

Czech Air Quality Monitoring and Receptor Modeling Study

J. P. PINTO* AND R. K. STEVENS†

*U.S. Environmental Protection Agency,
Research Triangle Park, North Carolina 27711*

R. D. WILLIS AND R. KELLOGG

*ManTech Environmental Technology, Inc.,
Research Triangle Park, North Carolina 27711*

Y. MAMANE

Technion-Israel Institute of Technology, Haifa, Israel

J. NOVAK AND J. ŠANTROCH

Czech Hydrometeorological Institute, Prague, Czech Republic

I. BENEŠ

District Institute of Hygiene, Teplice, Czech Republic

J. LENIČEK

Regional Hygiene Station, Usti nad Labem, Czech Republic

V. BUREŠ

*Technical Services for Air Pollution Control,
Prague, Czech Republic*

An ongoing air quality monitoring program in the Czech Republic has provided nearly continuous data for the concentrations of aerosol and gas-phase pollutants since its inception in February 1992. In addition to PM-2.5 concentrations, the concentrations of sulfate, organic carbon, elemental carbon, trace elements (Al–Pb), and polynuclear aromatic hydrocarbons (PAHs) were also measured. Fine particulate matter (PM-2.5) was composed mainly of organic carbon and sulfate with smaller amounts of trace metals. Coarse particle mass concentrations were typically between 10 and 30% of PM-2.5 concentrations. The chemical composition of emissions from power plants, residential space heating, local factories, and motor vehicles was also characterized. The ambient monitoring and source characterization data were then used in receptor modeling calculations, the results of which indicate that residential space heating and power plant emissions accounted for most of fine particle mass concentrations observed during winter air pollution episodes. Motor vehicles, incinerators, and windblown dust contributed to the balance of the fine particle mass. Peak 24-h average TSP and SO₂ concentrations (1100 and 800 µg/m³, respectively) obtained at the main monitoring site at Teplice in northern Bohemia during a severe air pollution episode in 1993 were within a factor of 2 of smoke and SO₂ concentrations (1800 and 1600 µg/m³) measured in London during the smog episode of December 5–9, 1952. That pollution episode was thought to have contributed to a substantial increase in mortality.

* Corresponding author fax: 919-541-1818; e-mail: pinto.joseph@epamail.epa.gov.

† Present address: Department of Environmental Protection, State of Florida.

Introduction

Burning coal, especially high-sulfur lignite, without effective pollution controls in the Czech Republic (CR) has resulted in high atmospheric concentrations of sulfur dioxide and particles since 1945 (1). During the 1980s, coal burning resulted in annual SO₂ and TSP (total suspended particulate) concentrations exceeding 100 µg/m³ in parts of the northwest CR and nearly that in the city of Prague (1). These elevated pollution levels are thought to contribute to health problems in the Czech Republic. Life expectancy in the Czech Republic has been estimated to be 3–6 years shorter than the European average (1).

Since January 1992, the Czech Ministry of Environment and the Czech Institute of Hygiene, in collaboration with the U.S. Environmental Protection Agency (EPA), have conducted an air quality field measurement program in the northwestern Czech Republic. The principal objectives of this program were to assess human exposure to toxic air pollutants, to relate ambient concentrations of pollutants to health risks (3, 4), and to determine the major pollution sources so that air pollution control measures could be implemented most effectively.

Aerosol and gas-phase air pollutant measurements were made at two sites during this study. The main monitoring site was located in Teplice (1990 population of 60 000), an industrial city in northern Bohemia, 90 km northwest of Prague and located in the same valley mentioned above. The second monitoring site was located in Prachatic (1990 population of 15 000), a town 130 km south of Prague. Aerosol composition data were collected in Prachatic only in 1993, while data reported here were collected from 1992 to 1994 in Teplice. Figure 1 shows the location of the major power plants and sampling sites in the CR. Six of the largest power plants in the CR are located in the northwestern CR, in a broad valley extending from the southwest to the northeast and surrounded by the Ore Mountains and the Middle Bohemian Mountains (2). In addition, many residents in the region use high-sulfur lignite coal to fire their stoves for heating during the winter.

