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THE IMPACT OF WIND ON PARTICLE MASS CONCENTRATIONS IN FOUR EUROPEAN URBAN AREAS

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ABSTRACT

The impacts of wind conditions to fine and coarse particle mass concentrations at four European urban centers, by multivariate regression analysis of particle measurements against categorized wind (direction and speed) conditions were estimated. Statistically significant associations (both positive and negative) were observed for all urban areas. Both fine and coarse particle mass concentrations were decreased for moderate and strong winds (speed > 2.2 m s^{-1}) in Athens and Helsinki. Weak winds had a positive influence on particle mass, accounting for up to 40% of fine particles in Athens. For the three coastal urban areas (Amsterdam, Athens and Helsinki), positive correlations were observed for sectors encompassing ports and areas with intense marine traffic. For Birmingham, the association of both particle fractions with the eastern sector indicated the influence of emissions from central UK and continental Europe. The method described here may be used to screen the orientation of sources near receptor sites.

KEYWORDS: Fine aerosol, coarse aerosol, harbors, regression analysis, shipping emissions.

INTRODUCTION

Several studies have addressed the complicated impact of weather patterns on the levels and, spatial and temporal variation of atmospheric particulate matter (Jones *et al*., 2010). Chaloulakou *et al*. (2003), estimated that low wind conditions may contribute up to 1.1 of PM_{10} and up to 1.5 μg m^{-3} of $PM_{2.5}$ mass concentrations in Athens, Greece. O'Dowd and Smith (1993) identified direct association between surface wind speed and sea salt aerosol generation. Strong winds also cause the resuspension of mineral dust particles from loose soil textures (Gillette, 1999) contributing, on average, 3 μg m^{-3} of particle mass levels on continental background areas (Kavouras *et al*., 2007). At the same time, high wind speed is frequently associated with low pressure systems and precipitation that usually result in the reduction of aerosol levels by dispersion and deposition (Vardoulakis and Kassomenos, 2008; Hussein *et al*., 2005).

The specific aim of this study was to determine the effect of wind speed by wind sector to particle mass concentrations measured in four European urban areas (Amsterdam, Athens, Birmingham and Helsinki) using an empirical, semi- quantitative approach. The approach utilizes a multivariate linear regression model to extract the associations between particle mass and aggregated categorized wind direction and speed. The number of the categories of wind conditions was set as a compromise between the ability to fully represent the non-linear associations of particle mass with wind and to obtain statistically significant regression results. This methodology has been successfully used to estimate the fractional contribution of local particulate sources in 70 background sites in the United States (Kavouras *et al*., 2007). The potential contribution of port activities and shipping emissions was identified for coastal urban areas by examining the outcomes of this analysis conjointly with emission inventories and land use data.

DATA AND METHODS

Aerosol sampling

Daily PM_{10} and $PM_{2.5}$ mass concentrations were continuously measured in urban background monitoring sites in Amsterdam, Birmingham and Helsinki (from October 2002 to March 2004) and in Athens (from October 2002 to December 2004). In Amsterdam, the site was located just outside the Amsterdam-Centrum area, near the Oosterpark, 100 m from the nearest street carrying 13400 cars/day. The monitoring site in Athens was at the Goudi Air Pollution Monitoring Station inside the University of Athens Dental School Campus, with 10,700 cars/day at the nearest street. The sampling sites in Birmingham and Helsinki were located on the University campus areas at Edgbaston and Kumpula districts, respectively. Detailed descriptions of the sampling sites are presented elsewhere (Lianou *et al*., 2011). Samples were collected using Harvard Impactors size selective inlets. Particle mass on conditioned Teflon filters (21 ± 0.5^{o}C and 35 ± 5 %RH) was determined by gravimetric analysis (Puustinen *et al*., 2007). Coarse particle ($PM_{10-2.5}$) mass concentration was the difference between PM_{10} and $PM_{2.5}$ measurements.

