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UTJECAJ ZMORCA NA ONEČIŠĆENOST ZRAKA NA SPLITSKOM PODRUČJU

THE INFLUENCE OF SEA BREEZE ON THE AIR POLLUTION IN THE SPLIT AREA

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U radu se istražuje utjecaj zmorca na koncentraciju NO_2, SO_2, PM2.5 i PM10 na splitskom području prema mjernim postajama Split-centar i Kaštel Sućurac. Za proučavanje su izdvojeni dani u kojima je obalna cirkulacija bila neporemećena. U izdvojenim danima polje tlaka zraka bilo je bezgradijentno. Posebno su istraženi čestina i brzina vjetra u takvim danima. Zmorac u Splitu ima SW smjer, a u Kaštel Sućurcu WSW i W. Koncentracije onečišćujućih tvari uspoređene su s brzinama vjetra. Najprisutnija onečišćujuća tvar je NO_2. Najviše ga ima u večernjim i jutarnjim satima dok zmorac još nije razvio veću brzinu ili mu se brzina smanjuje približavanjem večernjeg zatišja. Što je brzina zmorca veća, koncentracija NO_2 je manja. Nastupom zmorca povećava se koncentracija lebdećih čestica zbog turbulencije vjetra unutar grada te mogućeg donosa morskog aerosola i nusprodukata pomorskog prometa. NO_2 dostiže veće vrijednosti u Splitu nego u Kaštel Sućurcu. Lebdeće čestice imaju veće koncentracije u Kaštel Sućurcu nego u Splitu. Vrijednosti svih onečišćujućih tvari ispod su graničnih vrijednosti za zdravlje ljudi.

KLJUČNE RIJEČI: zmorac, obalna cirkulacija zraka, onečišćenost zraka, lebdeće čestice

The paper investigates the impact of sea breeze on the NO_2, SO_2, PM2.5 and PM10 concentrations in Split area towards the Split-centre and Kaštel-Sućurac measurement stations. The study included the days when the coastal circulation was undisturbed. In all isolated days, the mean sea level pressure field was with low gradients. In particular, the frequency and wind speed on selected days have been explored. The sea breeze in Split has SW direction, and in Kaštel-Sućurac WSW and W direction. The concentrations of pollutants are compared to wind speeds. The most polluting substance is NO_2, most of it is in the evening and in the morning when the sea breeze has not yet developed the speed or its speed decreases by approaching the evening's sea breeze lull. As the sea breeze speed increases, the concentration of NO_2 decreases. As the sea breeze occurs, the concentration of floating particles increases due to wind turbulence within the city, and the possible release of sea aerosols and by-products of sea traffic. NO_2 reaches higher values in Split than in Kaštel-Sućurac. Floating particles are present in Kaštel-Sućurac than in Split. The values of all pollutants are below the limit values for human health.

KEY WORDS: sea breeze, coastal air circulation, air pollution, floating particle

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INTRODUCTION

The coastal air circulation is caused by the local differential warming and cooling of land and sea at a relatively small area. It represents a daily-periodic system of the winds of the coastal area the land breeze and sea breeze. For their shifts the air temperatures over the sea and the mainland are equalized (TROŠIĆ, 2002; TROŠIĆ ET AL. 2006). The sea breeze appears during daytime in the lower part of the coastal circulation, and it flows from the sea to the land. Its influence along the eastern coast of the Adriatic depends largely on local topography (FILIPČIĆ, 1994; PERICA, OREŠIĆ, 1997; FILIPČIĆ, 1999). The sea breeze on the eastern Adriatic coast begins to blow between 9 and 11 h local time, and it reaches its maximum at about 14 h local time, because then the temperature difference between the mainland and the sea is the highest, i.e. the inclination of isobaric surfaces is the greatest (ŠEGOTA, 1976, ŠEGOTA, FILIPČIĆ, 1996). Coastal circulation reaches the height of 500 to 800 m (BRITVIĆ, 1990; MARIĆ, 1998). In tepid latitudes coastal circulation is most frequent in the warmer part of the year. On the synoptic scale, sea breeze is characterized by a slightly elevated air pressure field, and the sea breeze occurs during stable weather conditions (PANDŽIĆ, LIKSO, 2005). The Etesian winds are also frequent then, which should be distinguished from the local coastal air circulation. Depending on the position of the coast and the islands, the Etesian winds can increase or decrease the sea breeze (LUKŠIĆ, 1995; BENCETIĆ KLAIĆ ET AL., 2009). Many factors, such as the synoptic conditions and the position of the shore, can influence the sea breeze development (ESTOQUE, 1962; PIELKE, 1974; ARRITT, 1989; ARRITT, 1993; GRISOGONO ET AL., 1998). This is indicated by many climatological studies, as well as the impact studies of the local topography (PENZAR, 1977; ORLIĆ ET AL., 1988; LUKŠIĆ, 1989; LUKŠIĆ, 1995; PENZAR ET AL., 1996; LUKŠIĆ, 2000-2001) on Croatian part of the Adriatic coast.

