Air Quality in Postunification Erfurt, East Germany: Associating Changes in Pollutant Concentrations with Changes in Emissions

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The unification of East and West Germany in 1990 resulted in sharp decreases in emissions of major air pollutants. This change in air quality has provided an opportunity for a natural experiment to evaluate the health impacts of air pollution. We evaluated airborne particle size distribution and gaseous co-pollutant data collected in Erfurt, Germany, throughout the 1990s and assessed the extent to which the observed changes are associated with changes in the two major emission sources: coal burning for power production and residential heating, and motor vehicles. Continuous data for sulfur dioxide, total suspended particulates (TSP), nitric oxide, carbon monoxide, and meteorologic parameters were available for 1990–1999, and size-selective particle number and mass concentration measurements were made during winters of 1991 and 1998. We used hourly profiles of pollutants and linear regression analyses, stratified by year, weekday/weekend, and hour, using NO and SO_2 as markers of traffic- and heating-related combustion sources, respectively, to study the patterns of various particle size fractions. Supplementary data on traffic and heating-related sources were gathered to support hypotheses linking these sources with observed changes in ambient air pollution levels. Substantially decreased (19–91%) concentrations were observed for all pollutants, with the exception of particles in the 0.01–0.03 μm size range (representing the smallest ultrafine particles that were measured). The number concentration for these particles increased by 115% between 1991 and 1998. The ratio of these ultrafine particles to TSP also increased by more than 500%, indicating a dramatic change in the size distribution of airborne particles. Analysis of hourly concentration patterns indicated that in 1991, concentrations of SO_2 and larger particle sizes were related to residential heating with coal. These peaks were no longer evident in 1998 due to decreases in coal consumption and consequent decreased emissions of SO_2 and larger particles. These decreases in coal combustion and the decreased concentrations of SO_2 and particles of larger size classes may have led to decreased particle scavenging and may be partially responsible for the observed increases in ultrafine particles. Traffic-related changes, such as increased numbers of trucks and increased use of diesel vehicles in Erfurt, were also associated with increased number concentrations of ultrafine particles. Morning particle peaks of all sizes were associated with NO and CO (markers for traffic) in both the 1991 and 1998 periods. There were significant differences in the ultrafine particle levels for morning hours between 1991 and 1998, suggesting that traffic was the cause of this increase. *Key words*: air pollution, coal combustion, environmental exposure, motor vehicles, particles, sulfur dioxide, ultrafine particles. *Environ Health Perspect* 109:325–333 (2001). [Online 7 March 2001] *http://ehpnet1.niehs.nih.gov/docs/2001/109p325-333ebelt/abstract.html*

The unification of East and West Germany in 1990 brought major social and political changes, particularly affecting East Germany (*1*). This restructuring resulted in significant changes in emissions of air pollutants. During the 1980s, the former German Democratic Republic was a major industrial power with a high level of manufacturing output. This high output was fed by huge inputs of natural resources, which in turn led to extreme emissions of pollutants (*2*). In 1990, the European Union (*3*) concluded,

> the environment in East Germany is in a catastrophic state. Water and air pollution is so bad that it is no longer simply a matter of cleaning up the environment but one of restoring the most basic living conditions.

Clean up in East Germany initially occurred as a by-product of unification; low demand for products and energy led to a collapse of almost the entire industrial and agricultural structure in East Germany, sharply decreasing emissions between 1989 and 1991. This was followed by air pollution controls implemented between 1992 and 1996 (*4,5*). A decade has passed since the German reunification, and it is timely to examine the results of these structural changes and decreased emissions on the air quality.

The sharp change in emissions in the past 10 years has been an opportunity for a natural experiment to evaluate the health impacts of air pollution. For example, decreases in total suspended particles (TSP) and sulfur dioxide between 1991 and 1995 were associated with decreased prevalence of infectious airway diseases in East German children (*6,7*). Additionally, Erfurt, a city in former East Germany, was the site of several large-scale epidemiologic studies during the 1980s (*8*) and early 1990s (*9*) concerning the health effects of air pollution. Since this time, the composition of Erfurt's ambient air has changed, and assessments of the effects of these changes on the health status of the former East German population are currently in progress (*10*).

Current particulate air pollution health effects research is focused on evaluating biologic mechanisms that may explain the epidemiologic associations (*11*). Exposure assessment and analysis of the composition of ambient particles and the impact of specific sources is of special interest. Most epidemiologic studies have assessed exposure of the study population in terms of particulate mass concentrations (i.e., in micrograms per cubic meter), which are measured for specific particle cut sizes. For example, the mass concentrations of particles with aerodynamic diameters (d_a) ≤ 10 μm or 2.5 μm provide PM_{10} or $PM_{2.5}$ concentrations, respectively. Seaton et al. (*12*) suggested that ultrafine particles (particles with diameters < 0.1 μm) could be a component of the ambient particle mixture that is responsible for the observed health effects and suggested a mechanistic hypothesis. Because ultrafine particles do not contribute significantly to the total mass of particles, measurements based only on mass concentrations do not accurately represent the concentrations of ultrafine particles; thus ultrafine particles have been quantified in terms of particle number concentrations. Several epidemiologic studies have evaluated the impacts of ultrafine particle levels. In Erfurt, an increase

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in the number of ultrafine particles was associated with a decrease in peak expiratory flows (PEF) of asthmatic adults, which was stronger than the effects of PM_{10} (*13,14*). In Erfurt, associations have also been found between ultrafine particles and medication use by asthmatics and symptom reporting by coronary heart disease patients (*15*). A study in Finland on the PEF of asthmatic children did not indicate a stronger association with ultrafine particles, but consistent and significant results were obtained with PM_{10} and black smoke exposure metrics (*16*).

From extensive monitoring conducted in Erfurt throughout the 1990s, including gravimetric methods and aerosol size spectrometry, the concentrations of particles both in terms of their mass and number have been followed over time. Detailed assessments of the size distribution of particles have also been undertaken. For example, during the winter of 1991–1992, the mean number concentration was largely composed of ultrafine particles (72%), whereas the bulk of the particle mass (83%) was due to accumulation mode particles, which are in the size range of 0.1–0.5 µm (*17*). Since this time, the mass concentrations of fine particles ($PM_{2.5}$) have decreased by 72%; however, the overall number concentrations have remained constant, with particles < 0.03 µm increasing by 82% (*18,19*). Due to the potential health risks associated with ultrafine particles, the source of the observed increases in recent years is of immediate concern. Additionally, identification of important sources contributing to ambient concentrations of ultrafine particles may provide useful information to support the development of reduction policies in this and in other locations.

Two source categories that are large contributors to air pollution and that have undergone significant change in East Germany since the unification include energy production (power plants and residential heating) and vehicles (*5*). As was common throughout East Germany before the unification, lignite (brown coal) was the major fuel, meeting approximately 70% of the region's energy requirements (*4*). Lignite has high sulfur content, and its combustion results in high emissions of SO_2 and particulate matter (PM) when burned (*20*). At the time of unification in 1990, lignite-fired power plants in the former East Germany operated with minimum emission controls. Since this time there have been large reductions in emissions of particles and SO_2 from these sources due to the shutdown of old plants, transition from the use of coal to liquid and gaseous fuels, reduction in the sulfur content of coal, and retrofits of lignite-fired power plants with flue gas desulfurization systems (*4,5,21*). West German plants had already achieved emission reduction in the 1980s; thus the unification of East and West Germany brought together two power-generation systems. Thuringia, the state in which Erfurt is located, is an industrial region. It still has higher atmospheric SO_2 concentrations relative to other German states (with values sometimes > 50 µg/m^3), but much lower than in 1990 or earlier, when annual SO_2 average concentrations were > 150 µg/m^3 (*5*).