Wind conditions regression analysis

Hourly meteorological conditions (wind direction and wind speed at 10 meters above ground level and precipitation) were obtained from local meteorological networks. For this analysis, measurements when precipitation occurred during the sampling period or the day prior to the sampling period were excluded from the analysis to reduce the permutations of the effect of rain on aerosol levels. The hourly wind direction and speed data were transformed into sixteen categorical bins (true=1, false=0) to reduce the number of permutations of wind conditions (direction and speed) and utilize wind direction in the quantitative analysis. Wind direction measurements were allocated into four groups, each one centered in the four cardinal directions (± 45^o) (315 < WD1 ≤ 45, 45 < WD2 ≤ 135, 135 < WD3 ≤ 225, 225 < WD4 ≤ 315). Each group was further split into four categories, namely: 0 < WS1 ≤ 2.2 m s^{-1}, 2.2 < WS2 ≤ 6.2 m s^{-1}, 6.2 < WS3 ≤ 10.2 m s^{-1} and WS4 > 10.2 m s^{-1}. Narrow widths of the categorical wind direction/speed bins would sufficiently describe the non-linear associations of wind conditions and particle mass; however, the large number of variables would reduce the degrees of freedom of the regression model and have a direct effect on the significance of the regression results (Chaloulakou *et al*., 2003; Vardoulakis and Kassomenos, 2007; Kavouras *et al*., 2007). The selected categorical bins generated a reasonable number of variables. The hourly categorical wind direction/speed data for each bin were summed over the 24-hr collection period (noon to noon) of particulate matter.

The relationship between particle mass concentrations (Y, in μg m^{-3}) and categorical wind conditions (X_i, i=1 through 16, unitless) is described as follows (Kavouras *et al*., 2007):

$$Y = a + \sum_{i=1}^{16}(b_i \cdot X_i) \quad (1)$$

where a (in μg m^{-3}) is the intercept and b_i (in μg m^{-3}) are the regression coefficients of the sixteen categorical wind conditions variables. The intercept is the mean particle mass concentration not related to local wind conditions. Ordinal least squares (OLS) analysis was used to estimate the best-fit coefficients in Equation (1) at the 95% significance level. The *F*-value and the adjusted R^2 were applied to assess the adequacy of the regression models. The variance inflation factor values for all independent variables were substantially lower than 10, indicating the absence of nonlinearities among them (Belsley *et al*., 1980).

RESULTS

Table 1 shows the mean (± standard error) of 24-hr $PM_{10-2.5}$ and $PM_{2.5}$ mass concentrations for non-rainy days in the four urban areas. The particle mass varied from 8.6 in Helsinki to 27.5 μg m^{-3} in Amsterdam for fine particles and from 5.9 in Helsinki to 32.6 μg m^{-3} in Athens for coarse particles.

Table 1. Mean (± standard error) of $PM_{10-2.5}$ and $PM_{2.5}$ and mean $PM_{2.5}/PM_{10-2.5}$ ratio for days without rain during the monitoring periods

Urban area	Period	n	$PM_{10-2.5}$ (μg m^{-3})	$PM_{2.5}$ (μg m^{-3})	$PM_{2.5}/PM_{10-2.5}$
Amsterdam	10/2002-3/2004	254	10.5 ± 0.3	27.5 ± 1.1	2.62
Athens	10/2002-12/2004	605	32.6 ± 0.7	25.4 ± 0.5	0.78
Birmingham	10/2002-3/2004	204	9.6 ± 0.5	18.0 ± 1.0	1.87
Helsinki	10/2002-3/2004	239	5.9 ± 0.3	8.6 ± 0.4	1.45

Figures 1 and 2 show the contributions (± 2·error) of wind conditions on $PM_{10-2.5}$ and $PM_{2.5}$ mass concentrations per sector in Helsinki, Athens, Amsterdam and Birmingham. The vertical axis represents the particle mass contributions (radius scaling in the polar plots). Positive values for weak