Knowing the coastal air circulation is important for many reasons. It is important in organized fire protection, sailing, fishing, etc. The important component of bioclimate is a sense of thermal comfort that depends on wind, air humidity, etc. (PENZAR ET AL., 1996). Favourable bio-climatic conditions during the sea breeze are important for life in the coastal area and appeal to tourists because the sea breeze alleviates the summer heat.

The monitoring of the sea breeze is especially important in protecting the environment from air pollution. Pollutant transfer studies show that pollutants enter the coastal air circulation and then circulate within the coastal circulation cell (LYONS, OLSSON, 1973; KEEN, LYONS, 1978; SIMPSON, 1994; OKE, 1987). For the diffusion of pollutants within the sea breeze, the

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topography of the coastal area, the shape of the coast, the position of industrial plants, etc. are of great importance (PIELKE ET AL., 1983; GROSSI ET AL., 2000). The major source of air pollution is traffic, that is, exhaust emissions from motor vehicles, which is a major problem in big cities. In the past, it used to be Athens and Los Angeles (SIMPSON, 1994), and today the Asian cities are dominant.

Pollutants are spread by airflow. Air pollution can be in the form of gas, vapour or aerosol of a variety of chemical composition and can also be chemically modified due to photochemical or catalytic reactions (PENZAR ET AL., 1996). Nitrogen and sulfur oxides are key components from which various chemical reactions in the atmosphere generate numerous compounds that have an undesirable effect on the ecosystem. Pollution transfer studies have shown that the pollution in the cities is highest during small wind speeds or sea breeze lows, more in the mountain lee than in windward, and is favoured by weak mechanical turbulence and thermal inversion (ŠEGOTA, 1976). The same conclusions were obtained by I. Bešlić et al. (2004) for concentrations of PM10 and PM2.5 floating particles in the residential part of Zagreb. T. Trošić Lesar and A. Filipčić (2017), for example, successfully predicted the mean hourly change of the PM10 concentration using the multiple linear regression model for the sea breeze cases in Split and Kaštel-Sućurac.

In addition to the pollution during the day, a big problem is the change of the direction of coastal circulation in the evening when the heavily polluted air returns to the same area (SIMPSON, 1994). A. Clappier et al. (2000) show that pollution in Athens is much higher than in Los Angeles due to the position of industrial plants in the wider area and due to the local orography. M. G. Evtyugina et al. (2006) studied the sea breeze along the coast of Portugal and concluded that inland areas also show higher levels of pollution than the densely populated industrialized areas of towns and villages near the coast due to the transmission of pollutants throughout the day. K. Bouchlaghem et al. (2007) measured the SO_2 concentrations on the coast of Tunisia. The results show that SO_2 is adversely affected by sea breeze, and that the maximum concentrations of SO_2 coincide with the time of the morning sea breeze lull.

The exploration of coastal circulation is of great importance because of the rugged coastline. This relates to the parts of the coast where we find potential sources of pollution. This relates also to the Split area; therefore it will be the subject of research in this paper. Split city region has prominent relief borders: the coastal region of Zagora is divided by mountains like a wall: Rilić, Biokovo, Mosor, Kozjak, Boraja and Trtar. Split is located in central Dalmatia on the Split (Marjan) peninsula (Fig. 1). The area of the town with Solin can be divided into the

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Split peninsula defined by the low hill Marjan (175 m), and Split-Klis hilly area shaped in the flysch and the area of Kaštela bay (45 m deep). For decades, the Split region has relied heavily on industry, transport, shipbuilding, tourism and agriculture. Kaštela are well known by their industrial zone with the largest potential sources of pollutants. Concentrations of these substances in relation to the the coastal air circulation have not been analysed so far in Croatian literature.

DATA AND METHODS OF RESEARCH

The paper analyses the data on concentrations of pollutants for the selected sea breeze cases in the cities of Split and Kaštela. This will enable a better insight into the daily transfer of pollutants in this area. The impact of coastal circulation on pollutant concentrations at selected stations, as well as in the area of Kaštela Bay will be studied. Until now, it has not been a subject of research, and because of the proximity of the industry it can have large impact on air pollution also in the wider area.