Concurrent with the changes in energy production, mobile sources have undergone transitions. Due to the absence of other available vehicles, the car fleet in East Germany was largely composed of "Trabants" prior to unification. These were small cars with 26 horsepower, two-stroke engines and visibly high exhaust emissions (*1*). After unification the fleet was replaced with vehicles produced largely by Western countries (*1*), which had comparatively modern engine technology, including three-way catalysts. Additionally, during this same period, diesel-powered vehicles have become more common throughout Western Europe (*22,23*). Further, most motorcycles during the preunification period were equipped with two-stroke engines.

Significant changes in fuels and combustion processes associated with energy production and motor vehicles in East Germany have occurred, and these have likely impacted the ambient pollutant concentrations over time. In this study we used a unique database of particle size measurements and continuous gaseous air pollutant data from Erfurt to initially describe their temporal patterns during the 1990s. Furthermore, we assessed the extent to which the observed changes in particle size distribution are associated with changes in specific emission sources—namely, energy- and traffic-related combustion sources.

Methods

Data collection. Erfurt (population approximately 201,100) is a city in the state of Thuringia, Germany, approximately 200 m above sea level and about 100 km east of the former east–west border. It lies on a flat plain, surrounded on all sides by a 100-m high ridge, except toward the north. This geography favors wintertime inversions, which result in elevated levels of ambient air pollution during this season. Local air pollution sources include motor vehicles, residential heating, small-scale industry, and district heating plants. Suburban areas contain large apartment complexes that are heated with steam supplied by a large coal-burning power plant located several kilometers northeast of the city center. The inner city has a large historical center where buildings were heated by individual coal furnaces until the time of reunification.

Ambient particulate, gaseous, and meteorologic data were collected in Erfurt throughout the 1990s and have been used previously in numerous epidemiologic and exposure assessment studies (*13,17,24–26*). Between 1 October 1991 and 31 March 1992, and between 1 October 1995 and 31 March 1999, measurements were made at the Institute of Hygiene site located approximately 1 km south of the Erfurt city center and approximately 40 m from the nearest major road. The spatial representativeness of this site has been analyzed in detail and is generally representative of the air quality within Erfurt (*26*). In previous work, PM_{10} and sulfate measurements from this site were found to be significantly correlated (Spearman rank correlations: 0.69–0.85) with those made at other locations within Erfurt, although some spatial variability was evident during periods of low wind speed.

The number concentrations (NCs) of ambient fine particles were determined with a mobile aerosol spectrometer (MAS) as previously described by Brand et al. (*27*). The MAS is a combination of two instruments that measure different size ranges. Particles with diameters between 0.01 and 0.5 µm are quantified using a differential electrical mobility sizer (DMPS). The DMPS consists of a differential electrical mobility analyzer (TSI Model 3071; TSI, St. Paul, MN, USA) used to classify the particles by their electrical mobility, which are then counted by a condensation particle counter (TSI models 3760, 3010). Particles with diameters between 0.1 and 2.5 µm are classified with an optical laser aerosol spectrometer (LAS-X; Model LAS-X; PMS Inc., Boulder, CO, USA). The instruments used at the Erfurt site are described in more detail elsewhere (*17,28*). Overall, the particle number distribution of the ambient aerosol (using the 13 different size channels from the DMPS and 15 channels for each of 4 size ranges from the LAS-X) was measured every 6 min. The mass distribution of particles was determined from the number distribution basing calculations on a mean particle density of 1.5×10^3 kg/m^3, and an assumption of spherical particles (*17*). Sulfate concentrations were determined from 24-hr $PM_{2.5}$ Harvard Impactor (with the addition of a citric acid-coated honeycomb denuder for acidity sampling) measurements taken on a daily basis during winter 1991 and every second day after 1995. Filters were analyzed for SO_4^{2-} by ion chromatography. We measured nitric oxide separately from nitrogen dioxide with a two-channel chemiluminescence monitor (models NH3OM, AC31M; Environment S.A., Poissy Cedex, France). Sulfur dioxide was measured via UV absorption (Model AF21M; Environment S.A.). Both monitors

operated on a continuous basis, producing 3-m averaged measurements. Temperature, relative humidity (Model FR3205-M; RCI, Planegg, Germany) and windspeed sensors were mounted on the sampling shelter, thereby providing continuous meteorologic data. Additionally, hourly measurements for SO_2, TSP, NO, and CO from as early as 1990 was available from governmental monitoring stations (Thueringer Landesanstalt fuer Umwelt).

Data analysis. The continuous number concentration data obtained from the DMPS and LAS-X channels were only stored for certain size classes, from which we chose three ranges for analysis. The smallest fraction included particles with diameters between 0.01 and 0.03 µm ($NC_{0.01–0.03}$), thus capturing the lowest measurable portion of ultrafine particles (ultrafine particles are usually defined as particles with sizes up to 0.1 µm). The second size class included particles with diameters between 0.03 and 0.5 µm ($NC_{0.03–0.5}$), covering the upper portion of the ultrafine mode and the lower portion of accumulation mode particles (which are defined as particles between 0.1 and 1.0 µm). The largest size class contained all particles in the size range between 0.5 and 2.5 µm diameter ($NC_{0.5–2.5}$), thus including the upper portion of the accumulation mode as well as fine-mode coarse particles. These size class divisions were slightly different from previous reports using these measurements, since the continuous data needed for hourly assessments was stored for limited size classes only (*13,14,17,28*).

From the calculated mass distribution, $PM_{2.5}$ concentrations were determined by adding over all particle size ranges between 0.01 and 2.5 µm. For all NC, $PM_{2.5}$, gaseous (NO, NO_2, CO, SO_2), and meteorologic variables (temperature, relative humidity, wind speed), the measurements from each hour were averaged. The resulting hourly measurements were then used to obtain 24-hr (daily) averages. For SO_4^{2-} only daily averages were available. We used daily averages to assess the distributions of individual pollutants as well as their temporal patterns. For most variables, measurements during the 1991 monitoring period were only taken from 1 October 1991 to 31 March 1992; no summer data were collected. In addition, gaseous measurements for January to March 1992 were not available. Due to these gaps in the data, two distinct time periods were chosen for further comparative analyses: 3-month time windows from October to December were compared for the years 1991 and 1998.

Analyses included *t*-tests for differences in mean concentrations between these years. We created hourly profiles by plotting the median values for each hour of the day for each time period (for NC, NO, CO, and SO_2). These data were stratified by weekday and weekend periods. To visually compare hourly time series for the different pollutants, the hourly median values were standardized by calculating their percentage of the daily average concentration. Hourly data were subsequently assessed by linear regression analyses using the univariate general linear model procedure in SPSS version 9.0 (SPSS, Chicago, IL, USA). We predicted NC fractions using year and

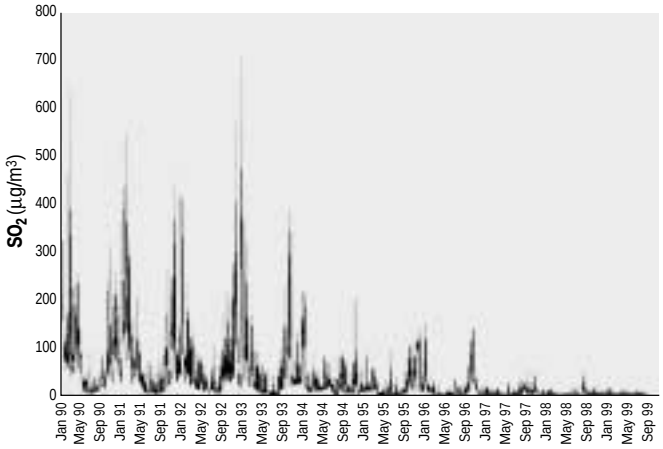

Figure 1. Daily SO_2 time series. Data from the Thueringer Landesanstalt fuer Umwelt monitoring network.

