Environmental Science
Processes & Impacts

RSC Publishing

PAPER

View Article Online
View Journal

Cite this: DOI: 10.1039/c3em00228d

Ultrafine particle concentrations in the surroundings of an urban area: comparing downwind to upwind conditions using Generalized Additive Models (GAMs)

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The aim of this study was to investigate the influence of an urban area on ultrafine particle (UFP) concentration in nearby surrounding areas. We assessed how downwind and upwind conditions affect the UFP concentration at a site placed a few kilometres from the city border. Secondarily, we investigated the relationship among other meteorological factors, temporal variables and UFP. Data were collected for 44 days during 2008 and 2009 at a rural site placed about 3 kilometres from Bologna, in northern Italy. Measurements were performed using a spectrometer (FMPS TSI 3091). The average UFP number concentration was 11 776 (±7836) particles per cm^3. We analysed the effect of wind direction in a multivariate Generalized Additive Model (GAM) adjusted for the principal meteorological parameters and temporal trends. An increase of about 25% in UFP levels was observed when the site was downwind of the urban area, compared with the levels observed when wind blew from rural areas. The size distribution of particles was also affected by the wind direction, showing higher concentration of small size particles when the wind blew from the urban area. The GAM showed a good fit to the data ($R^2 = 0.81$). Model choice was *via* Akaike Information Criteria (AIC). The analysis also revealed that an approach based on meteorological data plus temporal trends improved the goodness of the fit of the model. In addition, the findings contribute to evidence on effects of exposure to ultrafine particles on a population living in city surroundings.

Received 6th May 2013
Accepted 5th September 2013

DOI: 10.1039/c3em00228d

rsc.li/process-impacts

Environmental Impact

This work adds to the current scientific knowledge in terms of environmental processes, giving an important contribution to understanding of the mechanisms of the source, movement and distribution of ultrafine particle (UFP) concentration in the environment. We assessed how downwind and upwind conditions affect UFP concentrations at a site placed a few kilometres from an urban area, and also determined the effect of other meteorological parameters and temporal variables on UFPs. We adopted an approach based on the analysis of the effect of wind direction in a Generalised Additive Model (GAM). This model represents a development in the environmental research field. In addition, the findings contribute to evidence on effects of exposure to ultrafine particles on a population living in city surroundings.

1. Introduction

In the last decade some studies have investigated the effects of meteorological variables on ultrafine particle (UFP, diameter <100 nm) concentrations using statistical models that simultaneously took into account various climatic factors, primarily temperature and wind speed.[1] However, literature about how to model and estimate ultrafine particles is still lacking, particularly in regard to wind direction. Recently, due to the high complexities of the relationship between UFP and meteorological parameters, Clifford *et al.* have proposed Generalised Additive Models (GAMs) as a suitable approach to this type of analysis.[2] Indeed the effects of meteorology on ultrafine particle number concentration (UFP-NC) may be non-linear. As such any attempt to model UFP-NC must acknowledge this potential non-linearity, ideally without making any *a priori* assumptions about the functional form of these non-linear effects (*e.g.* exponential or quadratic polynomial).

The increase of knowledge of such environmental processes has been always strictly connected with other research fields. Indeed, exposure to ultrafine particles is an important public

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health threat. Over the last few decades, a large number of epidemiological studies have reported convincing relationships between the exposure to particulate matter and health effects.[3] Beginning in the late 1990s, some studies discovered inverse associations between ultrafine particle levels and the respiratory health of people with asthma.[4] Subsequent studies showed an association between UFPs and various health outcomes such as mortality and respiratory and cardiovascular morbidity.[5] However, research efforts are still important to understand the mechanism of action by which small particles affect human health as well as population exposure patterns. UFPs have two types of sources: primary and secondary. A primary source of UFPs is the exhaust gas from combustion of fossil fuels, with a primary role of traffic in urban environments.[6] These particles are in the size range of 30–100 nm. Secondary sources generate UFPs by nucleation events due to chemical–physical reactions in the atmosphere, typically associated with the oxidation of atmospheric gas, which range from 1 to 10 nm, or by condensation of low volatility vapours emitted in traffic exhaust originating from vaporised engine oil in the combustion process, and are typically in the size range 10–30 nm.[7]

In close proximity to the source of emission, the size distribution of UFPs will depend on variables such as the type of combustion fuel, the speed and load of the vehicle, the speed at which pollutants are emitted from vehicles, and turbulence caused by vehicle motion.[8] However, the fluctuations of a variety of meteorological and emissions variables cause remarkable spatial and temporal variations of UFP concentrations measured away from the source of emission. The prevalent wind is the main factor determining the speed and direction of the UFP plume.