The data available for studying the concentrations of pollutants during the sea breeze are from Split-centre station in Split and Kaštel-Sućurac station in the town of Kaštela. The primary purpose of the station Split-centre is to observe pollution levels as a result of traffic, but also of the industry. There are family houses and smaller industrial facilities in its vicinity. The air temperature, wind speed and direction data for Split-centre were taken from the Split-Marjan station, and for the station in Kaštel-Sućurac from the station itself where the level of pollution is measured. The wind speed sensor is placed at a height of 10 m from the ground.

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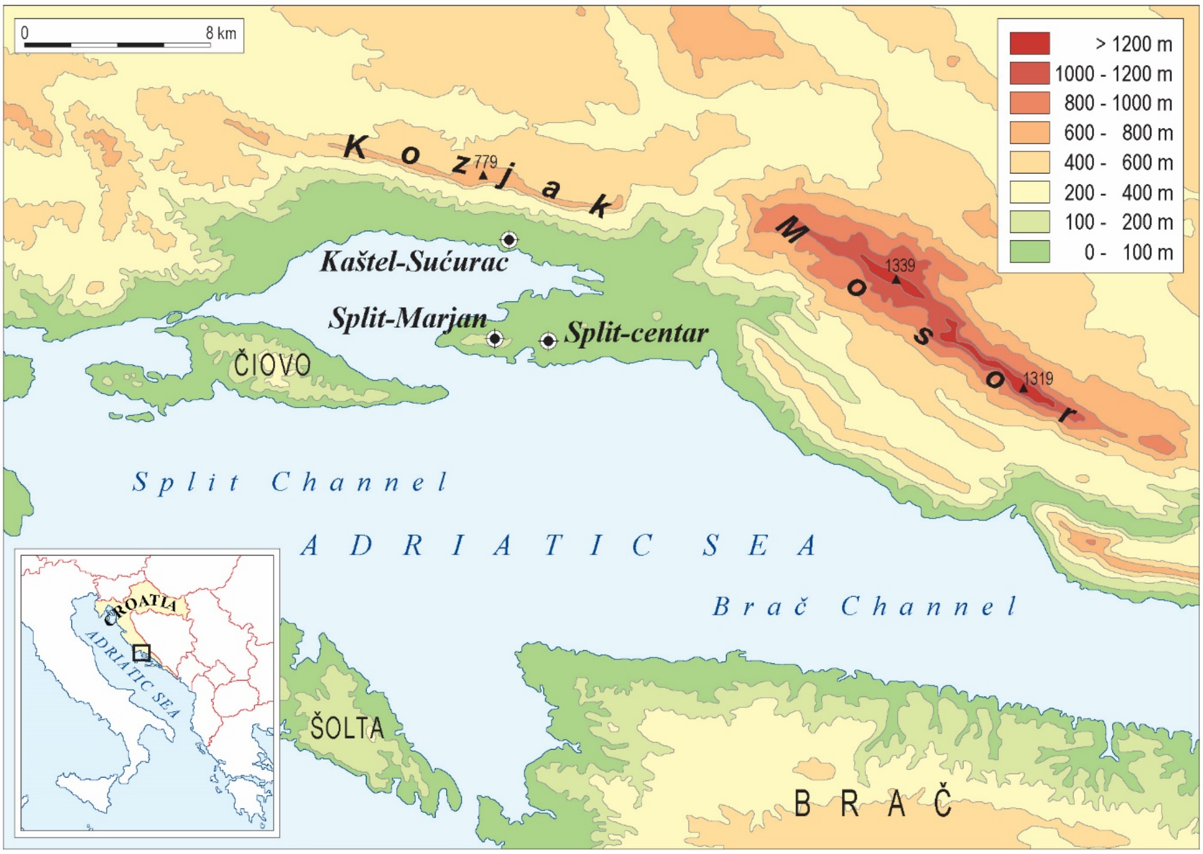

Figure 1 *The position of the stations for the pollutant measurements Kaštel-Sućurac and Split-centre, as well as the position of the meteorological station Split-Marjan (cartographic processing: I. Rendulić)*