Teplice was selected for this study because its residents could be exposed to high air pollution levels caused by emissions from local industry, power plants, lignite mines, residential space heating, and motor vehicles under adverse meteorological conditions. Pollutant emissions are often trapped within the valleys of the northwestern CR during periodic wintertime inversions. Prachatic was selected as a control location for health studies because of its location and low levels of targeted pollutants. Aerosol samples were analyzed for sulfate, crustal elements (e.g., Al, Si), toxic metals (Cr, Cu, Ni, Zn, As, Se, Pb), organic carbon (OC), elemental carbon (EC), and volatile and semivolatile PAHs. The composition of particles emitted by power plants, home heating stoves, glass factories, hospital incinerators, and motor vehicles were characterized for use in receptor modeling calculations to determine their relative importance as air pollution sources.

In this paper, we will focus on the results of ambient aerosol and SO₂ measurements obtained from February 1992 through February 1994 and the results of receptor modeling calculations used to estimate the relative contributions of the major air pollution sources to aerosol samples collected at the Teplice monitoring site. Concentrations of CO, NO_x, O₃, HNO₃, HCl, and HF were also measured in ambient air at Teplice. Further details regarding the complete set of aerosol and gas-phase pollutant measurements, including

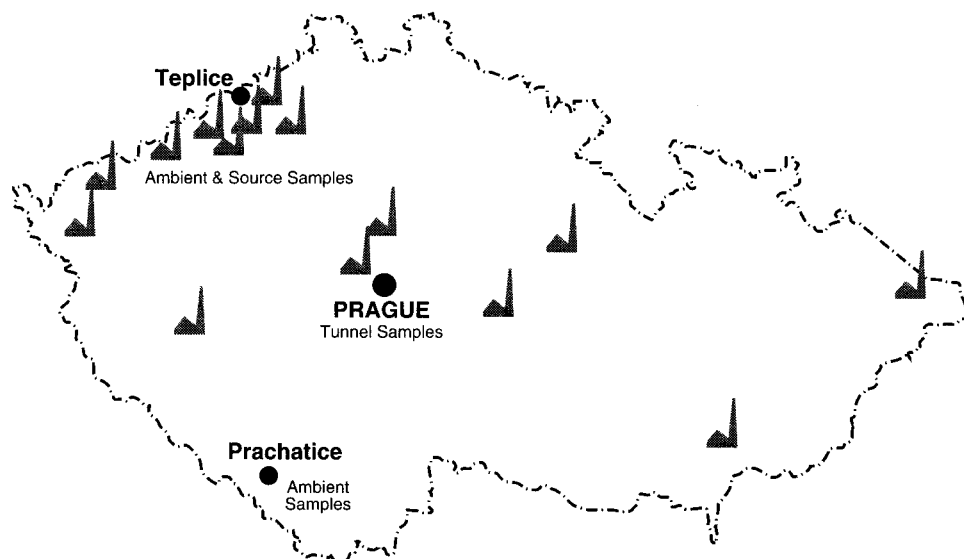


FIGURE 1. Map of the Czech Republic. The locations of the main monitoring sites (Teplice and Prachatic) along with the major power plants and source sampling sites are shown.

results for indoor-outdoor sampling can be found in Stevens et al. (4). Methods used in the collection of ambient data and source composition profiles are first described in the paper. Ambient concentrations of a number of key pollutants are then presented. Results of receptor modeling results, aided by scanning electron microscopy (SEM), for determining the relative contributions of major air pollution sources at the monitoring site in Teplice follow. Inferences about the mechanisms for the oxidation of SO_2 to sulfate in the airshed are discussed next. A comparison of SO_2 and total suspended particulate matter (TSP) concentrations obtained in Teplice during a severe air pollution episode in February 1993 with those obtained in London during the smog episode of December 1952 is also presented.