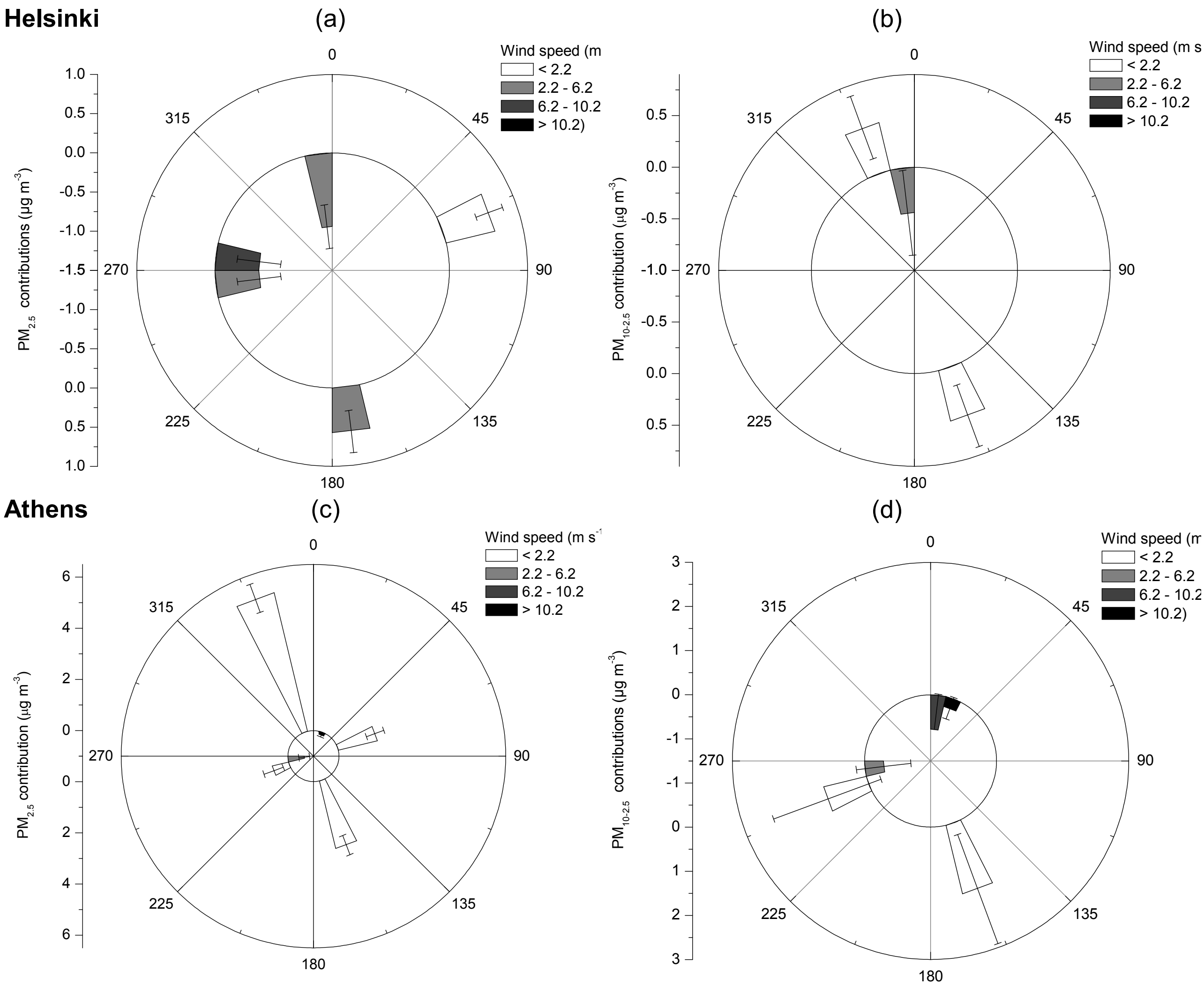

Figure 1. Polar plots of contributions to $PM_{2.5}$ (a, c) and $PM_{10-2.5}$ (b, d) mass concentrations obtained from the regression analysis of measured particle mass concentrations and wind conditions in Helsinki and Athens