Pollutant concentration data, especially near industrial sites, are very scarce and often unavailable. This paper uses the data for the period 2007-2009. Although this period is not long in the climatological sense and it does not apply to the recent years, it is sufficient to observe the general rules. Available pollutant measurements were the hourly concentration measurements of NO_2, SO_2 and floating particles PM10 and PM2.5. Nitrogen monoxide (nitrogen (II) oxide, NO) is a colourless and odourless gas produced during fuel combustion at high temperatures. After mixing with air, it quickly reacts with oxygen to produce nitrogen dioxide NO_2. The nitrogen dioxide disintegrates by absorption of sunlight to form the NO molecule, and the oxygen atom reacts with oxygen and gives the ozone molecule. The whole reaction is repeated while there is sunlight. As a consequence, a photochemical smog is created and the ozone concentration is increased in the lowest layer of the atmosphere. The concentration of nitrogen dioxide and nitrogen monoxide in the air is determined by chemiluminescence using the method HRN EN 14211: 2005. The chemiluminescence method

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is based on the reaction of ozone and nitric oxide which then oxidizes to NO_2. At the Split-centre and Kaštel-Sućurac stations, the ML 9841B analyzer is used with a high precision measurement over a large temperature range with a 0.5 ppb instrument accuracy.

Sulfur dioxide is generated by the burning of fossil fuels (coal and oil) and the melting of mineral raw materials containing sulfur, in industrial processes and road traffic. Volcano eruptions are the largest natural source of sulfur dioxide. The consequence of the presence of sulfur oxides in the atmosphere is the acid rain, or is the occurrence of sulfuric (sulfuric) acid. It affects the respiratory system and can cause eye irritation and exacerbates illnesses such as asthma and bronchitis (PENZAR ET AL., 1996). Sulfur dioxide measurement is performed using a UV fluorescence method using a ML9850B instrument that is reliable for a large temperature range with an instrument accuracy of 0.5 ppb.

The term "floating particles" refers to PM10 and PM2.5 particles (PM is the abbreviation for particulate matter). PM10 denotes particles of 10 μm or less in diameter, while PM2.5 have 2.5 μm diameter or less. Primary particles (PM10) are mainly generated by combustion of motor vehicle fuels, and combustion in non-industrial fireplaces i.e. small fireplaces in households. Because of their ability to penetrate the respiratory tract they can affect people's health (BEŠLIĆ ET AL., 2004). By entering the organism the particles are retained in various parts of the respiratory system, causing inflammatory changes, reduced resistance to infections and various allergies. The depth of penetration and the amount of decomposed particles depend on the particle size and the breathing mode. Secondary particles (PM2.5) can affect the emergence of serious pulmonary and vascular diseases (PENZAR ET AL., 1996). Total floating particles are measured by a high-volume sampling method using a Verewa F701 analyser, with readings of every 15 min. The measurements are carried out in such a way that particles are drawn over the filters, and the volumetric method determines their flow. The particles are then stopped on the filter and radiometric measurements using Beta-emitter (C-14) and Geiger-Müller counter are made. The measurement is based on the fact that beta-radiation is weakened by passing through the substance.

For the investigation of the impact of sea breeze on concentrations of pollutants to be relevant, it was necessary to separate the days in which the coastal circulation was undisturbed. In such days, in the morning hours, there is a visible change of wind direction from the land to sea breeze, after the appearance of morning lull. In the evening, after the appearance of sea breeze lull, the wind direction changes from the sea breeze toward land breeze. The difference in the air temperature between the morning and evening lull was not allowed to be higher than

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1 °C (TROŠIĆ, 2002), and mean daily cloudiness during climatological conditions during the day was not allowed to exceed 4/10 (TELIŠMAN PRTENJAK, GRISOGONO, 2002). The synoptic situations were reviewed at 12 UTC, which can be seen on maps of the German Meteorological Service (Europäischer Wetterbericht, 2007-2009). In all isolated days, the mean sea level pressure field was with low gradients. Above southern Europe there were no closed baric systems, and the pressure was slightly elevated, 1014-1020 hPa. On the upper level charts the pressure field was with low gradients with a weak geostrophic wind (up to 10 m s^{-1}).

In the period 2007-2009, 41 such days were selected in Split (tab.1) and 58 days in Kaštel Sućurac (tab.2). In these days, concentrations of pollutants were also simultaneously measured. In the year 2007, there were no available measurements of all pollutants in Kaštel-Sućurac, so the number of selected days is equal to zero in all months.

