Ann Ist Super Sanità 2010 | Vol. 46, No. 3: 242-253
DOI: 10.4415/ANN_10_03_04

Evaluation of the temporal variation of air quality in Rome, Italy from 1999 to 2008

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Summary. The main objective of this study was to asses the temporal variation (1999 trough 2008) of air quality in Rome, focusing on airborn concentration of selected pollutants (PM_{10} and $PM_{2.5}$ mass concentration and particle number concentration, PNC, carbon monoxide, CO, nitrogen oxides, NO and NO_2) used for health effects assessment in epidemiological analyses. Time series analysis using Seasonal Kendall test has been applied. A statistically significant decreasing trend was found for primary gaseous pollutants and total particle number concentrations. Moreover a decreasing trend was assessed for PM_{10}, $PM_{2.5}$ and NO_2 measured at traffic oriented sites even if the estimated reduction was lower compared with NO, CO and PNC. The urban background PM_{10} and NO_2 concentrations seem to be practically unchanged since 1999 as no statistically significant trends were found. All the pollutants show higher slope of the estimated trend line at traffic oriented sites compared with those observed at the urban background. Thus a reduction of the intra-city concentration variability throughout the years occurred.

Key words: trends, particulate matter, air pollution.

Riassunto *(Valutazione della variazione temporale della qualità dell'aria a Roma, Italia dal 1999 al 2008)*. Il principale obiettivo di questo lavoro è stato quello di valutare la variazione nel tempo (dal 1999 al 2008) della qualità dell'aria a Roma, con particolare riferimento alle concentrazioni di alcuni inquinanti aerodispersi (concentrazione di massa del PM_{10} e del $PM_{2.5}$, concentrazione numerica delle particelle, PNC, monossido di carbonio, CO, ossidi di azoto, NO e NO_2) usate negli studi epidemiologici per valutare gli effetti sulla salute correlati all'esposizione. La stima dei trend è stata eseguita usando il test non parametrico di Kendall corretto per la stagionalità. È stato individuato un trend statisticamente significativo di riduzione delle concentrazioni degli inquinanti gassosi primari (NO, CO) e del numero di particelle (PNC). Un trend statisticamente significativo di riduzione è stato individuato anche per PM_{10}, $PM_{2.5}$ e NO_2 limitatamente ai siti di monitoraggio orientati al traffico, sebbene meno rilevante rispetto a quanto osservato per NO, CO e PNC. I livelli di concentrazione di PM_{10} e NO_2 nel sito di fondo urbano sembrano invece rimanere praticamente invariati dal 1999 e non è possibile individuare alcun trend statisticamente significativo. Poiché per tutti gli inquinanti è stata evidenziata una pendenza maggiore della linea di tendenza stimata nelle stazioni di traffico rispetto a quella di fondo, è possibile concludere che nel corso degli anni si è verificata una riduzione della variabilità spaziale dell'esposizione all'interno della città.

Parole chiave: trend, materiale particolato, inquinamento atmosferico.

INTRODUCTION

Rome is the country's largest and most populated municipality with 2 724 347 residents (half of the population of the Lazio region [1]) in 1285.3 km^2. It is characterised by alternate zones of very high urbanisation and population density and zones where urbanization is lower or absent.

Services and commerce are the main working activities, and emission of air pollutants from industries is relatively low (there aren't heavy industries in the area around Rome and the nearest big power plant is at about 100 km from the urban area), compared with those originating from road traffic sources. In addition to the people living in the cities themselves, there are also people living in the 121 little towns in the surroundings who commute on a daily basis to the main city.

Traffic flow is rather high during work days with rush hours occurring twice everyday (morning and late afternoon). Traffic flow reaches 20 000 vehicles per hour in the busiest streets [2].

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During the period studied (1999 through 2008) important changes have been observed in the traffic flow composition that could have had an important impact on air pollution [3]. The number of vehicles has increased little compared with the previous decades, reaching 1 923 397 cars registered in 2008 (about 0.7 cars per inhabitant, likely the highest ratio in European capitals).

The number of cars which meet the most stringent emission standards (Euro III and Euro IV) has increased remarkably: at the beginning of the study there weren't Euro III and Euro IV cars yet.

In 2005 the Euro III cars were 617 235 (33% of the fleet) and Euro IV 200 478 (11%); in 2008 the Euro IV cars began the majority (760 216, 40% of the fleet) while Euro III were 399 483 (21%). As a whole Euro III and Euro IV cars represent the 61% of the car fleet in 2008. The pre-Euro cars decreased by 74% in 2008 compared with the year 2000 figures. The positive impact on air pollution that this pattern suggests, could be partially limited by the contemporaneous sharp increase in the number of diesel cars and light duty vehicles, LDV, (mainly diesel powered): 711 400 diesel cars in 2008, reflecting an increase of 300% compared to 2000; 151 380 LDV, an increase of 62% compared to 2000.

Overall, remarkable increase in motorcycle sales figures occurred. This phenomenon can be explained by the need for fast mobility throughout the cities, which is hard to achieve either by passenger car, mainly due to traffic congestion, or by public transport which is rarely competitive, with the exception of the underground.

As in several other large urban areas in Italy and Europe, air pollution in Rome still remains of high concern due to limit values exceedances as well as the failure to reach the target and objectives stated by the European directives and National laws [4, 5]. In 2009 the annual mean and 24-hours limit value for particulate matter less than 10 μm in aerodynamic diameter (PM_{10}), the nitrogen dioxide (NO_2) annual mean limit value and the ozone (O_3) target value, long term objective and information threshold have been exceeded. This occurred in spite of policies being pursued to reduce emission, both at national and local level, and regardless of the estimated reduction of primary pollutant emission showed by national and local emission inventories.

Detection of a statistically significant trend in atmospheric concentrations of air pollutants is of great interest as it could support policies toward the attainment of the targets, as well for epidemiological studies focused on long term exposure to air pollutants.

The main objective of this study was to assess the temporal variation (1999 through 2008) of air quality in Rome, focusing on airborne concentration of selected pollutants: PM_{10} and $PM_{2.5}$ (particulate matter less than 2.5 μm in aerodynamic diameter) mass concentration, ultrafine particle number concentration, PNC, carbon monoxide, CO, nitrogen oxides, NO and NO_2) which have been analyzed for determining whether there is a monotonic (single-direction) trend over time not limited to a visual estimation but following a formal, quantitative procedure.