Experimental Methods

Ambient Air Sampling and Analysis. The versatile air pollution sampler (VAPS) (5) was used to collect ambient aerosol and acidic gas samples. The VAPS system (Figure 2) is a modified dichotomous sampler consisting of an inlet (with d_{50} cut point of $8 \mu\text{m}$ aerodynamic diameter, d_a) and a virtual impactor, which separates the airflow into two channels that collect fine particles ($<2.5 \mu\text{m}$ d_a), and a third channel that collects coarse particles ($2.5-8 \mu\text{m}$). Inorganic constituents were measured in the first fine-particle channel, and organic constituents were measured in the second fine-particle channel. An annular denuder (6) and a 47-mm Teflon filter pack, located downstream of the denuder, were used on the first fine-particle channel. The denuder was coated with a solution of sodium carbonate to remove acidic gases (SO_2 , HCl , HNO_3 , HF) from the sample airstream. The denuder was then extracted with deionized water after sampling, and the extracts were analyzed by ion chromatography (IC). Fine-particle mass (FPM) was determined gravimetrically, elemental composition was determined by X-ray fluorescence (XRF), and inorganic anions were determined by IC from particle deposits on the Teflon filter. On the second fine-particle channel, a 47-mm quartz filter was used to collect organic carbon (OC), elemental carbon (EC), and semivolatile PAHs. Concentrations of OC and EC were determined using thermal-optical analysis techniques (7). A polyurethane foam (PUF) trap was used downstream of the quartz filter to collect PAHs that evaporate from the quartz filter or were originally in the gas phase. PAH concentrations were obtained by extraction of the PUF trap and the quartz

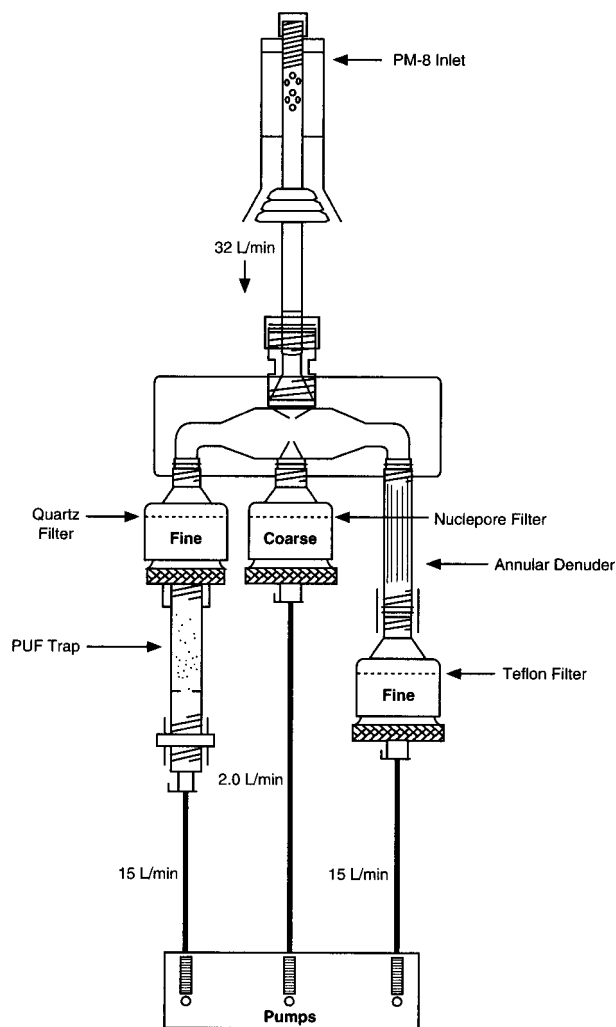


FIGURE 2. Schematic diagram of the versatile air pollution sampler (VAPS) used in the study. The VAPS is a modified dichotomous sampler that collects two fine particle samples and a coarse particle sample.

filters followed by high-performance liquid chromatography analysis (8). A Nuclepore filter pack was used on the third