Table 1 Number of days with undisturbed coastal circulation in Split

Godina	**Travanj**	**Svibanj**	**Lipanj**	**Srpanj**	**Kolovoz**	**Rujan**	**Ukupno**
Year	**April**	**May**	**June**	**July**	**August**	**September**	**Total**
2007.	7	0	2	5	2	1	17
2008.	0	3	3	5	0	0	11
2009.	0	4	2	3	2	2	13
Ukupno Total	7	7	9	13	4	3	41

Table 2 Number of days with undisturbed coastal circulation in Kaštel-Sućurac.

Godina	**Travanj**	**Svibanj**	**Lipanj**	**Srpanj**	**Kolovoz**	**Rujan**	**Ukupno**
Year	**April**	**May**	**June**	**July**	**August**	**September**	**Total**
2007.	0	0	0	0	0	0	0
2008.	0	5	3	1	0	1	10
2009.	1	11	6	9	11	10	48
Ukupno Total	1	16	9	10	11	11	58

For the selected days the frequency and velocity of the wind were investigated (Figures 2-5). At the station Split-Marjan, the sea breeze has a SW direction and the wind speed is from 2-3 ms^{-1}. In Kaštel-Sućurac, sea breeze comes from WSW and W, and reaches the speed of about 2 ms^{-1}. The hodograph (Fig. 5) has an ellipsoid shape, which is characteristic for channelled flow.

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The direction of the sea breeze in Kaštel-Sućurac can be explained by the position of the station in the Kaštela Bay. In addition, the nearby islands also may influence the direction of the sea breeze. Namely, the position and shape of the bay and its relation to the position of the island and the coast can significantly influence the development of the sea breeze (GRISOGONO ET AL., 1998).

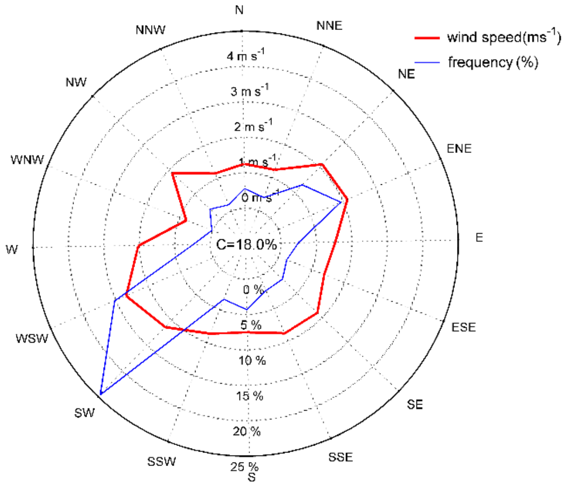

Figure 2 *Wind rose for selected days with undisturbed coastal circulation from 2007 to 2009 at the meteorological station Split-Marjan (from the hourly measurements of wind speed and direction)*

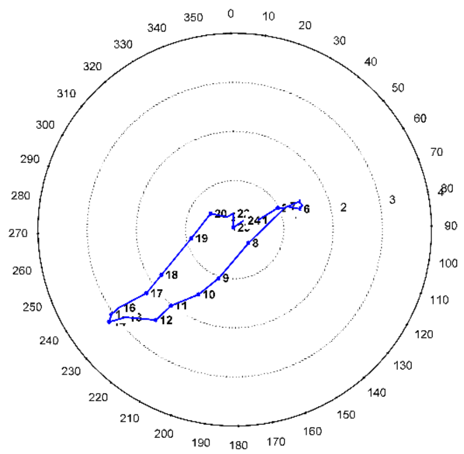

Figure 3 *Hodograph of wind for selected days with undisturbed coastal circulation from 2007 to 2009 at the meteorological station Split-Marjan. The circles denote the wind speeds (1-4 m s^{-1})*

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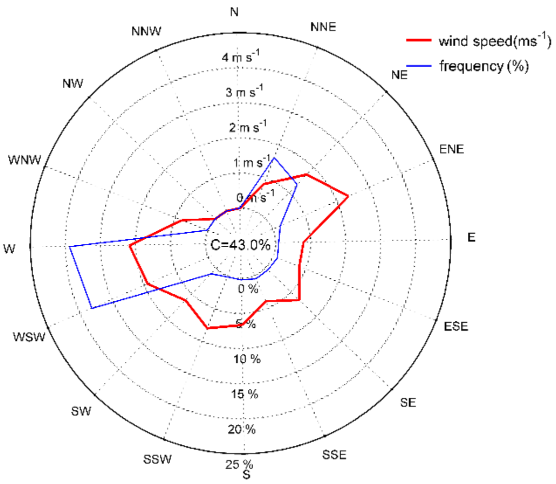