MATERIALS AND METHODS

Data used for the purpose of this study came from two main sources. The first one is the measurement station located in the front yard of the Istituto Superiore di Sanità (ISS) (2 km east of the city center) which has been operating since 1978, measuring classic pollutants such as NO, NO_2, CO, O_3, sulphur dioxide (SO_2), lead (Pb) and total suspended particles (TSP) [6].

Since the early nineties it has become the monitoring of PM_{10}, $PM_{2.5}$ as well as the coarse fractions of PM (PM_{10}-2.5). From 2001, PNC has been continuously measured, starting from the framework of the HEAPPS study [7, 8], until today.

Reference methods for the sampling and measurements of air pollutants as outlined in the air quality Directives [9, 10] have been strictly followed. Two low-volume samplers, model SKYPOST PM (TCR Tecora, Italy), were used for PM_{10} and $PM_{2.5}$.

One operated at 1 m^3/h, equipped with an omnidirectional aerosol inlet designated as reference for PM_{10} by the US Environmental Protection Agency [11] and a WINS impactor [12] to separate the particles into the $PM_{2.5}$ fraction. The second operated at 2.3 m^3/h, equipped with an omnidirectional aerosol inlet designated as reference for PM_{10} according to CEN standard EN 12341 [13]. PM was collected daily on a 47-mm-diameter glass fiber filter (Pall Corporation, USA) or polymethylpentane-ringed, 2.0 μm pore size, 47-mm-diameter polytetrafluoroethylene (PTFE) filters (Gelman, USA). Mass concentrations of both fractions of PM were determined gravimetrically using a Sartorius model M5P 000V001 electrobalance (readability: 0.001 mg). PNC was measured by a TSI model 3022A condensation particle counter (CPC). This type of counter is able to monitor particles larger than 0.01 μm in diameter but still has a 50% counting efficiency at 0.007 μm [14, 15].

CO measurements were carried out using a CO gas filter correlation analyzer model 300 (Teledyne Advanced Pollution Instrumentation API Inc.), following the EN 14626:2005 standard method, based on nondispersive infrared spectroscopy.

NO_2 measurements were carried out using a chemiluminescent NO_x analyzer model 200A (Teledyne API Inc.), following the EN 14211:2005 standard method based on chemiluminescence. Standard Operating Procedure details can be found in previous papers [7, 8, 16].

To have a wider view of spatial variability throughout the city, the data collected for regulatory purposes from the regional air quality monitoring network have been used. Those data were shared

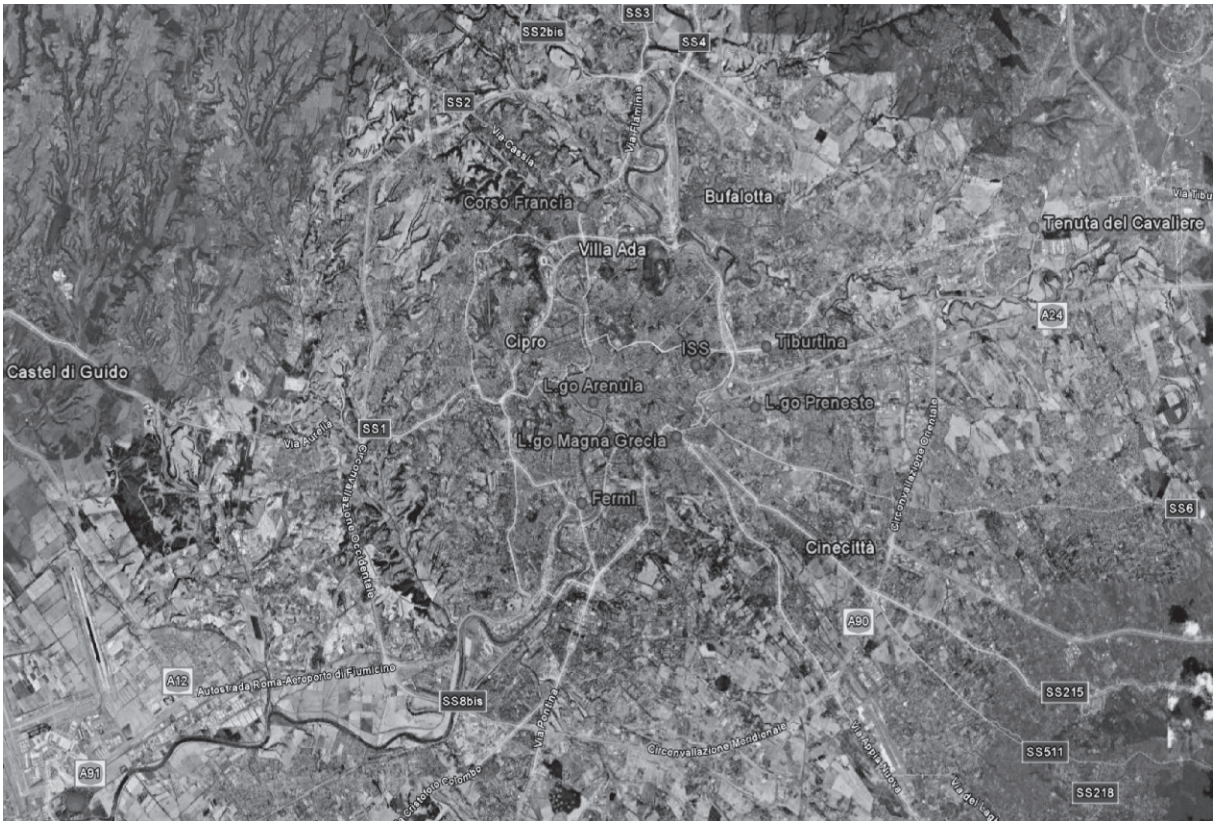


Fig. 1 | *Rome monitoring network (2008 configuration). Only monitoring stations within the communal border are showed. Traffic oriented sites; Corso Francia, Tiburtina, L.go Preneste, L.go Arenula, L.go Magna Grecia, ISS, Fermi; urban background sites: Villa Ada, Cipro, Bufalotta, Cinecittà, Castel di Guido, Tenuta del Cavaliere. All the stations belongs to the regional network except for the ISS station which is a research multipurpose station managed by the Italian National Institute of Health. Only data from five monitoring station were used in this study (ISS, Arenula, Magna Grecia, Fermi, Villa Ada, see "Material and method" section).*