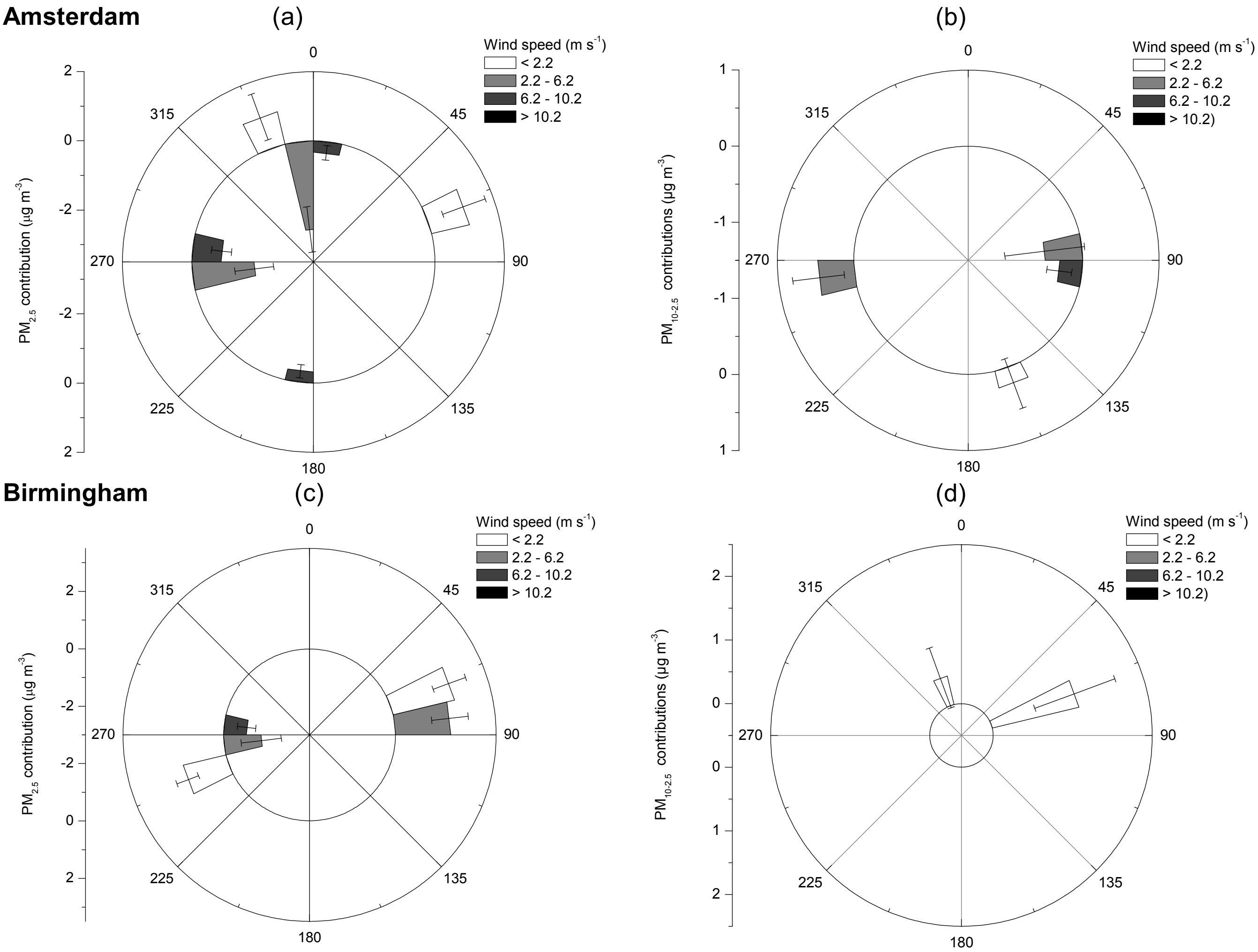

Figure 2. Polar plots of contributions to $PM_{2.5}$ (a,c) and $PM_{10\text{-}2.5}$ (b,d) mass concentrations obtained from the regression analysis of measured particle mass concentrations and wind conditions in Amsterdam and Birmingham