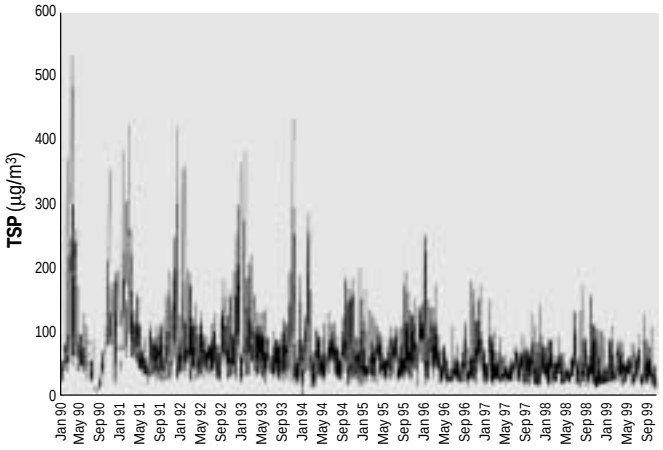

Figure 2. Daily TSP time series. Data from the Thueringer Landesanstalt fuer Umwelt monitoring network.

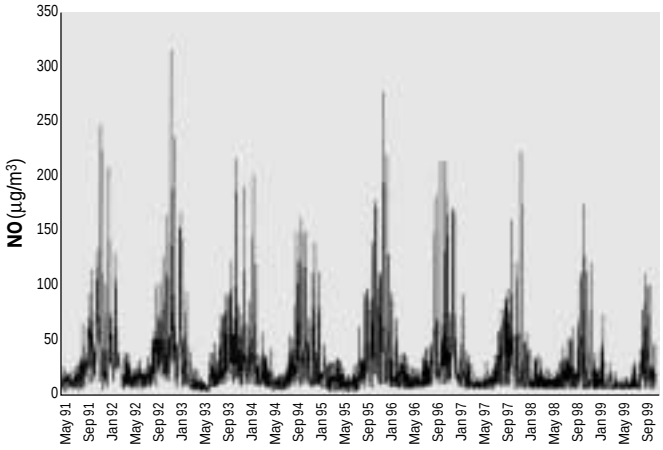

Figure 3. Daily NO time series. Data from the Thueringer Landesanstalt fuer Umwelt monitoring network.

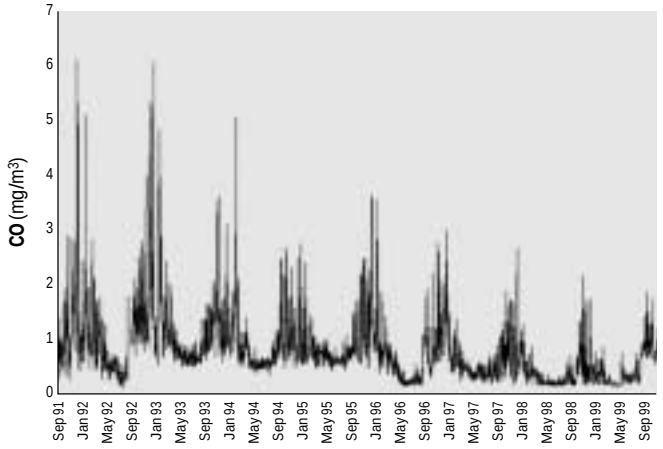

Figure 4. Daily CO time series; data from the Thueringer Landesanstalt fuer Umwelt monitoring network.

weekend/weekday as fixed factors and NO and SO_2 concentrations as covariates. Autocorrelation of measurements over time was addressed by using the hour of the day as an additional fixed factor. We also investigated effects of two-way interactions between the independent variables. Variables were not transformed for these analyses. For ease of comparison, effects of NO and SO_2 were assessed in the same models, although there was a relatively high correlation between them (Spearman rank correlation, 0.67); their effects as covariates in separate models were similar.

Results

Trends over time. Due to incomplete hourly data for several of the pollutants at various times within the period 1990–1999, we focused analyses on two shorter periods for which complete data were available: October–December of 1991 and 1998. These periods were also selected because they represent the extremes of long-term trends in concentrations for most pollutants (Figures 1–4). SO_2 and TSP (Figures 1 and 2) exhibited steady declines in concentration between 1991 and 1998, largely due to the minimization of winter extremes. NO and CO (Figures 3 and 4) concentrations also decreased, but to a lesser extent. Analyses were restricted to winter periods because the typical seasonal pattern in Erfurt exhibits high concentrations in winter months and lower concentrations in summer months.

Table 1 presents summary statistics for each variable for 1991 and 1998. The variables were not normally distributed, with the exception of temperature and relative humidity. $NC_{0.03–0.5}$, NO, and NO_2 were approximated by lognormal distributions, whereas all other metrics were not better represented when log transformed. Both arithmetic and geometric descriptive statistics are reported for consistency. Based on comparison of arithmetic means, concentrations decreased significantly for all pollutants (*t*-test, $p \leq 0.001$). For example, we observed reductions of 58% for TSP, 74% for $PM_{2.5}$, 50% for SO_4^{2-}, and 91% for SO_2. In contrast, $NC_{0.01–0.03}$ increased by 115%. The $NC_{0.01–0.03}$:TSP ratio changed from 78 to 394, which is a 5-fold increase from 1991 to 1998. The $NC_{0.01–0.03}$:$PM_{2.5}$ ratio changed by a factor of 8, from 83 to 686. These ratios emphasize the drastic shift of the aerosol size distribution toward the smaller, ultrafine particles from 1991 to 1998. Relative humidity was slightly higher in 1998 relative to 1991 ($p < 0.001$), while ambient temperatures were similar during the two periods. The mean windspeed was lower in 1998 than in 1991.

From these initial observations we developed several hypotheses to evaluate the impact of various sources that have changed over time. First, the main fuels used for energy production have changed from brown coal to natural gas. Lowering the PM emissions from high-sulfur coal combustion could lead to the decreases observed for $NC_{0.03–0.5}$ and $NC_{0.5–2.5}$. Such decreases could in turn lead to decreases in particle scavenging, thus allowing the $NC_{0.01–0.03}$ fraction to increase. Particle scavenging may occur when large numbers of fine and coarse particles provide surface area onto which ultrafine particles diffuse and readily aggregate. Reductions in the emissions of these larger particles could reduce the available surface area and therefore increase the atmospheric residence time of ultrafine particles. Second, there has been an influx of Western-style vehicles employing modern engine technology in eastern Germany. It has been hypothesized that improved efficiencies associated with modern combustion engines result in more complete combustion and larger proportions of particles < 0.03 µm. Increases in the number of diesel vehicles in Erfurt could also contribute to increased ultrafine particle counts.