A number of studies have analysed the spatial variability of UFPs within urban areas. Cyrys *et al.* investigated the spatial variation of particle number concentrations at four background sites in Augsburg, Germany.[9] During a 10 day period in winter the site located in the suburb showed a concentration that was 25–30% lower than the three sites placed near the city centre. Similar results were found in spring even though with lower concentrations in all sites. Even larger differences were found by Hussein *et al.* who evaluated the differences in particle number concentration between a central traffic site and three suburban sites within the Helsinki urban area.[1]

A few studies have focused on comparing UFP concentrations measured at urban and rural areas tens of kilometres away. Laakso *et al.* found that the average total number concentrations decreased with distance from an urban area.[10] They measured the concentrations of UFPs of 16 600, 2100 and about 920 particles per cm^3 in the Helsinki urban area, in a rural area at a distance of about 100 km, and an Arctic background site, respectively. Stanier *et al.* have monitored UFPs at a central sampling site and at an upwind location (Florence, PA) at 38 km from the city centre for approximately 2 months.[11] These authors showed that number concentrations at the urban site were, on average, almost twice the concentrations measured at the rural site.

To our knowledge, no study has been focused on the effect of wind direction and conducted in the surroundings of urban areas, *i.e.* at distance of a few kilometres from the city border, to analyse whether and to what extent the concentrations of UFPs emitted in an urban area have a direct impact on UFP levels in its surroundings. However, this type of analysis is difficult because the relationship among UFPs, wind direction and meteorological variables is complex and highly non-linear.[12] For this reason, in this study we used a GAM model. The main objective was to assess how downwind and upwind conditions affect UFP concentrations at a site placed a few kilometres from the city border, but also to assess the effect of other meteorological parameters and temporal variables on UFPs. Secondarily, the aim of this study is to estimate the extent to which an urban area affects exposure of the population living in its surroundings.

2. Methodology

2.1. Study area

The city of Bologna is located in the midland of Emilia-Romagna (Fig. 1), a region in northern Italy where air quality limits are frequently exceeded.[13] The area is characterized by a

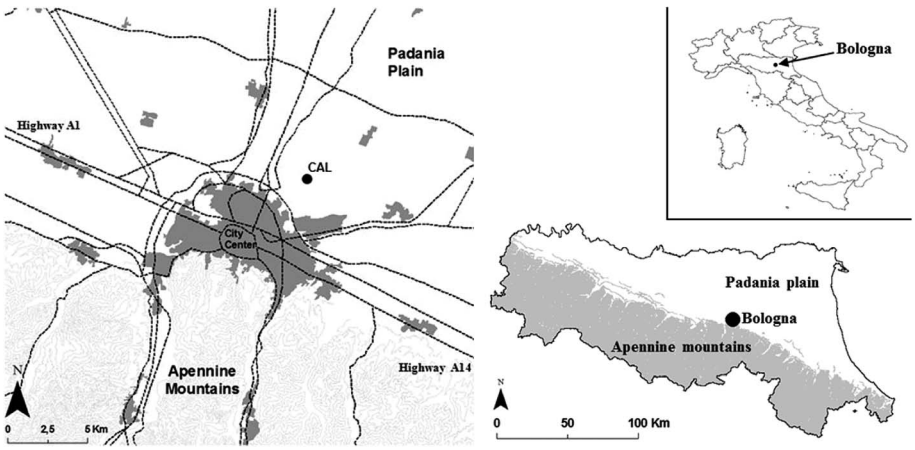

Fig. 1 Map showing the urban area of Bologna. The dotted lines represent the major high traffic roads.

sub-continental climate with cold winters and hot summers (typical monthly mean temperatures ranging from 1 to 26 °C), high humidity levels (typical monthly mean relative humidity ranging from 60% to 84%) and low wind intensities (typical annual mean wind intensities of about 2 m s^{-1}).

The city has approximately 370 000 inhabitants and is characterised by high traffic loads (black dotted lines in Fig. 1). Two major highways (A14 and A1) pass nearby and have a daily traffic volume of approximately 280 000 vehicles (of which 67 000 are heavy vehicles). In addition, there is the local traffic, estimated to be about 250 000 vehicles on a typical workday.

2.2. Measurement periods

The UFP measurement site (Calamosco, CAL) was located in a rural area about 6 kilometres from the city centre and approximately 3 kilometres north of the city boundaries. The distance to the closest road (low traffic) is about 150 meters. The site is not directly influenced by traffic or other relevant point sources. Three measurement periods took place between 4th to 15th July 2008, 13th to 30th 2009, and 24th February to 9th March 2009, with a total duration of 44 days.

The size distribution was monitored using a Fast Mobility Particle Sizer spectrometer (FMPS Model 3091-TSI). The size range was from 5.6 nm to 560 nm, classified in 32 evenly spaced logarithmic bins. The aerosol data were subject to quality assurance and the data validation was carried out. The FMPS was cleaned and maintained regularly by trained technicians and sent to the reference laboratory annually for calibration. For the present study, the original measurements at a time resolution of one minute were averaged on hourly basis. Measurements were made continuously 24 hours a day. UFP-NC was calculated by integrating the size distribution over the diameter range 10–100 nm.[2] The data of the first channels (from 5.6 to 9.96 nm) were excluded from the analysis because of the high percentage of observations under the detection threshold.