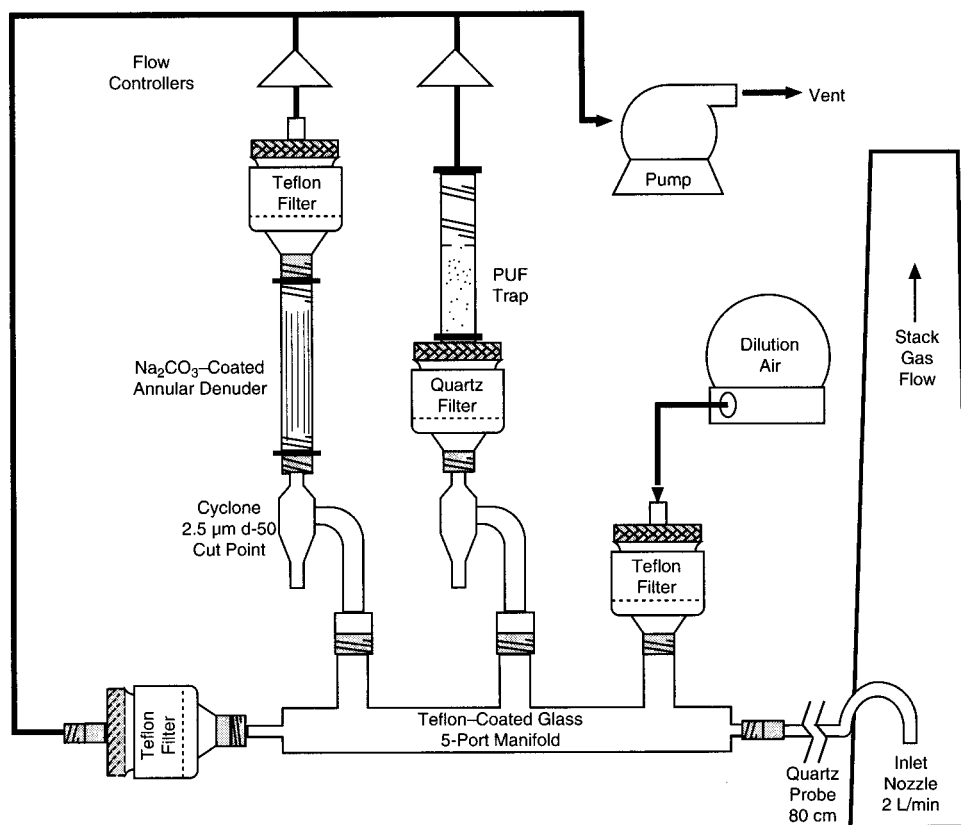


FIGURE 3. Schematic diagram of dilution source probe (DSP) used to collect stack samples.

TABLE 1. Mean Aerosol Composition in Teplice, Winter and Summer 1992

species	February–March		May–July	
SO ₂ (µg/m ³)	135 (20) ^a		31.1 (4.7)	
species	February–March		fine (May–July)	coarse (May only)
	fine	coarse		
Particulate Matter (µg/m ³)				
total mass	68.0 (1.9)	26.5 (2.7)	36.5 (1.2)	9.0 (1.5)
organic carbon	30.3 (5.0)	na ^b	na	na
elemental carbon	3.3 (0.5)	na	na	na
metal oxides	4.4 (0.3)	7.3 (0.8)	1.5 (0.11)	5.9 (0.8)
sulfate	14.7 (1.8)	2.0 ^c (0.5)	12.8 (1.6)	1.0 (0.4)
sum of PAHs	0.058 (0.002)	na	na	na
benzo[a]pyrene	6 × 10 ⁻⁴ (2 × 10 ⁻⁵)	na	na	na
no. of samples	43	32	35	21
Trace Elements (ng/m ³)				
Al	270 ^d (101)	930 (220)	110 ^d (50)	700 (212)
Si	570 (93)	1800 (320)	260 (44)	1300 (330)
S	3570 (250)	490 ^c (120)	3100 (230)	250 ^c (97)
Cl	700 (51)	99 ^c (28)	130 (13)	27 ^c (6.6)
K	220 (16)	130 (13.5)	110 (8.9)	110 (12)
Ca	120 (11)	490 (27)	90 (7.4)	360 (27)
Ti	28 ^d (8.3)	76 (10.5)	15 (4.5)	57 (9.0)
V	5.7 ^d (2.7)	–0.5 ^c (1.9)	2.5 ^d (1.5)	1.0 ^c (1.0)
Cr	3.5 ^d (1.3)	1.5 ^c (1.1)	1.3 ^d (0.7)	1.5 ^c (0.6)
Mn	12 (1.8)	13 (1.5)	8.2 (1.1)	8.9 (1.0)
Fe	260 (24)	630 (45)	150 (14)	440 (44)
Ni	2.1 ^d (1.3)	2.5 ^c (0.9)	0.8 ^d (0.8)	0.4 ^c (0.4)
Cu	12 (1.9)	6.7 (1.2)	17 (2.0)	12 (1.7)
Zn	110 (11)	17 (4.0)	69 (7.0)	15 (2.7)
As	23 (3.2)	2.9 ^c (1.2)	3.7 ^d (0.6)	1.5 ^c (0.6)
Se	5.0 (1.0)	0.5 ^c (0.5)	1.9 ^d (0.3)	0.2 ^c (0.2)
Br	19 (2.2)	8.7 (1.2)	9.8 (1.2)	5.6 (0.7)
Pb	95 (9.4)	9.9 ^c (3.1)	57 (6.1)	8.0 (1.5)