As there was no direct measure for attributing changes in these sources to changes in ambient particle concentrations, we evaluated the hour-to-hour changes in the concentrations of gaseous pollutant markers in relation to each particle size fraction. Gaseous pollutants and particle size fractions showing similar temporal structures were assumed to have arisen from the same sources. For our purposes, hourly SO_2 data were used as an indicator for coal-fired power generation and residential heating,

Table 1. Descriptive statistics for each pollutant by year.

								1998/1991 Ratio	
Measurement	Year	No.[a]	AM	SD	Range	GM	GSD	AM	GM
$NC_{0.01–0.03}$	1991	70	7,896	7,252	369–25,657	4,159	4	2.15	3.48
(*n*/cm^3)	1998	75	16,982	9,672	4,045–51,620	14,491	2		
$NC_{0.03–0.5}$	1991	70	10,593	8,624	1,121–39,073	7,324	3	0.63	0.74
(*n*/cm^3)	1998	75	6,669	4,896	1,436–25,177	5,427	2		
$NC_{0.5–2.5}$	1991	70	122	128	8–625	78	3	0.20	0.18
(*n*/cm^3)	1998	75	25	35	2–186	14	3		
TSP[b]	1991	70	101.34	77.76	19.85–421.83	79.68	2.05	0.42	0.47
(µg/m^3)	1998	75	43.05	28.56	12.00–157.00	37.55	1.73		
$PM_{2.5}$	1991	70	95.01	85.57	10.51–402.10	67.85	2.36	0.26	0.29
(µg/m^3)	1998	75	24.75	21.28	3.74–102.98	19.87	2.02		
SO_4^{2-}	1991	87	8.24	6.84	1.01–32.59	7.16	2.06	0.50	0.51
(µg/m^3)	1998	38	4.11	5.25	0.39–22.29	3.68	2.09		
NO	1991	69	47.43	47.06	6.38–231.82	32.73	2.45	0.59	0.46
(µg/m^3)	1998	90	22.75	33.90	0.30–189.09	11.35	3.36		
NO_2	1991	69	39.43	18.34	11.95–106.57	36.40	1.61	0.81	0.81
(µg/m^3)	1998	90	28.80	16.48	6.35–81.70	25.76	1.74		
CO[b]	1991	92	1.45	1.09	0.21–6.18	2.27	1.43	0.33	0.63
(mg/m^3)	1998	92	0.48	0.42	0.10–2.15	1.43	1.27		
SO_2	1991	69	91.23	84.04	6.35–365.15	62.52	2.53	0.09	0.11
(µg/m^3)	1998	90	6.58	7.32	0.33–50.87	5.63	2.09		
Temperature	1991	92	4.22	5.04	–7.20–14.30	—	—	0.99	—
(°C)	1998	90	4.18	5.19	–6.86–15.27	—	—		
RH	1991	92	75	9	55–94	—	—	1.17	—
(%)	1998	90	88	6	67–100	—	—		
Windspeed	1991	92	2.19	1.23	0.00–5.70	2.96	1.49	0.68	0.79
(m/sec)	1998	90	1.49	0.89	0.38–4.83	2.35	1.39		

Abbreviations: AM, arithmetic mean; GM, geometric mean; GSD, geometric standard deviation; RH, relative humidity. Different numbers of samples were collected for the various particle measurements, but for comparison purposes, summaries of NC, TSP, and $PM_{2.5}$ were based only on days for which each of these measurements were available.
[a]Number of 24-hr averages out of a possible 92 days for each variable. [b]Data from Thueringer Landesanstalt fuer Umwelt–Kraempferstrasse site; all other measurements were from Institute of Hygiene/GSF site.

and NO and CO data were used as indicators for traffic-related emissions.

Temporal patterns in energy production. Ambient sulfur oxide gases are formed largely when fuels containing sulfur, such as coal and oil, are burned (*20*). In East Germany before the unification, sulfur-rich coal was typically used as a source of energy for industry and heating purposes (*4*). Homes were either heated by individual coal-burning ovens or through long-distance steam pipelines fed by coal-fired power plants. SO_2 was used as a marker of such heating-related combustion.

In 1991, morning SO_2 peaks were more prominent during the week compared to the weekend (Figure 5). This pattern is consistent with morning peaks being due to energy use by industry during the workday. Evening SO_2 peaks occurred to the same extent on weekends as during the week, and these were thought to be related to home heating. These curves are consistent with typical home heating patterns during this period. The 1998 profiles were much weaker, with an altered hourly pattern. No longer were morning and evening peaks observed; rather, only one prominent and prolonged mid-day increase was present.

Supplemental data on energy production were gathered to help explain the observed changes in SO_2 concentrations. Over the past 9 years, the percentages of homes heated locally (60%) compared to those heated by long-distance steam pipelines (40%) did not change, but the sources of energy used by either route did (Table 2). In the Erfurt area there were four power plants in operation before unification, three of which were subsequently shut down between 1990 and 1993; the fourth plant was retrofitted. Between 1991 and 1998, the use of coal by power plants decreased by 84% and was replaced by a 3-fold increase in natural gas consumption. Also, between 1992 and 1995, the use of coal for residential heating decreased by 45%. Additionally, the sulfur content of coal was reduced from 2% in 1991 to 1.7% in 1993, after which the percentage remained constant. The decreased use of coal and increased use of gas can explain the large difference in SO_2 concentration between 1991 and 1998 in Erfurt. The small SO_2 elevations during the day in 1998 could be due to the remaining coal that was still used at this time or due to regional transport.

Temporal patterns in traffic. NO and CO are important air pollutants associated with motor vehicle emissions (*20*). On-road motor vehicles contribute up to 35% of the total nitrogen oxides (NO_x) emissions in the United States, of which more than 95% of NO_x from light-duty, gasoline vehicles is released as NO (*29*). Hourly NO and CO profile plots (Figures 6 and 7) support the use of these pollutants as traffic indicators in Erfurt. During the week, morning NO and CO peaks were observed that corresponded to the morning rush hour. In 1991, these morning NO and CO peaks occurred slightly earlier than morning SO_2 peaks, supporting the difference in sources of these pollutants: NO and CO arising from travel to work and SO_2 being emitted from factories in operation during the workday. Unfortunately, no traffic count data were available with which to directly verify the observations.

Peaks in NO and CO concentrations did not occur to the same extent on weekends, likely due to less traffic on the road. Over time, the weekday morning traffic peak shifted: in 1998, the peak occurred an hour later and was longer. This could be due to changes in work patterns that delay and lengthen the rush hour. Before unification, morning shifts in large factories usually started at 0700 hr. After the shutdown of the large plants and the implementation of flexible work hours, traffic was likely shifted toward start of regular business hours around 0900 hr.

We collected supplementary data on vehicles in Erfurt to help explain the reductions in NO and CO concentrations and differences in the timing of concentration peaks between 1991 and 1998. In Erfurt, the total number of vehicles (cars, motorcycles, trucks) over the last 7 years did not change (Table 3). However, a large increase (268%) in the percentage of trucks and a large decrease in the number of vehicles equipped with two-stroke engines, including a drastic decrease in the number of motorcycles (83%), was observed. The fraction of trucks with diesel motors has been steadily increasing in Germany in the past 20 years, from 58% in 1980 to 88% in 1999 (*30*). Since 1990, the number of diesel trucks in Gemany has doubled (*30*).