The hourly mean values of temperature, wind speed and direction, relative humidity, and solar irradiance together with the hourly cumulative rainfall were provided by the Hydro-Meteo-Climate Service of the Regional Agency for Prevention and Environment of Emilia Romagna for the period of study.

2.3. Generalised Additive Models

A Generalised Additive Model (GAM) is a multivariate method of analysis able to model the non-linear effects of a number of covariates, particularly capable of assessing any simultaneous and combined contribution to the response variable. The non-parametric form of the model is its main feature: it means that any non-linear relationship will be modelled flexibly without having to specify the non-linear functional form. Because of this, GAMs are commonly used in different research fields. This modelling approach is an extension of the Generalised Linear Model (GLM), where parametric regression terms are replaced by non-parametric functions such as scatter plot smoothers.[14] In this study the non-parametric regression is used to approximate the synergistic contribution of a number of covariates, such as wind direction, to the response variable (PNC) without making any *a priori* assumptions about the underlying processes or trends, which may be highly complex. Analyses were performed using the package mgcv in R (version 2.15.3, Copyright © 2013, The R Foundation for Statistical Computing).[15,16] Different GAM models were tested in this study. The available meteorological variables were: wind direction (wdrn), wind speed (wspd), temperature (temp), humidity (hum), solar radiation (solar), and rainfall (rain). We also tested temporal variables as hour of the day (h), day of the week (dow), monitoring period (period) and the one hour lagged logarithm of UFP (laguf1) number concentration (particles per cm^3). The forms of the models were:

$$\text{Model 1 } E(Y) = \beta_0 + f_1(\text{wdrn}) + f_2(\text{wspd}) + f_3(\text{temp}) \quad (1)$$

$$\text{Model 2 } E(Y) = \text{model 1} + f_4(\text{laguf1}) \quad (2)$$

$$\text{Model 3 } E(Y) = \text{model 2} + f_5(\text{h}) + f_6(\text{dow}) \quad (3)$$

$$\text{Model 4 } E(Y) = \text{model 3} + f_7(\text{hum}) + f_8(\text{solar}) \quad (4)$$

$$\text{Model 5 } E(\text{Y}) = \text{model 4} + f_9(\text{rain}) + f_{10}(\text{period}) \quad (5)$$

where Y is the logarithm of UFP-NC (particles per cm^3) and each of the f_i represent a smooth function to be fit as either thin plate regression splines or cyclic penalised B-splines,[17,18] with the exception of the day of the week and period, which were treated as a factor term. Table 1 shows the variables inserted in the models together with their base functions used in the GAMs. The Gaussian family distribution with an identity link function was used. The response variable was the logarithm of UFP-NC because previous studies have reported that concentrations of air pollutants (together with a number of environmental variables) are log-normally distributed.[19] Our data confirmed this hypothesis. The skewness and kurtosis test was used to assess the normality assumption of the log UFP-NC distribution ($p = 0.138$).

We set out to investigate the relationship between meteorological variables and UFPs. Our base model (model 1) included only the wind direction, wind speed, and temperature as independent variables. These are considered in several papers to be

Table 1 Predictor covariates and their base functions for GAMs used in eqn (1)–(5)

Variables	Covariate	Basis
Wdrn	Wind direction	cp[a]
H	Hour of the day	cp
Laguf1	log UFP-NC lag 1	tp[b]
Temp	Temperature	tp
Hum	Relative humidity	tp
Wspd	Wind speed	tp
Solar	Solar insolation	tp
Rainfall	Rainfall	tp
Dow	Day of the week	Factor
Campaign	Monitoring campaign	Factor

[a] Cyclic, penalised B-spline. [b] Thin plate regression spline.

the most important predictors for UFP-NC.[12] Subsequently, the one hour lagged UFP concentrations were included in the model (model 2), to take into account the high autocorrelation of UFP-NC ($R = 0.86$ at lag 1). As previously demonstrated by Clifford *et al.*, a significant improvement of the model has been achieved by inserting an autoregressive term.[2] In model 3 we considered also other temporal variables as hour of the day and day of the week. Humidity and solar radiation were added in model 4. Lastly, in model 5 we also tested the inclusion of rainfall and an indicator of the each specific period.

The choice of the best model was done *via* Akaike Information Criteria (AIC) and R^2.

We ran the GAM models using the R package mgcv. The package uses splines to create smooth functions that use the generalised cross-validation (GCV) technique to minimise both the model deviance and the bias. We adopted an automatic choice to determine the most appropriate parameters (degrees of freedom, knots) for the splines. Diagnostic checks on the models were performed using the gam.check procedure included in the package mgcv. In addition, the Partial Auto Correlation Function (PACF) was inspected to test for residual autocorrelation. We used Analysis of Variance (ANOVA) to test whether smooth terms in the fitted model were statistically significant.