(ws1) and moderate-stronger winds (ws2-ws4), depending on the aerosol fraction, provided an estimate of local fugitive sources (e.g. traffic for $PM_{2.5}$, mechanical resuspension for $PM_{10\text{-}2.5}$) and regional sources (sulfate, nitrate for $PM_{2.5}$)/windblown (sea spray, mineral dust for $PM_{10\text{-}2.5}$), respectively. Negative values suggested that winds, from the given direction, triggered transport of particles away from the urban area. Note that only wind categories with statistically significant association (p-value < 0.2) are reported here. All regression models were statistically significant (at p-value < 0.001) with R^2 values from 0.3 to 0.6. Weak winds exhibited strong positive influence on both fine and coarse particle mass in all cities. Wind direction was different for positive influence on fine particles than those on coarse particles. Stronger winds (ws3-ws4), overall, reduced particle mass levels; however, in some cases in Birmingham, Amsterdam and Helsinki, positive association was observed with moderate winds (ws2).

Weak north and east winds, over less populated areas and agricultural fields, increased $PM_{2.5}$ mass by 2.0 μg m^{-3} (~7%), in Amsterdam. In addition, weak winds from the south and west also contributed, on average, 0.7 μg m^{-3} on $PM_{10\text{-}2.5}$ mass (~16%). The harbor and densely populated communities are included in the south and west sectors. Stronger east (ws2-ws3) and south winds (ws2-ws3) were associated with negative coefficients for coarse and fine particles. In Birmingham, east winds were positively correlated with both $PM_{10\text{-}2.5}$ (1.4 μg m^{-3}) (~24% of measured $PM_{10\text{-}2.5}$) and $PM_{2.5}$ (4.0 μg m^{-3}) (46% of measured $PM_{2.5}$) concentrations. South winds also contributed 1.5 μg m^{-3} on fine particle mass. North winds contributed positively of $PM_{10\text{-}2.5}$ levels for wind speeds.

In Athens, weak wind conditions from all directions accounted for approximately 40% of fine particle mass, with north and south winds being responsible for 8.3 μg m^{-3} of $PM_{2.5}$. Regarding $PM_{10-2.5}$, east and west weak winds showed a positive association contributing 2.6 μg m^{-3} (~8%) on $PM_{10-2.5}$ mass. In Helsinki, wind conditions accounted for 1.0 μg m^{-3} of $PM_{10-2.5}$ (north and south weak winds (ws1)) and 1.2 μg m^{-3} of $PM_{2.5}$ (east weak and south moderate winds).

DISCUSSION

The mean $PM_{2.5}$ and $PM_{10-2.5}$ mass concentrations for days without precipitation in the four urban areas were comparable to those estimated for the entire monitoring period (Lianou et al., 2011). The $PM_{2.5}$/$PM_{10-2.5}$ ratios for Amsterdam, Birmingham and Helsinki were 2.62, 1.87, 1.45, attributed to primarily, traffic emissions in urban areas (Putaud *et al*., 2004). In Athens, the ratio value (0.78) demonstrated the influence of coarse particle sources including road dust and mechanical/windblown resuspension from disturbed soil surfaces (Putaud *et al*., 2004).