In terms of cars, the total number has doubled in East Germany since 1991 (*5*). In Thuringia, the number also increased by a factor of 1.6 (*30*), although in Erfurt, the number of private cars increased only slightly (15%) between 1990 and 1998 (Table 3). However, the composition of the private car fleet has likely changed dramatically in eastern Germany due to modern technology and diesel motors. For example, even though the proportion of diesel cars in Thuringia (7–8%) has been lower than in the country (13%) during the 1990s, the total number of

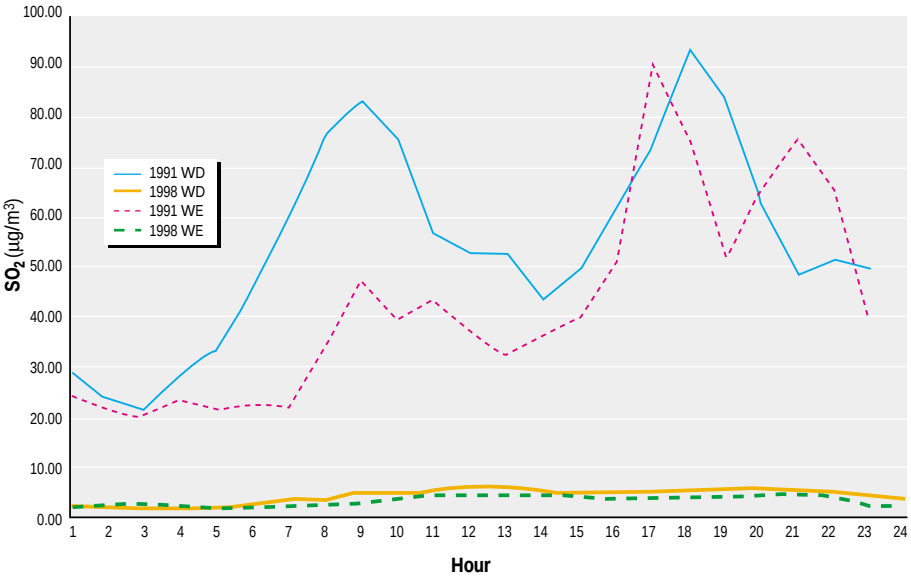

Figure 5. SO_2 hourly profiles for 1991 and 1998, showing weekends (WE) and weekdays (WD) separately.

Table 2. Temporal changes in energy sources.

Energy source/type	Measure	1991	1998	1998/1991 Ratio
Home heating	Percent of homes	40	40	1
Steam pipe (long distance)[a]				
Residential (total)[a]	Percent of homes	60	60	1
Residential (coal)[b]	Percent of homes	60	33	0.55
Power plant				
Coal[a]	Tons	565,172	92,243	0.16
Oil[c]	Tons	12,620	13,716	1.09
Gas	$m^3 \times 10^3$	26,492	102,439	3.87

Residential refers to heating source present in the residence itself.
[a]Data from Schmidt (*42*). [b]Data from the microcensus in 1992 and 1995 (*43*). [c]Data from Strom und Fernwaerme GmbH (*44*).

diesel vehicles has more than doubled since 1992, increasing from 11% to 16% of the total on-road vehicle population (*30*).

Relating emissions to particle levels. The emissions from heating and traffic (as assessed by SO_2 and NO/CO) were related to particulate measurements to determine the effect of these sources on the ambient particulate mix. Standardized weekday hourly profile plots (which combined the number concentrations of the three size classes of particles and the gases) were created (Figures 8–11) and regression models using these variables (except CO) were constructed (Table 4). We conducted visual and statistical analyses to assess the patterns over time (between 1991 and 1998), between weekdays and weekends and between the hours of a day.

From the regression models, the $NC_{0.01–0.03}$ fraction was significantly lower in 1991 compared to 1998, as also shown by the ratio comparisons (Table 1). As indicated by the weekend/weekday factor (Table 4), all particle fractions were significantly lower on weekends compared to weekdays. This supports traffic and energy production (when combining industry and residential sectors) as contributors to all size fractions, as they are both expected to be higher during the week. Both visual assessments of the hourly profiles and regression models indicated significant positive associations between all particle size fractions and both of the gaseous pollutants, NO and SO_2. Considering interactions between winter and the gases (winter × NO, winter × SO_2), the effects of both NO and SO_2 on $NC_{0.01–0.03}$ were significantly different in 1991 compared to 1998. This suggests that the influences of both traffic and energy production on $NC_{0.01–0.03}$ have changed over time.

Reviewing these results and the literature, there are several reasons to suggest that traffic is the major factor associated with the observed increase in $NC_{0.01–0.03}$. First, due to their tendency to aggregate, ultrafine particle number concentrations are more likely to be elevated near sources (*31*). Considering the location of the monitoring stations in Erfurt, the influence of power plants on ultrafine particles in this study is likely small compared to the influence of vehicles. Second, Figures 8 and 10 show that morning NC peaks of all particle sizes in both years occur earlier than the morning SO_2 peak. Thus, particles at this time of day were not likely associated with power plant emissions. Instead, the temporal patterns of the curves coincided well with the curves for NO and CO, suggesting that traffic was associated with the morning NC peaks of all size fractions. Indeed, from the winter × hour interactions, $NC_{0.01–0.03}$ in 1991 was significantly lower in the morning (approximately 0700–1000 hr) compared to the same hours in 1998. This supports the hypothesis of a change in source for $NC_{0.01–0.03}$, especially during the morning hours. This is consistent with motor vehicles as a major source because motor vehicles have undergone a changed hourly pattern over time.

The hourly profile plots also suggest that home heating was associated with the evening trends of larger particles. The $NC_{0.03–0.5}$ and $NC_{0.5–2.5}$ fractions followed the SO_2 level with respect to timing in the afternoon. In 1998, there were no prominent afternoon peaks for these larger fractions or for SO_2, which could be expected if home heating with coal were much reduced in this year. Therefore, SO_2 in 1991 was found to be associated with large particle levels in the evening, which we believe to be due to residential coal burning. For both 1991 and 1998, all morning particle peaks were associated with NO and CO. Because winter × hour interactions indicated the hours of 0700–1000 hr as being significantly different for $NC_{0.01–0.03}$ between the two years, we suggest that changes in motor vehicle emissions between 1991 and 1998 (composition of vehicle fleet, driving patterns, emission controls) have contributed to the increase in $NC_{0.01–0.03}$.

Table 3. Temporal changes in vehicle fleets.

Region (reference) Vehicle category	Engine type	No. of vehicles (% of total) 1992	1995	1998	1998/1992 Ratio
Thuringia[a]					
Cars	Total	751,673	1,165,813	1,204,304	1.60
	Gas	699,467	1,047,013	1,083,216	1.55
	Diesel	51,716 (7)[b]	83,695 (7)	96,278 (8)	1.86
Buses + trucks	Total	53,555	84,215	145,957	2.72
	Gas	9,222	12,901	20,176	2.19
	Diesel	44,236 (83)	80,786 (96)	125,315 (86)	2.83
All categories	Total	805,228	1,250,028	1,350,261	1.67
	Gas	708,689	1,059,914	1,103,392	1.56
	Diesel	95,952 (11)	164,481 (13)	221,593 (16)	2.31
Erfurt[c]					
Cars		80,553[d]	95,738	92,588	1.15[e]
Motorcycles		17,644[d]	2,163	2,958	0.17[e]
Trucks		2,381[d]	8,166	8,763	3.6[e]
All categories		104,012[d]	108,299	106,528	1.02[e]

[a]Data from Kraftfahrt-Bundesamt (*30*). [b]Percentages for Thuringia data refer to percentage of vehicle category composed of vehicles with diesel engines. [c]Data from Statistisches Jahrbuch Theuringen 1996/97 (*45*). [d]Number for 1990. [e]Ratio for 1998/1990.