3. Results

Table 2 shows the range of UFP concentrations and the observed meteorological variables measured in the rural site of Calamosco during the three monitoring periods. Similar results were found in the second and third periods with mean concentrations equal to 12 894 and 12 806 particles per cm^3. The concentrations measured during the first period in July 2008 were much lower (8801 particles per cm^3) than those found in January and March 2009 (periods 2 and 3). Rainfall was rare and scarce during the measurement periods, and all meteorological parameters showed values representative of their typical annual ranges. Fig. 2 (left panel) shows the "wind rose" during the monitoring periods. Wind tended to blow from the south-west with a peak at approximately 225°. Fig. 2 (right panel) shows strong analogies between the wind rose during the monitoring periods and the wind rose for the whole period of 1st of Jan 2008 to 31st of December 2009.

As a preliminary analysis we generated a scatter plot for the log UFP-NC and wind direction. As expected no particular trend or relationship was found between the two variables enforcing our choice of a non-parametric approach to model the relationship between the UFP-NC and the wind direction along with other predictors.

Table 3 shows a comparison of the performances of the models with regard to R^2 and Akaike Information Criteria (AIC). R^2 increased from 0.25 (model 1) to 0.81 (model 4 and 5). Model 4 showed the lowest AIC (only slightly lower than model 5) and it was chosen as the best model. The variables rainfall and campaign in model 5 were found not to be statistically significant while each term in the model 4 was statistically significant at a level of significance of 5% (p-values were <0.001).

Model 4 has been chosen as our best model, and the following graphs and figures are reported for this model only.

Normality of the residuals was tested with diagnostic plots (Fig. 3). The frequency histogram and the Q–Q plot showed a

Table 2 Descriptive statistics of environmental parameters (hourly data)

	N	Mean	SD	Min	25° perc.	Median	75° perc.	Max.
First period (4/7/2008 to 15/7/2008)								
UFP (number per cm^3)	256	8801	5122	1412	5332	7504	11 187	33 376
Temp (°C)	256	25.5	4.3	14.9	22.2	25.4	29.3	33.2
Hum (%)	256	52	14	24	40	52	64	87
Wspd (m s^{-1})	256	2.7	1.4	0.6	1.6	2.3	3.4	7.4
Wdrn (degrees)	256	206	72	5	164	205	265	350
Solar (W m^{-2})	256	318	335	0	0	198	652	917
Rain (mm h^{-1})	256	0	0	0	0	0	0	0
Second period (13/1/2009 to 30/1/2009)								
UFP (number per cm^3)	400	12 894	8183	2877	6897	10 913	16 210	60 894
Temp (°C)	400	4.2	2.1	−0.2	2.7	4	5.5	11.4
Hum (%)	400	87.6	7.5	59.5	85	89.5	93	96.5
Wspd (m s^{-1})	400	2.3	1	0.6	1.5	2.2	2.9	6.8
Wdrn (degrees)	400	235.8	80	8.5	197.1	269	285.5	349.5
Solar (W m^{-2})	400	35.8	71.4	0	0	0.8	43.5	401.7
Rain (mm h^{-1})	400	0.1	0.4	0	0	0	0	2.2
Third period (24/2/2009 to 9/3/2009)								
UFP (number per cm^3)	306	12 806	8591	3349	7487	10 140	15 478	62 842
Temp (°C)	306	8.1	3.3	−0.3	6.3	8.2	9.8	16.5
Hum (%)	306	74	19	27	60	80	90	95
Wspd (m s^{-1})	306	2.1	0.9	0.2	1.4	2	2.7	4.9
Wdrn (degrees)	306	188	93	9	102	197	267	358
Solar (W m^{-2})	306	92.6	160.1	0	0	0.8	100.2	624.9
Rain (mm h^{-1})	306	0.1	0.4	0	0	0	0	5.1