In Amsterdam, positive contributions of south and west winds to coarse particle mass may be partially attributed to harbor operations, including unloading and transport of dusty loose materials, west of Amsterdam (Moreno *et al*., 2007) and/or sea spray (van Jaarsveed and Klimov, 2011). Hoek *et al*., (2011) determined that the port was an important determinant of coarse particles in the city. Coarse particle emissions from harbor operations and ships along the coast (west of Amsterdam) ranged from 30.66 to 83.55 Mg (EMEP, 2004).The association of fine particles with north/east winds indicated the influence of emissions within the city of Amsterdam (i.e. traffic) and shipping emissions along the North Sea area (Sax and Larsen, 2004; Dalsoren *et al*., 2009; Matthias *et al*., 2010). Shipping emissions in coastal areas nearby Amsterdam for 2004 varied from 741 Mg to 926 Mg for primary $PM_{2.5}$, from 4707 to 7860 Mg for SO_2 and from 6670 to 11585 Mg for NO_x (EMEP, 2004). Secondary sulfate and nitrate in Amsterdam accounted from 12.4 μg m^{-3} in the summer to 18.4 μg m^{-3} in the winter (Lianou *et al*., 2011).

The western sector of Athens includes the ports of Pireaus, Perama and Elefsina. The three ports carry out the vast majority of sea transports in Greece with coarse particles emissions of 26.17 Mg (EMEP, 2004) as well as major industrial operations (i.e. oil refineries, smelters, waste disposal). Regarding fine particles, wind conditions were typical of the sea-land breeze that circulates air pollutants in the valley indicating that local emissions account for half of measured fine particle mass. Moderate north winds (ws2) reduce coarse particle mass concentration (statistically significant). This effect for coarse particles may be augmented because their sources are already located south of the receptor site, while the sources of fine particles are present throughout the valley. In addition it should be noted that periods with strong winds (ws3-ws4) reduce both fine and coarse particle mass concentrations. Similar conclusions, with respect to coarse particles, were drawn for Helsinki. The nearest part of Helsinki port was located less than 3 km south of the site, with most of the port-related activities being within 6 km south of the site. Regarding fine particles, the dependence on east and south winds may be explained by increased emissions in the Gulf of Finland that also host two big cities, St. Petersburg (250 km east of Helsinki) and Tallinn, Estonia (65 km south of Helsinki). In addition, the city center with high traffic, is located south from the measurement site. Note that secondary inorganic particles account from 2.2. to 5.6 μg m^{-3} of $PM_{2.5}$ (Lianou *et al*., 2011). The association of both fine and coarse particles with east winds (weak and moderate) in Birmingham indicated the influence of sources in central/eastern UK and western Europe. This was corroborated by the $PM_{2.5}$/$PM_{10-2.5}$ ratio of the contributions of weak east winds (1.56).

The method used relied on the statistical associations of particle mass and categorical wind conditions, thus, the results are subjected to several limitations. The primary limitation is that the associations were fixed over time, although emissions of particles and their precursors may be seasonally variable. Secondly, because of the coarse resolution of wind conditions (90^o), each wind condition sector may include areas with different characteristics (e.g. both industrial areas and residential communities). In conjunction with the strict significance criterion for our regression model, these two limitations would lead to weaker regression results and underestimating the possible effect of specific wind conditions. Thirdly, the boundary layer height also influences PM levels; however, its stability and height depends heavily on wind conditions. Thus, this analysis provided a conservative estimate of the relationships of particle mass with wind conditions. These limitations may be eliminated by using larger air pollution and meteorological databases with finer resolution and controlling for seasonal variability.

This analysis provided initial evidence on the possible role of harbor activities and shipping emissions on particle levels in coastal areas. Shipping emissions account from 0.9 to 1.7 million tons of particulate matter annually, with nearly 70% of the emissions occurring within 400 km of the coast (Contini *et al*., 2011; Moldanova *et al*., 2009). Saxe and Larsen (2004) showed that shipping emissions contribute 0.1-0.2 μg m^{-3} to annual $PM_{2.5}$ levels in Copenhagen. Shipping traffic through the straits of Gibraltar/Bay of Algeciras was estimated at 1.4–2.6 μg m^{-3} for PM_{10} (3–7%) and 1.2–2.3 μg m^{-3} for $PM_{2.5}$. It is estimated that shipping emissions contribute as much as or even higher than industrial emissions (Pandolfi *et al*., 2011). This is due to the use of low-quality residual fuels, containing high amounts of sulphur and heavy metals (Pandolfi *et al*., 2011) that are typically high in sulphur (~2-5%) and porphyrins which contain Ni and V (average ash content of 0.072%) (Viana *et al*., 2009; Agrawall *et al*., 2008). In northern Germany, Denmark and southern Sweden the sulphate and nitrate aerosol concentration may be increased by 50% or more because sulphur rich bunker oil is used (burned) in ship engines (Matthias *et al.*, 2010).