in the framework of the Exchange of Information protocol (EoI) between the member countries of the European community (Decision 97/101/CE), managed for Italy by Institute for Environmental Protection and Research (ISPRA), by means of the BRACE data base. Long term continuous data for the whole set of pollutants (PM_{10}, CO, NO and NO_2) are available from a restricted number of monitoring stations belonging to the current configuration of the network *(Figure 1)*. These are three traffic-oriented stations (Magna Grecia, Fermi, Arenula) and the historical urban background monitoring station of the city, located in one of the largest public parks of Rome, called Villa Ada. Reference measurement methods were used for CO, NO and NO_2 whereas beta attenuation mass monitor, certified as equivalent to the reference method was used for sampling and measurement of both PM_{10} and $PM_{2.5}$. QA/QC procedures were strictly followed, as stated in the legislation in force for the air quality monitoring regional network [17].

Daily averages for CO, NO, NO_2 and PNC have been calculated starting from hourly averages measured at each site, if at least 16 hourly averages were available.

Each station and pollutants had missing daily averages data. To fill the gaps for each pollutant A, first the q stations with yearly data coverage higher than 90% were selected ($1 \le q < n$). Then a daily variability profile p_d was calculated as follows, based on the observation that relationships between observed concentrations of the same pollutants (expressed as Pearson correlation coefficient) at the different selected sites were always higher than 0.7:

$$p_d = \frac{\sum_{k=1}^{q} [A]_{d,k}}{\sum_{k=1}^{q} \beta_{d,k} \cdot [A]_{y,k}}$$

Where:

$\beta_{d,k} = 0$ if $[A]_{d,k}$ = missing

$\beta_{d,k} = 1$ if $[A]_{d,k}$ = not missing

$[A]_{d,k}$ = daily average concentration of the pollutant A measured at the station k

$[A]_{y,k}$ = yearly average concentration of the pollutant A measured at the station k

Then the data series for the q stations and the missing data days j were completed as follows:

$$[A]_{j,k} = p_j \cdot [A]_{y,k}$$

To complete the time series of the n-q stations with yearly data coverage less than 90%, first an overall pollutants daily average was calculated from the *q* stations:

$$[A]_d = \frac{1}{q} \cdot \sum_{k=1}^{q} [A]_{d,k}$$

Thus a correlation line was built for the z (n-q) station with data coverage < 90%, starting from the i daily average available:

$$[A]_{i,z} = a \cdot [A]_{i,d} + b$$

The calculated slope (a) and intercept (b) was finally used to estimate the lost values j at each z station:

$$[A]_{j,z} = \hat{a} \cdot [A]_j + \hat{b}$$

With such an approach it was possible to improve the data coverage to 100% for CO, NO and NO_2 for all the monitoring stations selected; only a few days were still missing for PM_{10} (no measurement available at all, 4 days out of 3652 days of observations).

Long term, though discontinuous, measurements of $PM_{2.5}$ are available only from the ISS station, which has operated since 1999, while at the regional networks monitoring of $PM_{2.5}$ began in 2006. For the ISS site 1538 daily averages are available (evenly distributed over the years). Based on these measurements and on the daily observed $PM_{2.5}/PM_{10}$ ratios we have built a retrospective estimation of $PM_{2.5}$ concentrations at the 5 historical stations used in this study.

PNC has been measured since April 2001. Previous data since 1999 has been retrospectively estimated in the framework of the HEAPPS study [7, 8], using concurrent measurements of air pollutants and weather data, as well as selected interactions between the two, to fit a regularized linear model also called ridge regression [18].

Trend analysis was carried out after grouping the measurements carried out in each traffic station into one averaged traffic stations pattern. Trend analysis for the urban background sites has been carried out only on the Villa Ada site time series, thus grouping was not necessary.

A site specific analysis was carried out only for PM_{10}.

Among the many different statistical approaches available for detecting and estimating trends that may be present in environmental variables of interest, we used the Seasonal Kendall (SK) test [19] an extension of the well-known non-parametric Mann-Kendall's test of association between two variables that is often used as a test of trends [20]. To deal with differences among seasons, the SK system separately tests the trend in each season, and then combines the results into one overall test. This overall test suggests whether there is a trend over time, blocking out all seasonal differences in the pattern of change [21].

The Kendall's τ correlation coefficient (ranging between -1 and +1), measures the strength of the monotonic association between Y and time. The p value summarizes the probability of getting the observed value of τ, or one more extreme, when the null hypothesis is true.

When p is small, the likelihood that there is no trend is also small, and null hypothesis is rejected.

Tests for trends have been of keen interest in environmental sciences over the last 30 years. Among other parametric and non-parametric tests, SK was recently used to assess trends of atmospheric concentrations of semivolatile compounds in the USA [22], particulate matter in four megacities in India [23] and concentrations of primary and secondary pollutants in Finland [24]. The SK test appears to maintain its stated α lev-

Table 1 | *Relationship between the different variables, expressed by the Pearson correlation coefficient*

	CO (UT)	CO (UB)	NO (UT)	NO (UB)	NO_2 (UT)	NO_2 (UB)	PM_{10} (UT)	PM_{10} (UB)	$PM_{2.5}$ (UT)	$PM_{2.5}$ (UB)	PNC (ISS)
CO (UT)	-	0.822	0.854	0.648	0.582	0.470	0.624	0.434	0.662	0.522	0.778
CO (UB)		-	0.855	0.817	0.542	0.646	0.649	0.515	0.727	0.628	0.756
NO (UT)			-	0.860	0.607	0.619	0.676	0.473	0.776	0.634	0.818
NO (UB)				-	0.449	0.637	0.622	0.515	0.726	0.653	0.706
NO_2 (UT)					-	0.705	0.549	0.430	0.537	0.457	0.503
NO_2 (UB)						-	0.497	0.469	0.572	0.545	0.570
PM_{10} (UT)							-	0.857	0.953	0.853	0.574
PM_{10} (UB)								-	0.810	0.927	0.381
$PM_{2.5}$ (UT)									-	0.893	0.691
$PM_{2.5}$ (UB)										-	0.577
PNC (ISS)											-

UT: urban traffic measurement stations (daily averages between the four monitoring station Magna Grecia, Fermi, ISS, Arenula); UB: urban background measurement station (Villa Ada). p- value < 0.001 per each variables pair.