Figure 4 *Wind rose for selected days with undisturbed coastal circulation from 2007 to 2009 at the meteorological station Kaštel-Sućurac (from the hourly measurements of wind speed and direction)*

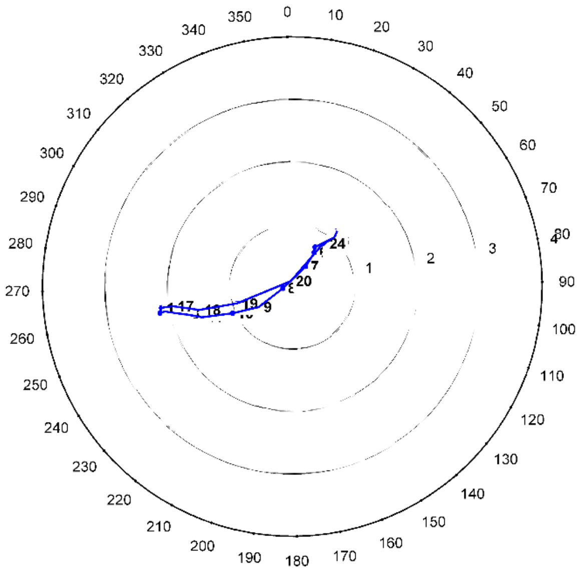

Figure 5 *Hodograph of wind for selected days with undisturbed coastal circulation from 2007 to 2009 at the meteorological station Kaštel-Sućurac (from the hourly measurements of wind speed and direction). The circles denote the wind speeds (1-4 m s^{-1})*

RESULTS AND DISCUSION

Data on velocity and the amount of wind in specific synoptic situations and the data of simultaneous air pollution measurements enabled the determination of the interdependence of

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these variables. At the Split-center station (Fig. 6), the most polluting substance is NO_2. The mean daily NO_2 concentration has the highest values in the evening and in the morning. These are the times when the wind speed is lowest because the sea breeze has not yet developed or its speed is reduced by approaching the evening's sea breeze lull. Road traffic and the position of the station in relation to the town morphology contribute to high NO_2 concentrations. As the sea breeze speed increases, the concentration of NO_2 decreases. This corresponds to the general rule that the wind "dilutes" the concentration of pollutants. However, at the same time with the strengthening of the sea breeze, the concentrations of floating particles are increased. This means that these particles are partly worn by sea breeze from the ground due to turbulence within the urban environment. Similarly, the exploration of the origin of floating particles in Rijeka (IVOŠEVIĆ ET AL., 2016a, IVOŠEVIĆ ET AL., 2016b) showed, among other things, the presence of sea aerosols and combustion products in marine traffic. These particles can be carried by sea breeze to the shore. Average concentrations of pollutants in the evening rise due to the return of contamination and its retention at the time of the eare visible, when the polluted air returns to the area of the city.

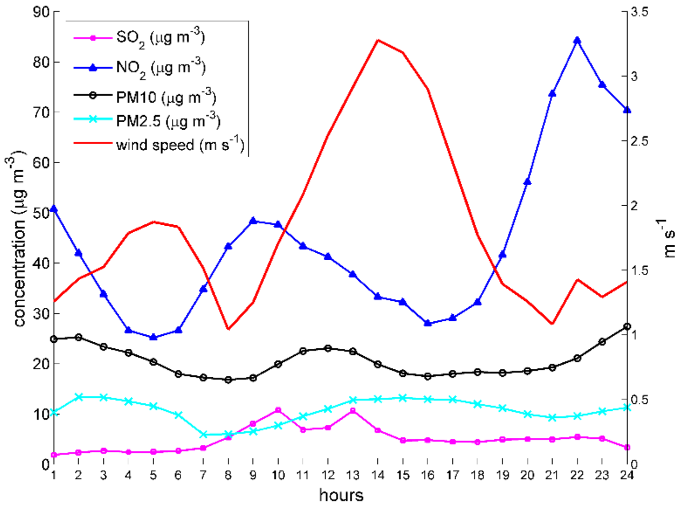

Figure 6 *The mean measured hourly values of nitrogen dioxide concentration (NO_2), sulfur dioxide (SO_2) and floating particles PM10 and PM2.5 in µg m^{-3} at the Split-centre station, and wind speeds at the Split-Marjan station in days with undisturbed coastal circulation*

The Kaštel-Sućurac station is located near the Adriatic highway, which also increases the concentration of NO_2 (Fig. 7). The highest concentration of NO_2 is achieved during the morning sea breeze lull. Even a small increase in the speed of the sea breeze reduces the NO_2

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concentration, but, as in Split, it increases the concentration of floating particles. In the periods of lull between the sea breeze and the land breeze the amount of floating particles is smaller. It may be noted that the land breeze during the night also reduces the concentrations of pollutants, but this impact is not as visible as in the case of a daytime sea breeze, because the absolute emissions of pollutants are greater during the day than over night.