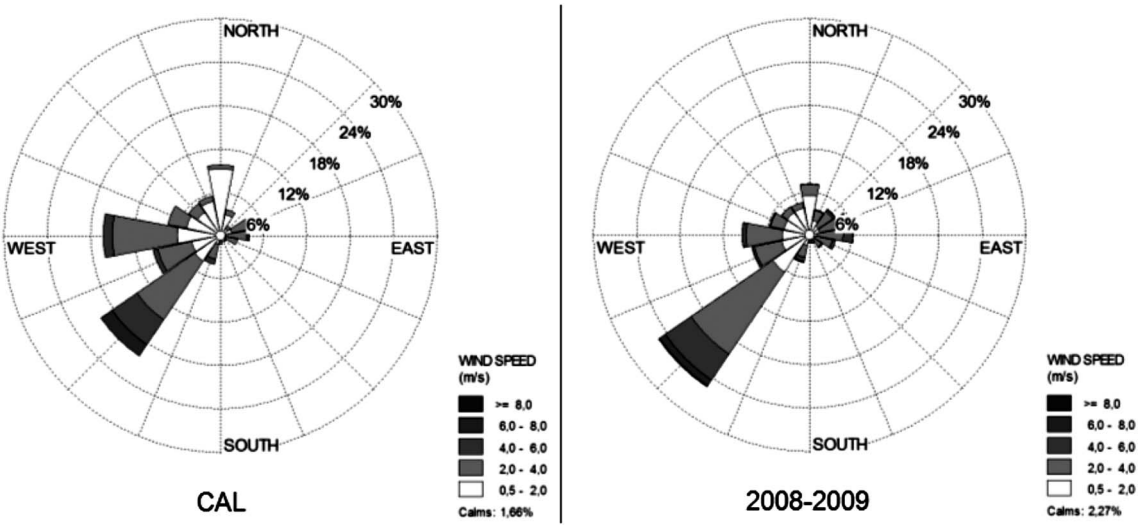

Fig. 2 Wind rose for the station of Calamosco (CAL) during the monitoring periods (left) and for the period Jan 2008 to Dec 2009 (right).

Table 3 AIC score and R^2 obtained from the generalised additive models

Outcome variable: log UFP number concentration	R^2	AIC
Model 1 = wind speed + wind direction + temperature	0.25	1508
Model 2 = model 1 + log UFP number concentration lag 1	0.76	389
Model 3 = model 2 + hour of the day + day of the week	0.80	287
Model 4 = model 3 + solar radiation + humidity	0.81	208
Model 5 = model 4 + rainfall + campaign	0.81	209

nearly normal distribution. High correlation was found between observed and fitted values ($R = 0.9$). The PACF analysis showed no residual autocorrelation (Fig. 4).

Fig. 5 shows the relationships between UFPs and each variable in model 4. Two marked daily peaks were present at 8:00 and 18:00 h, corresponding to the traffic rush hours. The relationship between UFPs and temperature was monotonically decreasing. Similar patterns were found with respect to relative humidity and wind speed. A U-shaped relationship was found for solar radiation. A strong positive relationship with UFP concentration the hour prior to measurement (the autoregressive term of UFPs) was evident. Lower UFP levels were observed on Sunday.

The relationships between UFPs and wind direction are described in more detail in Fig. 6. UFP levels were found to be highly affected by the wind direction. An increase in UFP concentrations of up to 25% was found when the urban area was upwind compared with when the wind came from the opposite direction. Minor changes in the results were found stratifying for each monitoring period.

Marked differences in the shape of the size distribution were also found with regard to wind direction. Fig. 7 compares the size distributions of particles when wind came from the urban area ($220 \pm 40^\circ$ N) and from the opposite direction ($40 \pm 40^\circ$ N), corresponding with clean rural areas. The size distributions were almost identical below 8 nm and above 100 nm. The shape of the size distribution between 8 nm and 100 nm was similar, although the specific characteristics of multimodality in each distribution differed in each group of data. The size distribution downwind of the urban area presented two marked peaks at 10–20 nm and a broad peak at 40–80 nm, which can be associated with the secondary formation of UFPs from condensation of semi-volatile gases emitted in the traffic exhaust and with UFPs directly emitted by traffic. On the other hand, the size distribution associated with cleaner areas showed a marked peak at 80–100 nm, which might be associated with background UFPs, and a small degree of primary UFPs associated with primary emissions by traffic.

4. Discussion and conclusions

Short term measurements of UFPs have been carried out in the surroundings of an urban area in northern Italy to verify whether and to what extent the urban area affects UFP concentrations and population exposure in areas several kilometres from the city borders. We adopted an approach based on the analysis of the effect of wind direction in a multivariate GAM model. The results showed a relevant increase in UFP levels at the rural site when the site was downwind of the urban area, compared with the levels observed when the wind came from non-urbanized areas.

The use of GAM played a key role in the analysis. Our findings confirmed the importance of the non-linear approach allowed by GAMs. In particular for wind direction and meteorological variables this modelling approach is needed. The relationships found in the GAM model between UFPs and the other predictors were generally in good agreement with the few studies based on multivariate techniques. Further to this, we have shown that the generalised additive model provided a good fit to the data (R^2 equal to 0.81). The GAM guarantees flexibility and permits the evaluation of the specific and synergistic contributions of different variables affecting UFP concentrations. We showed that a combined contribution of wind direction, wind speed and temperature explained 25% of