Table 2 | *Pearson's correlation coefficients among pairwise PM_{10} monitoring stations*

	Arenula (UT)	Magna Grecia (UT)	Fermi (UT)	ISS (UT)	Villa Ada (UB)
Arenula (UT)	-	0.856	0.857	0.807	0.837
Magna Grecia (UT)		-	0.850	0.814	0.819
Fermi (UT)			-	0.818	0.816
ISS (UT)				-	0.797
Villa Ada (UB)					-

UT: urban traffic station; UB: urban background station.

els as well as maintain high power with different trend functions. It is also easy to implement [25].

RESULTS AND DISCUSSION

Relationships between observed variables

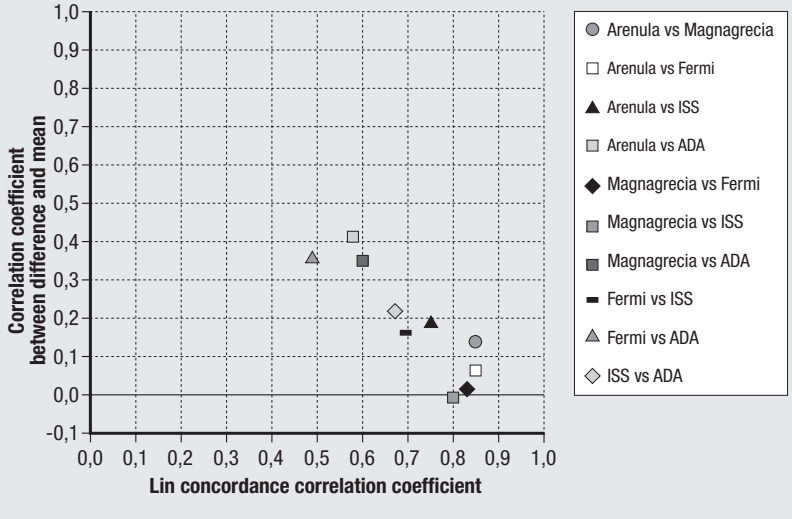
Table 1 shows the correlation matrix (expressed as Pearson's coefficient of pairs of daily averages) between the observed variables measured at the four traffic oriented sites, Magna Grecia, Fermi, Arenula, Istituto Superiore di Sanità (ISS), and at the urban background station Villa Ada. Each variable observed at traffic oriented sites is the average of the daily averages measured at individual sites.

All the observed variables, whether measured at traffic oriented sites or the urban background site, show statistically significant correlations ($p < 0.001$). Such behaviour reflects two main drivers in the variability of air pollutant concentrations: common sources and meteorological condition variability [26, 27]. Differences in strength of the observed association reflects the important differences in pollutants' reactivity after emission, pollutants formation in the atmosphere from precursor and existence of specific sources for some pollutants [28].

The combustion related, primary pollutants CO and NO, mainly arising directly from anthropogenic sources, were highly correlated with each other (0.854 at traffic oriented sites; 0.817 at urban background site) and with the measured PNC (0.778 and 0.818 respectively) confirming the finding coming from other studies in European urban environments and Rome as well [8, 29-31].

Daily $PM_{2.5}$ and PM_{10} levels were found instead to be poorly correlated with the daily PNC and the others gaseous compounds. The mass concentration pattern of PM and NO_2 is due to their characteristics of being largely secondary pollutants meaning that they are formed in the atmosphere from precursor pollutants, directly emitted from the main sources.

Assessing spatial variability of the data is another important task before attempting to analyse trends. In *Table 2* Pearson's correlation coefficients among pairwise PM_{10} monitoring stations are reported. The Pearson's coefficient of correlation was always higher than 0.8; this means that the station's daily pattern of variability is superimposable on each other. *Figure 2* shows the correlations between PM_{10}station pairs during the study period, used to assess homogeneity of the data following the method proposed within the frame-



Note: The concordance correlation coefficient [33] evaluates the degree to which pairs of observations fall on the 45° line through the origin.
Arenula, Magna Grecia, Fermi and ISS: urban traffic stations; Villa Ada: urban background station.

Fig. 2 | *Pair correlations between monitors. PM_{10} measurement, ($\mu g\ m^{-3}$, daily mean), during the study period (1999-2008).*

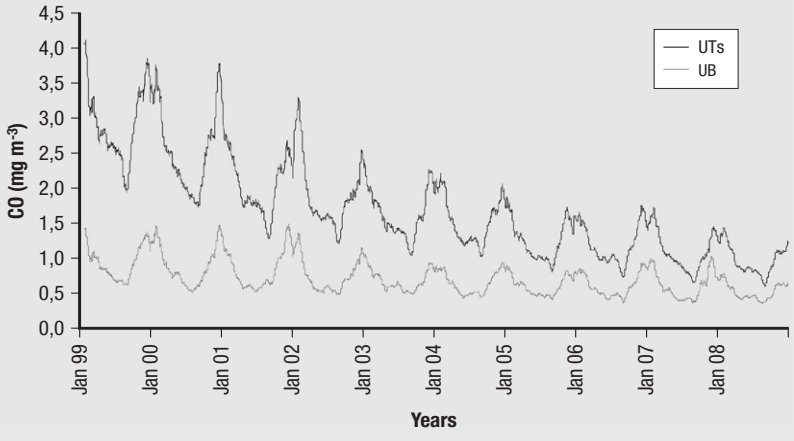


Fig. 3 | *Thirty-day moving average daily mean of carbon monoxide concentrations (mg m^{-3}) measured at the urban traffic stations (Fermi, Arenula, Magna Grecia, ISS - grouped citywide average UTs) and at the urban background station (Villa Ada, UB).*

work of the MISA2 study [32]. Particularly high homogeneity, stated by high Pearson correlation coefficient, high Lin concordance coefficient (which evaluates the degree to which pairs of observations fall on the 45° line through the origin [33]) and low correlation between daily difference and mean of pair measures, was found between traffic stations. These findings confirm those reported elsewhere [34, 35] and allow grouping the measurements carried out in each traffic oriented site into one averaged traffic oriented sites pattern.

