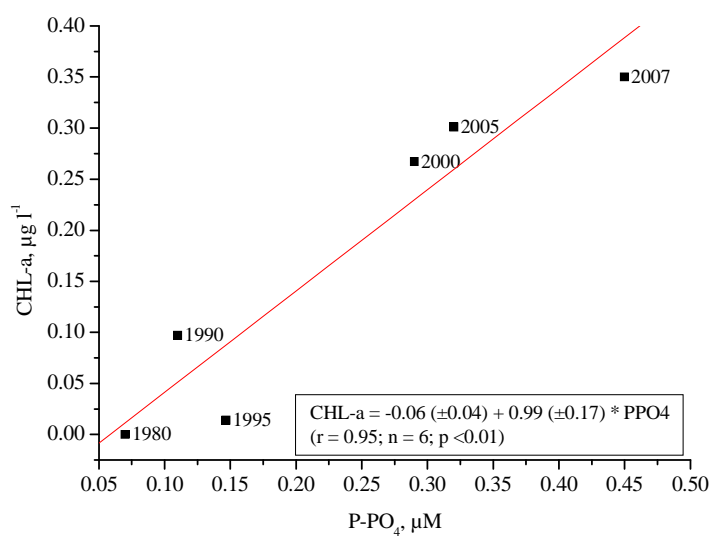
Figure 3

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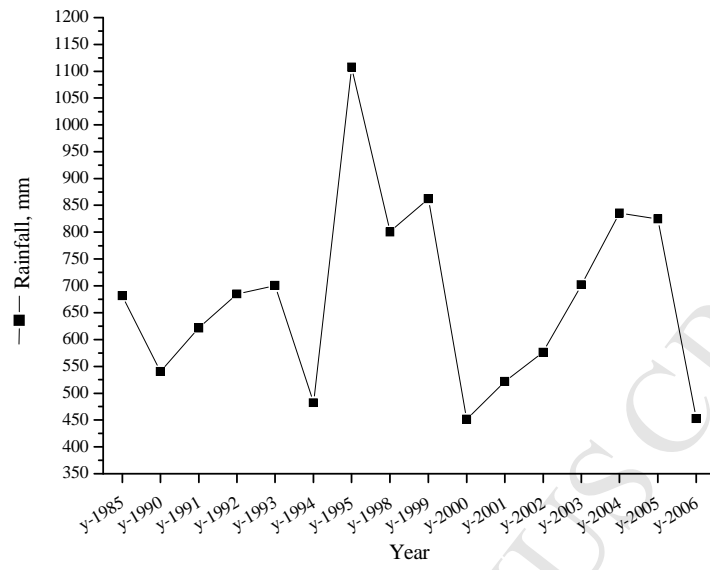
Figure 4



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Figure 5

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Figure 6