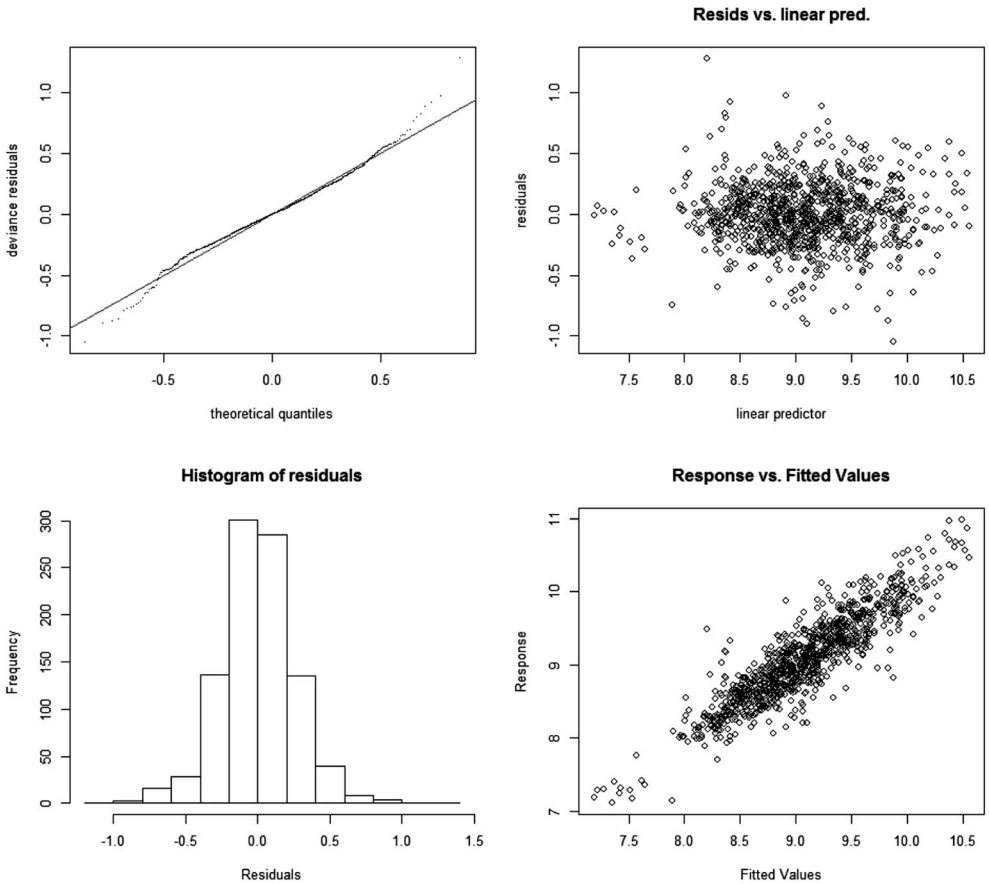

Fig. 3 Diagnostic plots from GAM (model 4).

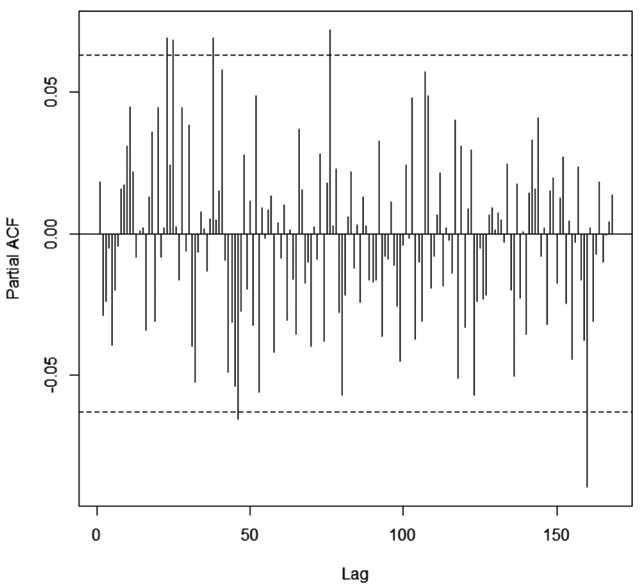

Fig. 4 Partial Autocorrelation Function (PACF) of the residuals from GAM (model 4).

the variability of the model, indicating that those variables are not the only important covariates in modelling UFP-NC. Incorporating temporal trends and other meteorological factors significantly improved R^2.

The inverse proportional relationship between the wind speed and UFPs was consistent with the literature.[1,12,20] As a matter of fact, high wind speeds increase air mixing and more particle losses due to deposition and scavenging.[6,21] Temperature showed an inverse relationship with UFP concentrations. This might be due to the evaporative loss of semi-volatile nanoparticles at higher temperatures.[22] The relationship between UFPs and solar radiation showed a U shape. The first part, with decrease in UFPs and increases in solar radiation might be associated with an increase in the height of the boundary layer that occurs at higher temperatures and irradiation conditions, and thus there is higher mixing of aerosol particles within a bigger volume. The second part of U-shape might be associated with an increase of new UFPs formed from the nucleation of photo-chemically generated compounds.