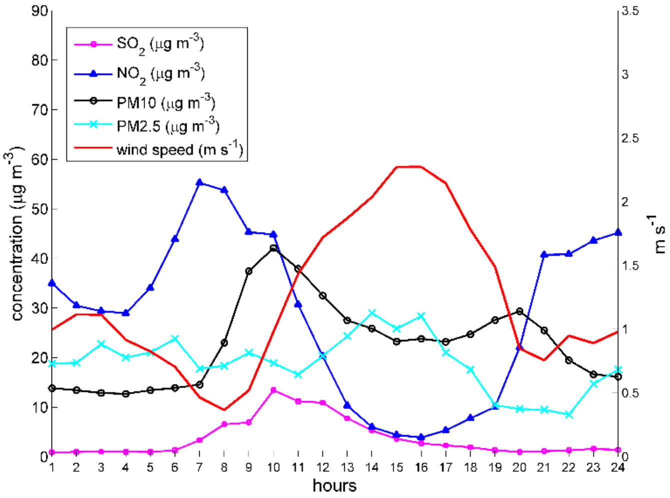

Figure 7 *The mean measured hourly values of nitrogen dioxide concentration (NO_2), sulfur dioxide (SO_2) and floating particles PM10 and PM2.5 in μg m^{-3} and wind speed at the Kaštel-Sućurac station during days with undisturbed coastal circulation*

The maximum daily value of the NO_2 concentration at the station Split-centre is 150.93 μg m^{-3}, and in Kaštel-Sućurac it is slightly lower, 109.95 μg m^{-3}, which are below the limit value for human health which is 200 μg m^{-3}. The maximum mean daily value is also below the average daily limit value for human health (80 μg m^{-3}).

Table 3 The borderline values (URL 1), 24-h mean values, as well as the polutant particle maximum daily values during sea breeze (NO_2, SO_2, PM10 and PM2.5) at the stations Split-centre and Kaštel-Sućurac on selected days with undisturbed coastal circulation from 2007 to 2009

	NO_2 (μg m^{-3})	SO_2 (μg m^{-3})	PM10 (μg m^{-3})	PM2.5 (μg m^{-3})
Granična vrijednost za 24-h srednjak **Borderline value for 24-h mean**	200	125	50	-
Granična vrijednost za 24-h maksimum **Borderline value for 24-h maximum**	80	350	-	-

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Split-centar		NO_2 (μg m^{-3})	SO_2 (μg m^{-3})	PM10 (μg m^{-3})	PM2.5 (μg m^{-3})
2007. – 2009.	**24-h srednjak** **24-h mean**	66,40	20,11	40,96	108,85
	24-h maksimum **24-h maximum**	150,93	198,08	60,16	500,62
Kaštel Sućurac		NO_2 (μg m^{-3})	SO_2 (μg m^{-3})	PM10 (μg m^{-3})	PM2.5 (μg m^{-3})
2007. – 2009.	**24-h srednjak** **24-h mean**	62,84	12,49	38,93	189,79
	24-h maksimum **24-h maximum**	109,95	73,79	139,29	501,23

The PM10 concentrations are slightly below the prescribed limit value for human health for a 24-hour mean of 50 μg m^{-3}, and in Split, for example, the maximum daily value is 40.96 μg m^{-3}, which is almost the limit value, while in Kaštel Sućurac it is smaller and has the value of 38.93 μg m^{-3}. The possible reason for this is larger retention of the PM10 particles in the city, due to the position of orography in the hinterland of Split as well as the possible partial transfer from the area of Kaštela when the particles from the Kaštela area entered the coastal circulation towards the city, where they also recorded the maximum daily values.