CONCLUSIONS

The associations of fine and coarse particle mass measured in four European urban areas with wind conditions were examined using a heuristic regression analysis method. Positive and negative impacts of wind conditions for both fine and coarse particle mass were identified. The negative dependence of moderate and strong winds for both fine and coarse particles verified the removal and transport of particles from the urban atmosphere. Weak winds accounted for 1.2 μg m^{-3} to 10.5 μg m^{-3} of $PM_{2.5}$ mass, and for 0.7 to 2.5 μg m^{-3} of $PM_{10-2.5}$ mass in all urban areas. The orientation of positive correlations for coarse particles with respect to harbors in Amsterdam, Athens and Helsinki indicated that emissions of loose material during port operations and sea spray contributed from 6% to 17% towards coarse particle mass. For fine particles, local emissions dominated fine particles in Athens, while regional sources of secondary inorganic aerosol (including shipping emissions) appeared to influence $PM_{2.5}$ levels in Amsterdam and Helsinki.

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REFERENCES

Belsley, D.A., Kuh, E., Welsch, R.E. (1980). Regression Diagnostics: Identifying Influential Data and Sources of Collinearity. New York: John Wiley and Sons.

Chaloulakou, A., Kassomenos, P., Spyrellis, N., Demokritou, P., and Koutrakis, P. (2003), Measurements of PM_{10} and $PM_{2.5}$ particle concentrations in Athens, Greece*,* *Atmospheric Environment*, **37**, 649-660.

Contini, D., Gambaro, A., Belosi, F., De Pieri, S., Cairns, W.R.L., Donateo, A., Zanotto, E. and Citron M. (2011), The direct influence of ship traffic on atmospheric $PM_{2.5}$, PM_{10} and PAH in Venice, *Journal of Environmental Management*, **92**, 2119-2129.

Dalsoren, S.B., Eide, M.S., Endresen O., Mjelde, A., Gravir, G. and Isaksen I.S.A. (2009), Update on emissions and environmental impacts from the international fleet of ships: the contribution from major ship types and ports, *Atmospheric Chemistry and Physics*, **9**, 2171-2194.

Gillette, D. (1999), A qualitative geophysical explanation for ‘‘hot spot’’ dust emitting source regions, *Beitraege zur Physik der Atmosphaere*, **72**, 67–77.

Hoek, G., Beelen, R., Kos, G., Dijkema, M., van der Zee, S., Fischer, P.H. and Brunekreef, B. (2011), Land use regression model for ultrafine particles in Amsterdam, *Environmental Science and Technology*, **45**, 622-628.

Hussein, T, Hämeri, K, Aalto P.P. and Kulmala, M. (2005), Modal structure and spatial-temporal variations of urban and suburban aerosols in Helsinki area, *Atmospheric Environment*, **39**, 1655–1668.

Jones, A.M., Harrison, R.M. and Baker, J. (2010), The wind speed dependence of the concentrations of airborne particulate matter and NOx, *Atmospheric Environment*, **44**, 1682-1690.

Kavouras, I.G., Etyemezian, V., DuBois, D.W., Xu, J. and Pitchford, M. (20090, Source reconciliation of atmospheric dust causing visibility impairment in class I areas of the western United States, *Journal of Geophysical Research-Atmospheres*, **114**, D02308.

Kavouras, I.G., Etyemezian, V., Xu, J., DuBois, D.W., Green, M. and Pitchford, M. (2007), Assessment of the local windblown component of dust in the Western United States, *Journal of Geophysical Research-Atmospheres,* **112**, D08211.