Relative humidity (RH) shows diurnal inverse correlation with temperature, and hence the effect of RH on UFPs should be one of a positive association. Olivares *et al.* measured high concentrations of UFPs during high humidity periods, and associated this with an increase of nucleation mode particles with increased relative humidity.[23] This was not observed in our

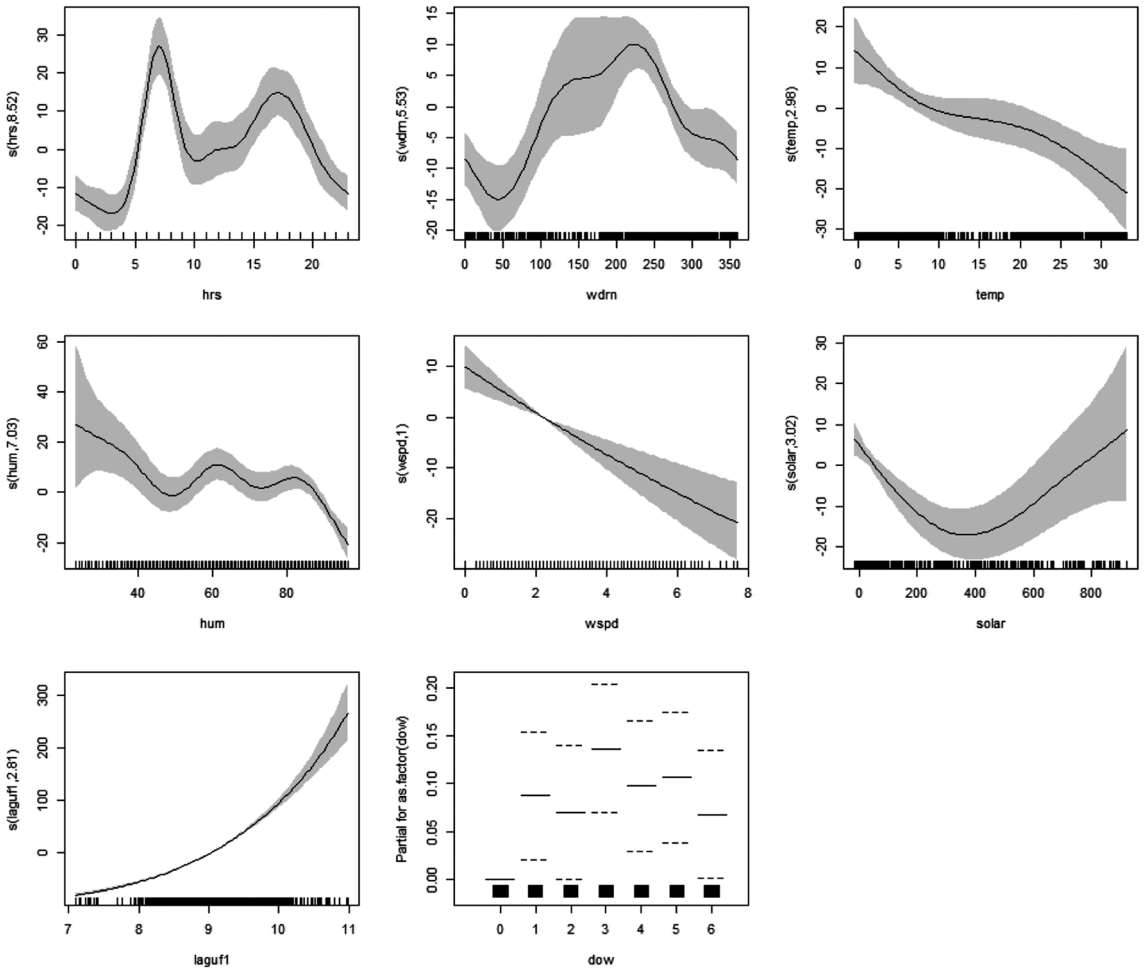

Fig. 5 Relationships between the percentage change in UFP levels (*Y* axis) and all covariates (*X* axis) at the rural station of Calamosco (model 4). For variables in the *X* axis refer to Table 1 for their covariate meaning. For each variable the smoothed function from GAM reported the degrees of freedom (*e.g.* s (h, 8.52) means spline for variable h having 8.52 degrees of freedom) and the 95% CI (grey shade). The range of the factor variable dow (day of the week) is 0–6 (0 = Sunday, 1 = Monday,…, 6 = Saturday), and Sunday is used as baseline and the dotted lines represent the 95% CI. Variable h defines the hour during the day (h) in local time between h and h+1.