A lower Lin concordance coefficient was found when comparing the urban background site with the traffic oriented sites, as expected, due to the relevant impact on measured concentrations (and variability) of the nearby sources.

Trend pattern

Figures 3-6 show the trend pattern for CO, NO, PNC, NO_2, PM_{10} and $PM_{2.5}$ along the ten-year study expressed as the monthly running average. These

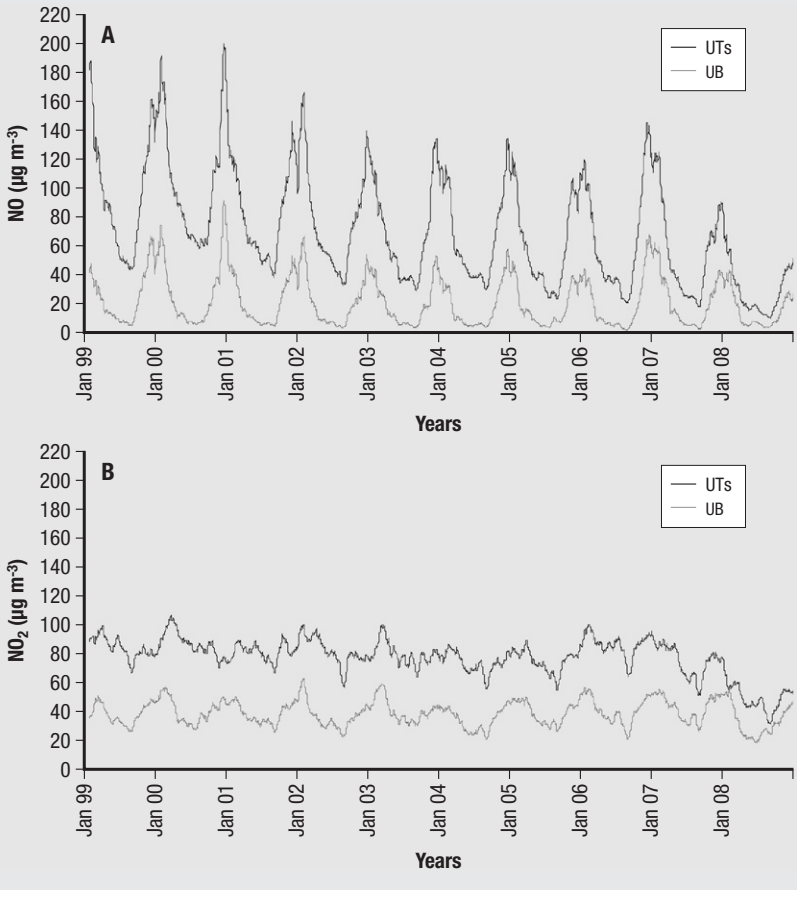


Fig. 4 | *Thirty-day moving average. daily mean of nitrogen oxides concentrations (µg m^{-3}) measured at the urban traffic stations (Fermi, Arenula, Magna Grecia, ISS - grouped citywide average, UTs) and at the urban background station (Villa Ada, UB). (A): nitrogen oxide (NO); (B): nitrogen dioxide (NO_2).*

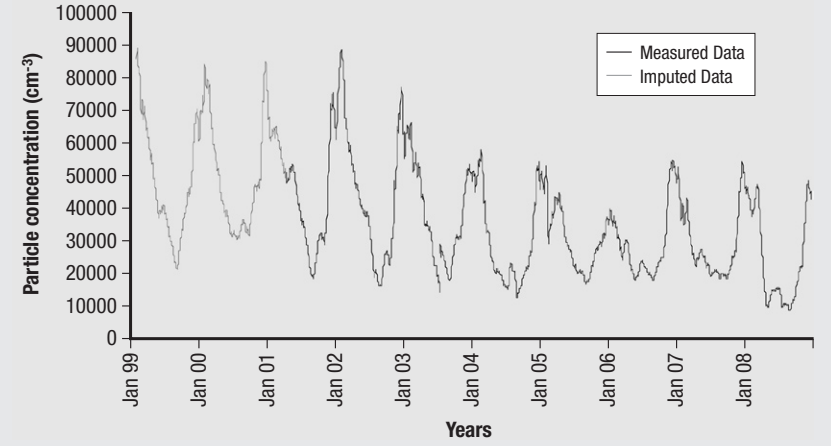


Fig. 5 | *Thirty-day moving average daily mean of particle number concentrations (counts cm^{-3}) measured at the urban traffic station ISS. Data were imputed From January 1999 to March 2001.*

kinds of graphic representations allow to visually check for trend while retaining the majority of the yearly variability due to seasonality. Data from traffic oriented sites (grouped) as well as from the urban background site (dotted line) are reported.

A trend toward reduction of concentrations was visually detected for CO, NO, PNC; the record shows quite a regular annual cycle, maximizing in winter and minimizing in summer. It can be argued that the intra-year variability observed could be due to the weaker atmospheric convective processes in winter: advection periods alternate to multi-day atmospheric stability conditions and diurnal mixing is weak and short leading to atmospheric stagnation, which allows pollutants to accumulate thus generating severe pollution episodes [36, 37]. During warm months convective mixing of the lower atmosphere occurs early in the morning and the lower atmos-