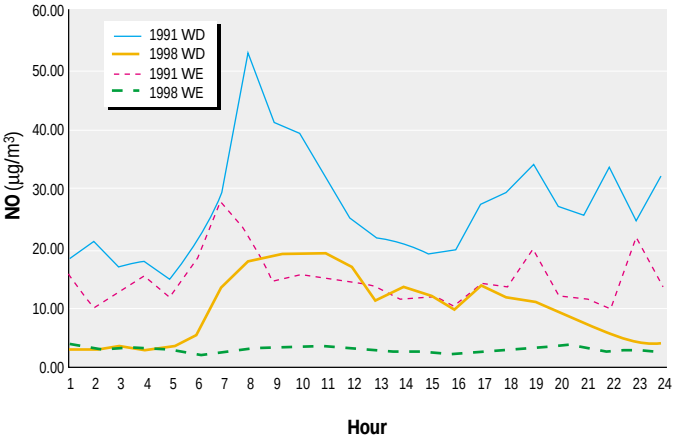

Figure 6. NO hourly profiles for 1991 and 1998, showing weekends (WE) and weekdays (WD) separately.

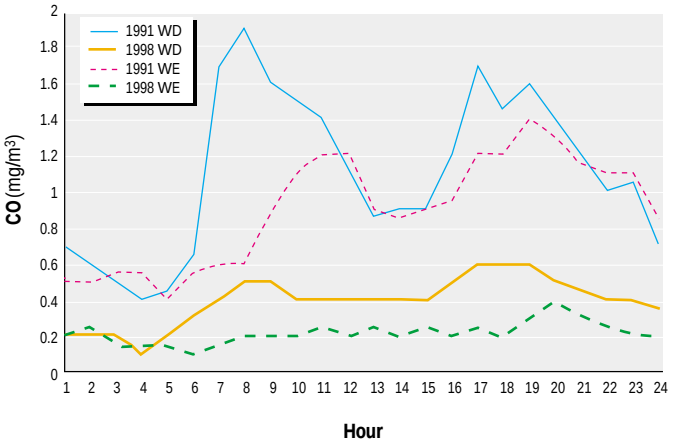

Figure 7. CO hourly profiles for 1991 and 1998, showing weekends (WE) and weekdays (WD) separately.

Discussion

Time–series plots revealed considerable seasonal variability of all pollutants in Erfurt, East Germany, with high concentrations during the winter months (October–March) compared to the summer months (April–September). Such seasonal patterns were expected considering the tendency for winter inversions in the Erfurt region and the use of coal and other fuels for heating during the winter. Due to less complete combustion under colder climatic conditions, automotive emissions are usually also increased during colder months of the year. These data also demonstrate the drastic change in ambient air quality that has occurred in Erfurt after the 1990 unification of East and West Germany.

As direct methods of establishing and quantifying the sources contributing to particulate matter, source apportionment techniques such as chemical mass balance or factor analysis methods can be used. Lacking a complete emissions inventory or analysis of particle constituents, we were not able to perform a direct source analysis at this time. However, by using markers of sources for which we had data and analyzing how these markers varied over the course of the day in 1991 compared to 1998, our goal was to assess to the extent to which changes in specific sources have affected various particle size classes. Regressions between NC and SO_2 and NO indicate major influences of power plants, residential coal combustion, and road traffic on the number concentrations of particles in Erfurt. A limitation in our analysis was the comparison of only two seasons of data, although these where chosen to be representative of a documented trend of changing air pollutant concentrations over the 1990–1999 period.

Before unification, lignite-fired power plants in former East Germany operated with minimum emission controls (*21*). Coal consumption data demonstrated that significant power plant retrofit programs and changes in residential heating worked to sharply decrease SO_2 concentrations in Erfurt. Decreases in the use of coal were reflected in decreased levels of ambient SO_2 and $PM_{2.5}$. Comparing to the current U.S. Environmental Protection Agency's (U.S. EPA) National Ambient Air Quality Standards (NAAQS), $PM_{2.5}$ levels were in excess of the 65 µg/m^3 standard 67 times during winter 1991 (October–March), compared to only 6 times during winter 1998. Similarly, the 24-hr NAAQS for SO_2 of 365 µg/m^3 was exceeded 6 times during winter 1991 (October–March) and was not exceeded at all after 1995.

Our first hypothesis for the observed increases in the smallest ultrafine particles considered decreased ultrafine particle scavenging due to fewer $NC_{0.03–0.5}$ and $NC_{0.5–2.5}$ particles in the ambient air. Due to their high diffusivity, particles in the $NC_{0.01–0.03}$ range are subject to enhanced aggregation particularly with larger particles because of their larger cross-section. The change of the ratio of $NC_{0.01–0.03}$ particles to $PM_{2.5}$ (TSP) concentrations by a factor of 8 (*5*), respectively, can be taken as a measure of such particle scavenging. Increases in $NC_{0.01–0.03}$ per mass of TSP over time

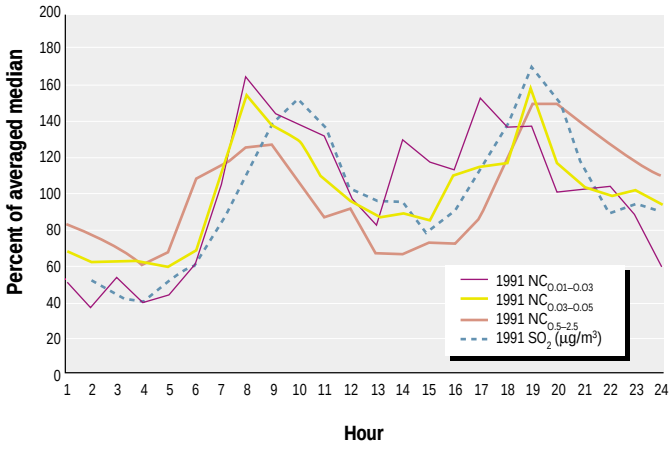

Figure 8. Standardized hourly profiles for particle number concentration and SO_2 in 1991 (weekdays only).

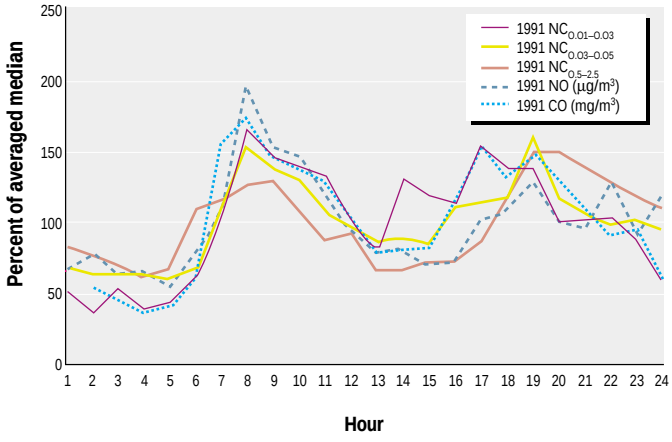

Figure 9. Standardized hourly profiles for particle number concentration, NO, and CO in 1991 (weekdays only).