Mean daily concentrations of SO_2 were 20.11 μg m^{-3} in Split and 12.49 μg m^{-3} in Kaštel-Sućurac, which are far below the prescribed human health limit value of 125 μg m^{-3}. The maximum daily value of the SO_2 concentration in Split and Kaštel Sućurac is also well below the prescribed limit for human health (350 μg m^{-3}). In Split, the maximum daily value of SO_2 was 198.08 μg m^{-3}, and in Kaštel-Sućurac 73.79 μg m^{-3}. PM2.5 particles have no prescribed daily mean and maximum values, and these values are not comparable. Of all analysed pollutants, only NO_2 with relatively high concentration showed statistical significance. Therefore, the correlation of NO_2 and wind speeds (Fig. 8) is shown separately in the example of Kaštel-Sućurac.

Student's t-test showed the correlation significance ($R = -0.69$) of this pollutant with wind speed at the significance level of $\alpha = 0.01$. The maximum concentrations of nitrogen dioxide were observed with an increase in wind speed. The decrease is slower for lower wind speeds of up to 1.5 m s^{-1}, and then it increases rapidly. The reason for this is that at higher speeds ventilation is more efficient.

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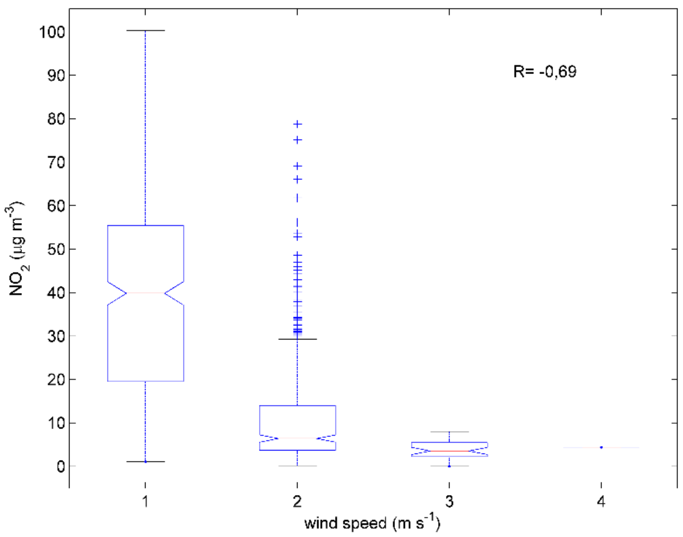

Figure 8 *Box plot, NO_2 concentration-wind speed. The x axis is divided into groups. The top and bottom rectangle show a 75% and 25% percentile, and the red line in the rectangle shows the median. Horizontal lines outside the rectangle show the largest (upper) and lowest (down) data within the M ± 1.5 IR interval where M is a median and an IR interquartile range. Crosses above the rectangle show data greater than M + 1.5 IR and below less than M-1.5 IR*

Pollutant transfer studies in cities have shown that the highest levels of pollution are during low wind or silence, more in the lee than in windward, and are favoured by inversion and poor mechanical turbulence (ŠEGOTA, 1976). This is in agreement with the study of the sea breeze cases in Kaštel-Sućurac, where the highest NO_2 concentrations are obtained at the lowest speeds with a relatively high correlation coefficient of -0.69.

CONCLUSION

The coastal air circulation is caused by local differentiated heating, i.e. by cooling of land and sea at a relatively narrow area. The surface air temperature has a significant effect on the heat flow between the atmosphere and the sea, as well as the properties and the motion of the air. In order to minimize adverse atmospheric impacts on the development of the sea breeze, on the basis of available measurements, the days with undisturbed coastal air circulation are selected. In order to study the concentrations of pollutants during the sea breeze, the available data used were from the measurement stations Split-centre in Split, and Kaštel-Sućurac in

Kaštela. Measurements included concentrations of SO_2, NO_2, and floating particles PM10 and PM2.5.

Sea breeze in Kaštel Sućurac has WSW and W direction, while in Split it has SW direction. The NO_2 dependence on the sea breeze speed showed a decrease in its concentration with an increase in wind speed. However, the concentrations of PM10 and PM2.5 increase with the development of the sea breeze. This is contributed by the wind turbulence within the city as well as the transfer of sea aerosols and combustion products in maritime traffic. The pollutants from Split and the Kaštela area, carried by the sea breeze, , do not have the ability to move further inland due to the orographic characteristics of the hinterland.

NO_2 concentrations are higher in Split than in Kaštel-Sućurac. The PM10 and PM2.5 concentrations were significantly higher in Kaštel-Sućurac than in Split, as a result of industrial pollution, but they were below the prescribed 24-h limit and tolerant values for human health. The results point to a great importance of exploration of coastal circulation, as well as to the need of a larger number of measuring stations and pollutant concentrations measurements, especially in cities and near industrial plants.

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