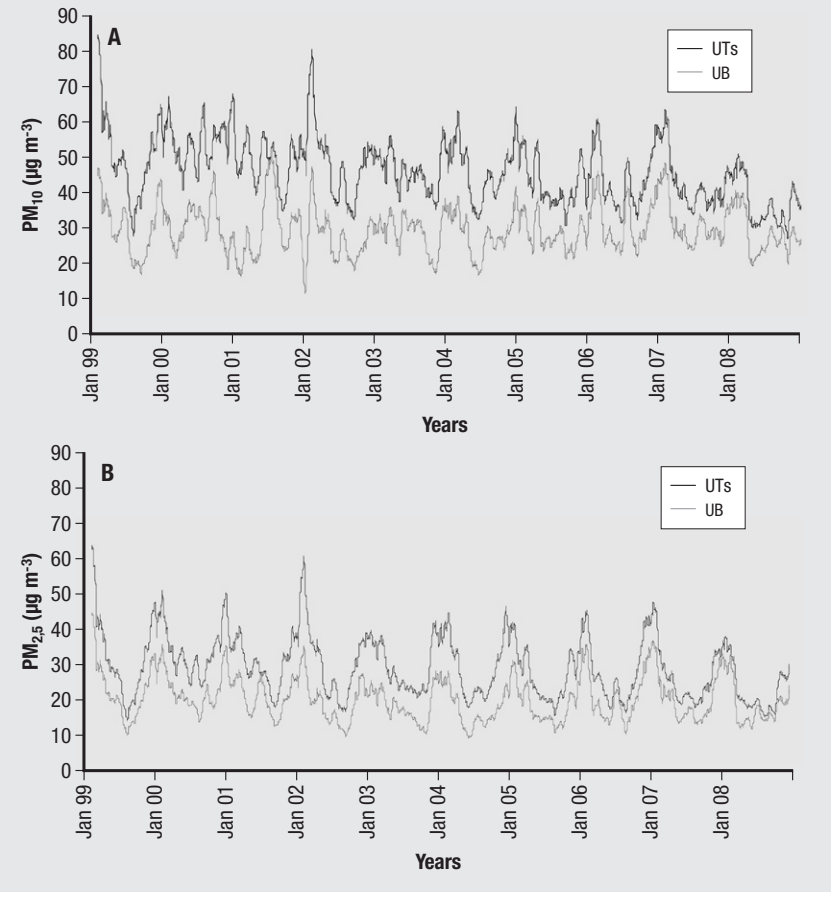


Fig. 6 | *Thirty-day moving average daily mean of particulate matter mass concentrations (µg m^{-3}) measured at the urban traffic stations (Fermi, Arenula, Magna Grecia, ISS - grouped citywide average, UTs) and at the urban background station (Villa Ada, UB). (A): PM_{10}; (B): $PM_{2.5}$.*

phere is already well mixed when the traffic flow increases; stability occurs mainly during night time when the traffic flow has already decreased [27]. This lead to winter traffic related air pollutant levels generally higher than in summer, even if during warm months photochemical smog episodes and Sahara dust advections days often occurs [38]. Moreover we can observe that the difference between levels measured in nearby streets (traffic-oriented sites) and at the urban background site tends to decrease along with the trend observed throughout the years. It is worth noting that the spatial variability throughout the city became smaller year by year. This finding could have an important impact on the estimated spatial variability of population exposure.

The NO trend appears quite similar to that of CO. It should also be noted that the behaviour seems to be more likely a two or three-step trend (first three years, second five years and last two years). We could cautiously suppose that these differences could be due to different impact of the change in vehicular fleet composition observed on the CO and NO emission rate.

PNC has been measured only at one site during the study period, thus we cannot extend the observed decreasing trend to the whole city. The HEAPPS study [7, 8], comparison carried out between two sites (ISS and an urban background site) shows that the absolute values found in the urban background site were lower (by about 50%) than the values found in the traffic-related site while maintaining a good correlation, suggesting dispersion of these particles also at relatively long distances from the primary sources. The burden of evidence allows to affirm that ultrafine particles act very much like primary gaseous compounds (CO and NO) though the spatial variability is mainly due to atmospheric dispersion as well as coagulation that contributes to the rapid decrease in particle number concentration with distance from the main sources (road traffic) [8]. Keeping in mind that several studies have shown that PNC is dominated by the ultrafine fraction of aerosol particles (*e.g.* those with aerodynamic diameter less than 0.1 μm) we are confident that there is a general tendency for a decrease in the number of ultrafine particles through the city coherent with the observed decrease of concentrations of primary combustion-related gaseous compounds.

It is not possible to extrapolate a clear trend visually merely from PM_{10} and NO_2 graphic patterns. For the traffic-oriented sites pattern toward reduction is smoothed (if any) compared with the primary gaseous compounds (NO, CO) and PNC. At the urban background site both PM_{10} and NO_2 trends appear negligible. For PM_{10} we should add the observation that there are several relative maxima during the warm season, unexpected if we consider only the particles of primary and secondary origin, likely due to Saharan dust advection. A clear effect of Saharan dust advection, mostly between May and November were lidar-assessed during 2001-2004. On average an added contribution of 12.1 ± 1.1 (8.3 ± 6.5) μg/m^3 with respect to a dust free 15-day running average PM_{10} ($PM_{2.5}$) record was estimated [39].

Seasonal Kendall test for trend results

Table 3 displays the results obtained by applying the seasonal Kendall test for trend (SK) for CO, NO, NO_2, PM_{10}, $PM_{2.5}$ and PNC time series.

The presence of a statistically significant trend is evaluated using the test statistic Z (normally distributed). The Kendall's τ correlation coefficient, the p value at 0.05 α level, the slope and the intercept of the estimated trend line are reported for each pollutant and for grouped traffic oriented sites and the Villa Ada urban background site. The estimated percent reduction of the yearly mean concentration in the whole period is also reported.

CO levels have been significantly decreasing since 1999 (Z= –13.306 at traffic oriented sites and –10.796

Table 3 | *Results of the seasonal Kendall (SK) test for trends for CO, NO, NO_2, PM_{10}, $PM_{2.5}$, PNC*

Pollutant	Monitoring site	Z	τ	p-value	Slope	Constant	Change overall (%)
CO	UTs	-13.306	-0.956	0.000	-0.190	2.615	-65%
	UB	-10.796	-0.776	0.000	-0.039	0.874	-40%
NO	UTs	-10.847	-0.778	0.000	-6.000	97.00	-56%
	UB	-6.425	-0.454	0.000	-0.833	20.58	-36%
NO_2	UTs	-5.878	-0.420	0.000	-2.000	91.00	-20%
	UB	-0.446	-0.033	0.655	-	-	negligible
PM_{10}	UTs	-4.592	-0.330	0.000	-1.000	49.50	-18%
	UB	-0.235	-0.019	0.815	-	-	negligible
$PM_{2.5}$	UTs	-6.479	-0.459	0.000	-1.000	33.00	-27%
	UB	-3.163	-0.224	0.002	-0.400	21.20	-17%
PNC	ISS	-8.908	-0.641	0.000	-2.754	49.790	-50%

at urban background site); the strength of the monotonic association is pointed out by the high values of the τ correlation coefficient (- 0.956 at traffic oriented sites and - 0.776 at urban background site, $p < 0.01$). Overall concentrations decreases by 65% at the traffic oriented sites, and by 40% at urban background site were estimated, *i.e.* 0.2 mg m^{-3} y^{-1} and 0.04 mg m^{-3} y^{-1} respectively.