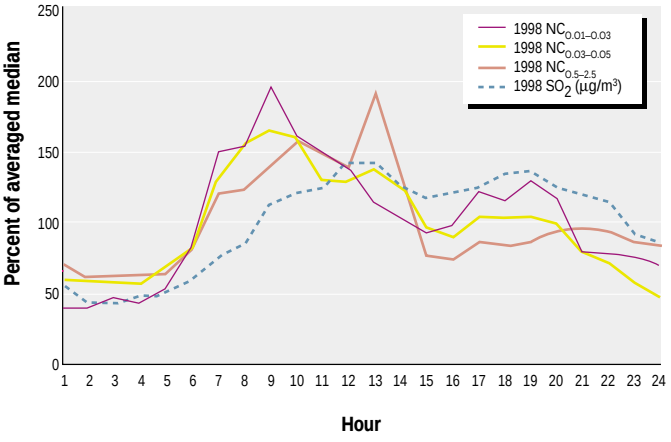

Figure 10. Standardized hourly profiles for particle number concentration and SO_2 in 1998 (weekdays only).

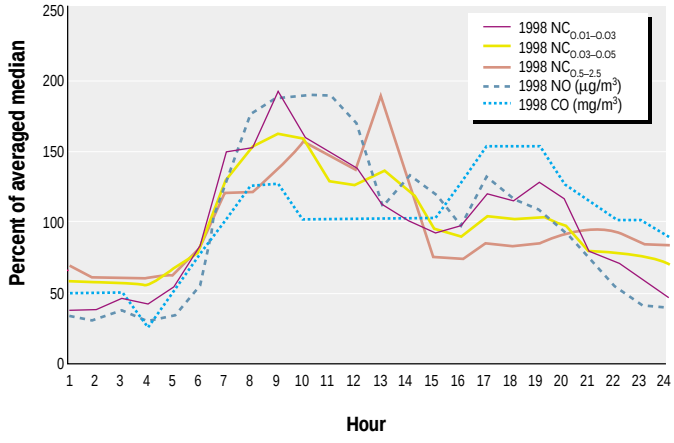

Figure 11. Standardized hourly profiles for particle number concentration, NO, and CO in 1998 (weekdays only).

suggests that decreased particle scavenging is a possible mechanism for retaining high numbers of small particles in the air. Our analysis suggested that residential coal burning was associated with afternoon peaks of larger particles in 1991. This relationship faded over time due to decreases in coal consumption and consequent decreases in the $NC_{0.03–0.5}$ and $NC_{0.5–2.5}$ fractions. If decreases in coal use have affected the concentrations of large particles, then changes in this emission source have potentially contributed to decreased scavenging of smaller particles.

The second hypothesis regarding the increase in $NC_{0.01–0.03}$ considered traffic as the cause, and we focus the discussion of this hypothesis in the following paragraphs. Vehicle emissions were found to be associated with the morning peaks of all particle size fractions. However, significant differences in the morning $NC_{0.01–0.03}$ in particular, between 1991 and 1998, suggested that vehicle emissions in 1998 were directly producing larger numbers of $NC_{0.01–0.03}$. This result is supported by a study conducted in the United Kingdom, which also found that ultrafine particles dominate the number count and that number count data give a clear indication of recent road traffic emissions (*32*).

Supplemental data indicated that the total number of vehicles did not change in Erfurt between 1991 and 1998; however, the composition of the vehicle fleet did change. Although there are no available data, the proportion of two-stroke cars in the Erfurt vehicle fleet has decreased dramatically since 1991. Additionally, trucks increased in number by a factor greater than 3, and motorcycles decreased by a factor greater than 5. The percentage of vehicles with diesel motors increased from 11% to 16% between 1992 and 1998, doubling in number. These changes are likely a combination of increased diesel usage in all of Germany and Western Europe as well as the societal and structural changes that have taken place as a result of unification.

Diesel emissions are a significant air pollution issue. Estimates from the U.S. EPA indicate that diesel vehicles contribute 27% of on-road NO_x and > 60% of on-road PM emissions (*33*). Kirchstetter et al. (*34*) found heavy-duty diesel trucks to be much higher emitters of NO_x, $PM_{2.5}$, SO_4^{2-}, and black carbon than light-duty vehicles. Additionally, heavy-duty diesel engines were found to emit 15–20 times more fine particles than light-duty vehicles per unit mass of fuel burned (*34*). There is also evidence suggesting diesel engines produce particles of smaller sizes. When assessing particles from all mobile sources, mass distributions peak between 0.1 and 0.2 μm particle diameter (*35*). Of two medium-duty diesel vehicles tested, Kleeman et al. (*35*) found particle number size distributions peaking at 30 nm and 50 nm particle diameter. Several studies have demonstrated new diesel vehicles as higher emitters of ultrafine particles in comparison to diesel engines with older technology (*32*,*36*). For example, Bagley et al. (*36*) found that a 1998 diesel engine delivered 15–35 times the number of particles than a 1981 engine produced, although the total mass was reduced. This was due to a 30- to 60-fold increase in the number of smaller primary particles (*36*).

As stated in our second hypothesis, increases in the number of trucks and other diesel-powered vehicles may be contributing

Table 4. General linear models predicting NC fractions (column headings indicate the dependent variables).