Lianou, M., Chalbot, M., Kavouras, I.G., Kotronarou, A., Karakatsani, A., Analytis, A., Katsouyanni, K., Puustinen, A., Hameri, K., Vallius, M., Pekkanen, J., Meddings, C., Harrison, R.M., Ayres, J.G., ten Brick, H., Kos, G., Meliefste, K., de Hartog, J. and Hoek, G. (2011), Temporal variations of atmospheric aerosol in four European urban areas, *Environmental Science and Pollution Research*, **18**, 1202-1212.

Lianou, M., Chalbot, M., Kotronarou, A., Kavouras, I.G., Karakatsani, A., Katsouyanni, K., Puustinnen, A., Hameri, K., Vallius, M., Pekkanen, J., Meddings, C., Harrison, R.M., Thomas, S., Ayres, J.G., ten Brink, H., Kos, G., Meliefste, K., de Hartog, J.J. and Hoek, G. (2007), Dependence of home outdoor particulate mass and number concentrations on residential and traffic features in urban areas, *Journal of the Air & Waste Management Association*, **57**, 1507-1517.

Matthias, V., Bewersdorff, I., Aulinger, A. and Quante, M. (2010), The contribution of ship emissions to air pollution in the North Sea regions, *Environmental Pollution*, **158**, 2241-2250.

Moldanova J., Fridell E., Popovicheva O., Demirdjian B., Tishkova V., Faccinetto A. and Focsa C. (2009), Characterisation of particulate matter and gaseous emissions from a large ship diesel engine, . *Atmospheric Environment*, **43**, 2632-2641.

Moreno, N., Alastuey, A., Querol, X., Artirano, B., Guerra, A., Luaces, J.A., Lorente, A. and Basora, J. (2007), Characterization of dust material emitted during harbor operations, *Atmospheric Environment*, **41**, 6331-6343.

O'Dowd, C. D. and Smith, M.H. (1993), Physicochemical properties of aerosols over the northeast Atlantic: Evidence for wind-speed-related submicron sea-salt production, *Journal of Geophysical Research-Atmospheres*, **5**, 1137–1149.

Pandolfi M., Gonzalez-Castanedo Y., Alastuey A., de la Rosa J.D., Mantilla E., Sanchez de la Campa A., Querol X., Pey J., Amato F. and Moreno T. (2011), Source apportionment of PM10 and PM2.5 at multiple sites in the strait of Gibraltar by PMF: impact of shipping emissions, *Environmental Science and Pollution Research*, **18**, 260–269.

Puustinen, A., Hameri, K., Pekkanen, J., Kulmala, M., De Hartog, J., Meliefste, K., Ten Brink, H., Kos, G., Katsouyanni, K., Karakatsani, A., Kotronarou, A., Kavouras, I., Meddings, C., Thomas, S., Harrison, R.M., Ayres, J.G., Van der Zee, S. and Hoek, G. (2007), Spatial variation of particle number and mass over four European cities, Atmospheric Environment, **41**, 6622-6636.

Saxe, H. and Larsen T. (2004) Air pollution from ships in three Danish ports, *Atmospheric Environment*, **38**, 4057-4067.

Van Jaarsveed, JA. and Klimov, D. (2011), Modeling the impact of sea salt particles on the exceedances of daily PM1 air quality standards in the Netherlands, *International Journal of Environment and Pollution*, **44**, 217-225.

Vardoulakis, S. and Kassomenos, P. (2008), Sources and factors affecting PM10 levels in two european cities: Implications for local air quality management, *Atmospheric Environment*, **42**, 3949-3963.

Viana M., Amato F., Alastuey A., Querol X., Moreno T., Dos Santos S.G., Herce M. D. and Patier R.F. (2009), Chemical Tracers of Particulate Emissions from Commercial Shipping, *Environmental Science and Technology*, **43**, 7472–7477.