Results of the quantitative trend assessment for NO confirm the "visual" inspection (Z = - 10.847 at traffic oriented sites and - 6.425 at urban background site) though if the rate of the decrease is lower (- 6 μg m^{-3} y^{-1} at traffic oriented sites, - 0.8 μg m^{-3} y^{-1} at urban background site, overall decreasing being - 56% and - 34% respectively) as well as the τ correlation coefficient (- 0.778 and - 0.454 respectively, $p < 0.01$) compared with those of CO.

As CO and NO, PNC measured at ISS shows a monotonic change over time (Z= - 8.908) stated by highly significant τ (- 0.641) and estimate decrease of 2754 count cm^{-3} y^{-1}, leading to a 50% overall decreasing.

Regarding NO_2 and PM_{10} at the traffic oriented sites the decreasing trend is still statistically significant: (Z= - 5.878 and Z= - 4.592 respectively) though the rate of decrease is much lower than in the previous cases ($\tau = - 0.420$ and $\tau = - 0.330$ at $p < 0.01$ respectively; estimated yearly decrease was 2.0 μg m^{-3} y^{-1} for NO_2 (overall decrease by 20%) and 1.5 μg m^{-3} y^{-1} for PM_{10}; overall decrease by 18%). More, in the urban background site, statistically significant trend for NO_2 and PM_{10} were not observed.

A similar pattern was found in a recent study in Finland [24] suggesting that during their study period (1994 trough 2007) the proportion of NO_2 in NO_x (*i.e.* the sum of NO and NO_2) in ambient air has increased as a results of the increased NO_2/NO_x ratio in vehicular exhaust, due to the emission of diesel vehicles and their increase in vehicular fleet. The Finland study confirms other recent studies findings [40-43]. It should also be taken into right account as another factor contributing to the change in the atmospheric NO_2/NO_x ratio, the effect of NO decreasing in the atmospheric equilibrium between NO, NO_2 and O_3: a decline in NO concentrations does not necessarily lead to a similar decline in NO_2 concentrations [44].

Attempting to explain the PM_{10} behaviour one should keep in mind that PM_{10} is a complex mixture of both particles directly emitted from primary sources and formed in the atmosphere from precursor (secondary inorganic compounds such as ammonium nitrate and organic compounds, mainly of secondary origin too) and that is enhanced in winter. Particles of secondary origins belong mainly to the fine particles (aerodynamic diameter ranging between 0.1 and 2.5 μm) and have a long lasting residence time in the atmosphere compared with those belonging to the ultrafine (aerodynamic diameter less than 0.1 μm) and the coarse particles (aerodynamic diameter ranging between 2.5 and 10 μm). It

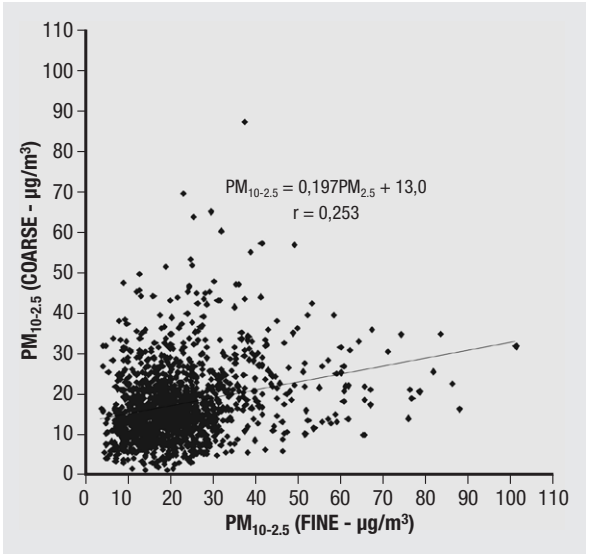


Fig. 7 | *Correlation between daily average particle mass concentrations fine ($PM_{2.5}$) and coarse ($PM_{10-2.5}$) fractions measured in Rome (1999-2008) at the ISS monitoring station. Number of pairs: 1508.*

is not easy to relate the behaviour of PM_{10} to the reduction of primary pollutants that acts as precursor of particles in the accumulation mode as well as to the reduction of particles of primary anthropogenic origin, that was effectively observed both in the emission inventory estimate [5] and from the trend analysis of the primary gaseous pollutants and PNC (dominated by ultrafine particles) already showed. Unfortunately long-lasting time series of sulphate, nitrate and ammonium ions, which could help explain the secondary inorganic particles role in the observed trend, are not available.

Enforcing the finding related to PM_{10} is interesting to observe the very weak linear relationship between the fine and the coarse fraction of PM *(Figure 7)* compared with the very good linear relationships between $PM_{2.5}$ and PM_{10} *(Figure 8)*. Only 20% of the coarse variability can be explained by the variability of $PM_{2.5}$ and as an average more than 10 μg/m^3 of PM_{10} in the coarse fraction could be originating from source different from those of $PM_{2.5}$. It is worth noting that a characteristic $PM_{2.5}/PM_{10}$ ratio of 0.6 could be estimated from the slope of the linear regression confirming previous data (*e.g.* [45]). The coarse fraction contribution to the PM_{10}overall behaviour could be partially affected by the frequency and intensity of the Saharan dust events that is obviously uneven year by year.

Regarding $PM_{2.5}$ the resulted trend should be considered with due caution as it is based mainly on calculated instead of measured concentrations. Anyhow we have found in this case a statistically significant trend at both traffic and background sites though if in the latter the strength of the monotonic association between $PM_{2.5}$ and time is very weak.