Independent variable	$NC_{0.01–0.03}$		$NC_{0.03–0.5}$		$NC_{0.5–2.5}$		Interaction	$NC_{0.01–0.03}$		$NC_{0.03–0.5}$		$NC_{0.5–2.5}$	
	t	*p*	*t*	*p*	*t*	*p*		*t*	*p*	*t*	*p*	*t*	*p*
Intercept	7.0654	0.0000	6.5667	0.0000	2.3433	0.0192	[Winter = 1991] × [hr = 2]	–0.1878	0.8510	0.2027	0.8394	0.2192	0.8265
[WINTER = 1991]	–2.7877	0.0053	–0.1602	0.8727	0.4035	0.6866	[Winter = 1991] × [hr = 3]	–0.1821	0.8555	–0.2520	0.8011	0.0289	0.9769
[WINTER = 1998]							[Winter = 1991] × [hr = 4]	–0.0954	0.9240	–0.6444	0.5194	–0.0706	0.9437
[WE_WD = 0]	–13.6873	0.0000	–7.9740	0.0000	–4.8550	0.0000	[Winter = 1991] × [hr = 5]	–0.4506	0.6523	–1.1000	0.2714	0.3192	0.7496
[WE_WD = 1]							[Winter = 1991] × [hr = 6]	–1.7233	0.0849	–1.6520	0.0986	0.6005	0.5482
[hr = 1]	–0.5876	0.5569	–0.8024	0.4224	–0.1992	0.8422	[Winter = 1991] × [hr = 7]	–3.6535	0.0003	–2.6818	0.0074	0.5908	0.5547
[hr = 2]	–0.4005	0.6888	–0.9770	0.3286	–0.1990	0.8422	[Winter = 1991] × [hr = 8]	–2.5480	0.0109	–1.8715	0.0614	0.3387	0.7349
[hr = 3]	–0.0677	0.9460	–1.0935	0.2743	–0.1581	0.8744	[Winter = 1991] × [hr = 9]	–3.5805	0.0003	–1.5650	0.1177	–0.6504	0.5154
[hr = 4]	–0.4670	0.6405	–1.0499	0.2939	–0.1826	0.8551	[Winter = 1991] × [hr = 10]	–2.2048	0.0275	–1.0131	0.3111	–0.5403	0.5890
[hr = 5]	0.5961	0.5512	–0.6479	0.5171	–0.4868	0.6264	[Winter = 1991] × [hr = 11]	–1.5440	0.1227	–0.8481	0.3964	–1.7663	0.0774
[hr = 6]	3.2035	0.0014	0.4958	0.6201	–0.2572	0.7970	[Winter = 1991] × [hr = 12]	–1.2945	0.1956	0.6457	0.5185	–0.5659	0.5715
[hr = 7]	6.8155	0.0000	2.4687	0.0136	–0.2490	0.8034	[Winter = 1991] × [hr = 13]	–0.5716	0.5676	–0.3380	0.7354	–1.2032	0.2290
[hr = 8]	5.9466	0.0000	3.8648	0.0001	–0.3009	0.7635	[Winter = 1991] × [hr = 14]	–0.2602	0.7947	–0.3428	0.7317	–0.6994	0.4844
[hr = 9]	7.5553	0.0000	3.4140	0.0006	–0.5550	0.5789	[Winter = 1991] × [hr = 15]	–0.2225	0.8239	0.9604	0.3369	0.0107	0.9915
[hr = 10]	5.4919	0.0000	3.1855	0.0015	–0.5865	0.5576	[Winter = 1991] × [hr = 16]	–0.0235	0.9812	0.5460	0.5851	–0.2223	0.8241
[hr = 11]	4.9918	0.0000	3.1058	0.0019	–0.3745	0.7081	[Winter = 1991] × [hr = 17]	–0.3983	0.6904	0.5648	0.5722	–0.8899	0.3736
[hr = 12]	5.0369	0.0000	2.3489	0.0189	–0.5219	0.6018	[Winter = 1991] × [hr = 18]	–0.0210	0.9832	1.1870	0.2353	–1.6704	0.0949
[hr = 13]	4.0586	0.0001	2.7164	0.0066	–0.0555	0.9558	[Winter = 1991] × [hr = 19]	–0.7550	0.4503	1.5163	0.1295	–1.6625	0.0965
[hr = 14]	3.8022	0.0001	2.5658	0.0103	–0.3450	0.7301	[Winter = 1991] × [hr = 20]	–0.8757	0.3812	0.8772	0.3805	–0.6627	0.5076
[hr = 15]	3.9226	0.0001	1.2383	0.2157	–0.8150	0.4152	[Winter = 1991] × [hr = 21]	0.0966	0.9230	0.5564	0.5780	–0.7743	0.4388
[hr = 16]	3.5315	0.0004	1.2033	0.2289	–1.0870	0.2771	[Winter = 1991] × [hr = 22]	–0.0650	0.9482	0.1081	0.9140	–0.6406	0.5218
[hr = 17]	3.4745	0.0005	1.1518	0.2495	–0.9125	0.3616	[Winter = 1991] × [hr = 23]	0.7958	0.4262	0.6871	0.4921	0.0073	0.9942
[hr = 18]	3.0559	0.0023	0.8676	0.3857	–0.8886	0.3743	[Winter = 1991] × [hr = 24]						
[hr = 19]	3.6616	0.0003	0.9204	0.3574	–1.2229	0.2215	[Winter = 1998] × [hr = 1–24]						
[hr = 20]	2.9145	0.0036	0.9693	0.3325	–1.3642	0.1726	[Winter = 1991] × [WE_WD = 0]	4.7869	0.0000	2.0255	0.0429	1.5249	0.1274
[hr = 21]	0.9281	0.3534	–0.0476	0.9620	–1.4565	0.1454	[Winter = 1991] × [WE_WD = 1]						
[hr = 22]	0.7690	0.4420	0.2630	0.7926	–1.1736	0.2406	[Winter = 1998] × [WE_WD = 0]						
[hr = 23]	–0.5833	0.5597	–0.4859	0.6271	–0.8348	0.4039	[Winter = 1998] × [WE_WD = 1]						
[hr = 24]							[Winter = 1991] × SO_2	–9.6593	0.0000	–7.4273	0.0000	–10.1383	0.0000
							[Winter = 1998] × SO_2						
SO_2	9.1031	0.0000	9.6561	0.0000	15.5278	0.0000	[Winter = 1991] × NO	–10.9911	0.0000	1.2034	0.2289	18.1812	0.0000
							[Winter = 1998] × NO						
NO	34.7273	0.0000	57.7616	0.0000	7.3894	0.0000	[WE_WD = 0] × SO_2	–1.3678	0.1715	–1.2127	0.2253	1.1589	0.2466
							[WE_WD = 1] × SO_2						
							[WE_WD = 0] × NO	1.6588	0.0973	3.0926	0.0020	1.6952	0.0901
							[WE_WD = 1] × NO						

Abbreviations: WE_WD = 0, weekend; WE_WD = 1, weekday. Independent variables, indicated in the rows, were regressed against each of the particle size fractions indicated in the columns. For the two periods, winter 1998 is compared to winter 1991 as the reference value. For day of the week, weekdays are compared to weekends as the reference value.

directly to increased ultrafine particle concentrations. Also, changes in engine technology leading to combustion that is more complete could be a cause for increasing numbers of ultrafine particles. In a review of diesel emissions studies, Yanowitz et al. (*37*) reported that emissions of CO and PM have fallen steadily in the last 10 years which is consistent with the findings discussed above (*35*,*36*). Engine technology that decreases PM increases the efficiency of combustion, thereby also lowering CO and total hydrocarbon emissions. However, no changes in average NO_x emissions of diesel vehicles have been observed (*37*,*38*). NO_x and PM emissions are inversely correlated, which is the main barrier to lowering diesel emissions (*37*).

In Erfurt, NO concentrations did not increase along with the increase in the number of trucks. In fact, NO concentrations decreased by > 50% between 1991 and 1998. This was likely due to changes in engine technology that took place during the 1990s. For example, introduction of the three-way catalysts on gasoline-powered, light-duty vehicles has been mandatory on all new cars sold within the European Union since January 1993. A trend analysis between 1986 and 1994 for 15 Swedish cities confirmed that the real-world efficiency of the three-way catalyst corresponds to a reduction in NO_x emissions from the average vehicle by at least 80–90% (*39*). Because the majority of NO_x from automobiles is in the form of NO, NO from these vehicles has decreased over time. Therefore, the observed 50% decrease in NO, although less than expected from the introduction of three-way catalyst-equipped, light-duty vehicles, may be explained by the increased number of trucks with comparatively high NO emissions.

Overall, decreases in the concentrations of larger particle fractions were largely associated with the decreases in coal combustion that occurred during the 1990s. Changes in vehicle composition and emissions were reflected in a changed hourly pattern of $NC_{0.01–0.03}$ and were thought to be the cause of increased number concentrations of these ultrafine particles. However, the exact contribution of vehicle and fuel types was not analyzed, and emission rates have been found to vary significantly with vehicle classification and driving conditions, among other parameters (*35*,*37*,*40*). There have been suggestions on how to distinguish between gasoline and diesel vehicles in source apportionment studies, for example, by using polycyclic aromatic hydrocarbon profiles created to represent the local fleet of vehicles (*41*).

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