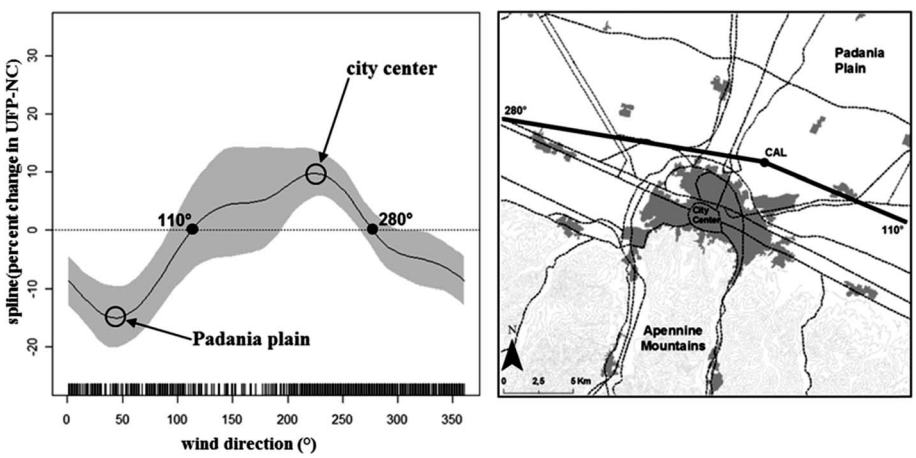

Fig. 6 Relationship between ultrafine particle number concentration (UFP-NC) and wind directions (model 4) at the rural station (CAL). *Y* axis represents the percent change in UFP-NC. The smoothed function from GAM output is reported on the left, while on the right the associated map shows the correspondence with the wind directions identified by GAMs.

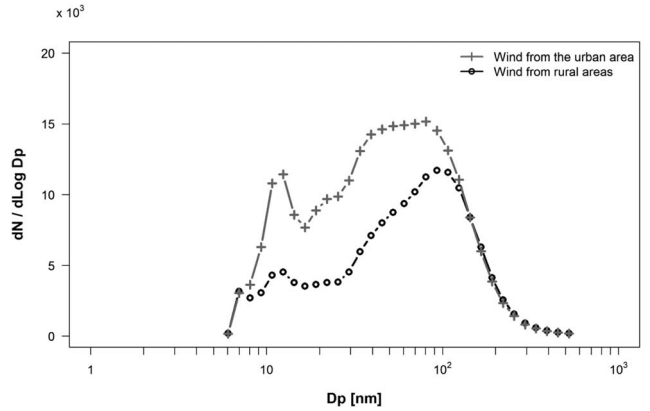

Fig. 7 Size distributions of particles coming from the urban area (220 ± 40° N) and from the rural areas in the opposite direction (40 ± 40° N).

results. However, Clifford *et al.* have seen that higher humidity results in a decrease of particles, similar to our findings.

We did not have long time series of data available but this did not seem to be a strong limitation. In fact, no internal restrictions are to be accounted for using GAM with a short time series. In contrast, GAM models add values to the short time series of the data. Inferring general conclusions from data with a small number of observations could be problematic. However, the distribution of the values of our exposure variables during our monitoring period was representative of the annual variability of all variables inserted in the model. The distribution of wind intensities and directions, our main variable of interest, shows remarkable analogies with the typical long-term distributions. The meteorological representativeness of our sampling period in addition to the good fit of our model to the data warrants confidence in our findings.

A limitation of our study was the availability of only one measuring site. Of course, downwind and upwind monitoring would have added value to our analysis. Further investigations using different locations could clarify whether the regional background concentrations would be isotropic in the absence of emissions from Bologna. Despite that limitation in our study design, the GAM showed a good fit to the data and highlighted the effect of wind direction and provided results consistent with previous studies for other covariates, using only one site and a short time series. Monitoring two, three or four sites at the same time might lead to more consistent findings.

This study also provides new information in terms of exposure to UFPs. To our knowledge no studies have been published on the impact of an urban area on UFP concentrations in its surroundings. We found only studies comparing UFP levels in urban and rural sites. These studies did not analyse the direct impact of city in a lee approach. Moreover, most of these studies considered rural sites very far from the urban areas, *i.e.* tens to hundreds of km away from the source of emission.

It is important to highlight that a number of studies which have investigated UFP concentrations close to high traffic roads have found a sharp decrease of particle concentrations with distance.[24] By contrast, we found a significant impact of the city of Bologna at a distance of several kilometres in terms of the increase of ultrafine particles when the wind blows from the urban area.

The findings of this study contribute to evidence on the effect of exposure to ultrafine particles of a population leaving in near proximity (some kilometres away) to an urban area. People residing there are significantly affected by the urban pollution plume. Masses of air downwind of urban areas present an increase in the concentrations and a decrease in the particle size of ultrafine particles in the atmosphere of local rural communities nearby urban conurbations, which might affect the health of the rural population.

Acknowledgements

The authors would like to thank Prof. Richard Morris (Department of Primary Care and Population Health, UCL Medical School), Claudio Maccone (ARPA Emilia-Romagna), Giovanni Bonafè (ARPA Emilia-Romagna), and Mauro Rossi (ARPA Emilia-Romagna). The UFP data were collected during the MONITER project approved and funded by the Region of Emilia-Romagna (DGR n. 466, 11/4/2007).

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