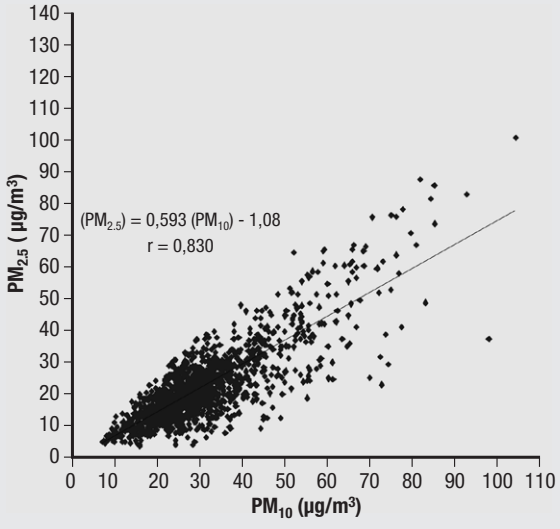


Fig. 8 | *Correlation between daily average particle mass concentrations $PM_{2.5}$ and PM_{10} fractions measured in Rome (1999-2008) at the ISS monitoring station. Number of pairs: 1508.*

Table 4 reports the trends analysis for PM_{10} measured at traffic oriented sites. We can see that the observed decreasing trend is coherent between sites, which show all a similar decreasing pattern. Thus the overall pattern previously observed is not affected, perhaps, by one station decreasing trend driving the overall trend. It should be noted that the higher yearly decrease was found at the Largo Arenula station, located inside the limited-traffic zone of the city centre (about 6 km^2 surface area; only resident, authorised commercial and public transport vehicles have access and free parking). During 2008 the monitoring station was moved from the original location to a new location at about 200 m from the latter, in a zone where the influence of combustion-related primary pollutants could be reduced compared with the previous location, nearby high traffic street. Also the Piazza Fermi station was moved from the original location to the opposite kerbside since June 2006.

Though if the SK accounts for seasonality in the data, it does not allow us to sort out if there is a single pattern of trends across all seasons or the overall trend arise from a strong trend in one or more season whereas not or opposite trend is detectable in the others.

The multi-season analyses for trend results are shown in *Table 5* for PM_{10} time series at traffic oriented sites and at urban background site. In this case the original Mann-Kendall test was applied to the yearly, season specific, daily averages of measured PM_{10} concentrations.

No statistically significant trend was observed during winter and summer whereas a significant trend was found during spring at both traffic and background sites ($Z = -2.988$, $p < 0.01$ and $Z = -1.968$, $p < 0.05$ respectively). A significant decreasing trend was found also during fall only at traffic-oriented sites ($Z = -1.984$, $p < 0.05$). The reason for such a "season" dependent trend could be due to the meteorological conditions that favour secondary components formation of PM during winter, as well as to the higher frequency of Saharan dust advection during summer. In other words there is a meteorological "driving force" that hides the reducing trend due to primary pollutant emission fall down.

Table 5 | *Results of the Kendall test for trends for PM_{10} season specific*

	Z	τ	*p*-value	Slope
Urban traffic				
Spring	-2.988	-0.756	0.003	-1.800
Summer	-0.996	-0.267	0.319	-
Fall	-1.984	-0.511	0.047	-1.250
Winter	0.000	-0.022	1.000	-
Urban background				
Spring	-1.968	-0.511	0.049	-1.080
Summer	-0.527	-0.167	0.598	-
Fall	0.000	0.022	1.000	-
Winter	1.082	0.289	0.279	-

CONCLUSION

Trend analysis, based on air pollution data measured in Rome since 1999 to 2008, shows a statistically significant decreasing trend for primary gaseous pollutants and total particle number concentrations.

Table 4 | *Results of the seasonal Kendall (SK) test for trends for PM_{10}*

Monitoring site	Z	τ	p-value	Slope	Constant
Arenula (UT)	-6.897	-0.527	0.000	-1.942	56.4
Fermi (UT)	-4.426	-0.335	0.000	-1.300	55.7
Magna Grecia(UT)	-4.719	-0.371	0.000	-1.100	50.2
ISS (UT)	-3.737	-0.304	0.000	-1.333	47.3
Villa Ada (UB)	-0.235	-0.019	0.815	-	-

Moreover a decreasing trend was assessed for PM_{10}, $PM_{2.5}$ and NO_2 measured at traffic oriented sites even if the estimated reduction was lower compared with NO, CO and PNC.

The urban background PM_{10} and NO_2 concentrations resulted practically unchanged since 1999 as no statistically significant trends were found.

The decreasing trend observed at the traffic oriented sites seems to be related mainly to the progressive shifts towards vehicles meeting the most stringent European emissions standard which was enhanced by national and local measures during the period study.

The lack of a decreasing trend in the urban background site could be explained by the role of secondary atmospheric process that lead to the formation of both particle and NO_2 irrespective of the reduction of the emission of gaseous primary pollutants. It was observed that the relative contribution of primary particles to the PM_{10} concentrations is different between traffic-oriented sites and urban background sites (about 30% and about 15% respectively [46]). Thus if the anthropogenic emission reduction mainly affects the primary particles concentrations, the overall effect to the PM_{10} concentrations could became negligible at the urban background sites.

The CO and NO downward trend (together with the well known reduction of "fuel related" pollutants SO_2, benzene and Pb) suggests an overall improvement of urban air quality in Rome.

Notwithstanding, no meaningful change has been observed in PM_{10} and NO_2 concentration measured at urban background sites since 1999. Thus, the long term population exposure to PM_{10} and NO_2 (likely well described by the urban background sites levels) seems to be unchanged in the study period.

The PM_{10} levels, over the whole set of available data, lie between the WHO interim target 2 (50 μg m^{-3} as annual mean) and target 3 (30 μg m^{-3} as annual mean).

To reach the most stringent WHO air quality guidelines (20 μg m^{-3} as annual mean) a 25% reduction of the yearly average measured should be achieved to comply with the target at least at the urban background site.

Finally it should be noted that all the pollutant show higher slope of the estimated trend line at traffic oriented sites compared with those observed at the urban background, thus an intra-city reduction of the exposure spatial variability throughout the years occurred. These finding should be taken into account while assessing long term exposures differences between people living near high traffic roads and people living in residential suburbs relatively far from main sources.

Conflict of interest statement

There are no potential conflicts of interest or any financial or personal relationships with other people or organizations that could inappropriately bias conduct and findings of this study.

Submitted on invitation.
Accepted on 22 April 2010.

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