^a Estimated analytical uncertainty shown in parentheses. ^b na, measurements not available. ^c Detected at the analytical uncertainty in fewer than half the samples. ^d Detected at 3 times the analytical uncertainty in fewer than half the samples.

TABLE 2. Mean Aerosol Composition in Teplice, Winter and Summer 1993

species	January–March		May–August	
SO ₂ (μg/m ³)	153 (23) ^a		na ^b	
species	January–March		May–August	
	fine	coarse	fine	coarse
Particulate Matter (μg/m ³)				
total mass	122 (3.0)	18.5 (4.9)	28.7 (1.2)	10.6 (1.1)
organic carbon	33.8 (5.0)	na	na	na
elemental carbon	2.3 (0.3)	na	na	na
metal oxides	6.5 (0.5)	14.0 (2.0)	1.9 (0.14)	7.3 (1.0)
sulfate	41.3 (5.7)	1.5 ^c (0.8)	10.2 (1.3)	0.3 ^c (0.3)
sum of PAHs	0.278 (0.012)	na	0.027 (0.001)	na
benzo[<i>a</i>]pyrene	8 × 10 ⁻³ (4 × 10 ⁻⁴)	na	5 × 10 ⁻⁴ (4 × 10 ⁻⁴)	na
no. of samples	66	62	37	36
Trace Elements (ng/m ³)				
Al	510 (130)	1900 (544)	160 (47)	790 (210)
Si	930 (180)	3100 (770)	290 (43)	1650 (400)
S	10000 (920)	370 ^c (190)	2460 (190)	69 ^c (64)
Cl	410 (27)	100 (18.3)	23 (4.3)	58 (6.2)
K	300 (24)	210 (23)	94 (7.6)	125 (7.1)
Ca	140 (12)	600 (45)	75 (6.3)	450 (33)
Ti	51 (7.4)	140 (22)	17 (4.4)	72 (11.0)
V	7.7 (1.8)	−0.2 ^c (1.8)	1.4 ^c (1.5)	1.8 ^c (1.1)
Cr	4.7 (1.0)	−0.3 ^c (0.7)	1.8 ^d (0.7)	0.2 ^c (0.6)
Mn	17 (1.7)	12 (1.5)	8.0 (1.0)	11 (1.1)
Fe	390 (42)	840 (84)	150 (14)	490 (48)
Ni	3.6 ^d (1.1)	0.8 ^c (0.7)	0.4 ^c (0.9)	0.8 ^c (0.9)
Cu	12 (1.7)	7.8 (1.2)	3.1 ^d (1.0)	4.0 (0.7)
Zn	160 (16)	20 (4.0)	34 (3.6)	9.8 (1.6)
As	34 (4.5)	3.6 ^c (0.9)	2.2 ^c (1.5)	1.0 ^c (0.6)
Se	6.7 (1.0)	0.3 ^c (0.3)	1.2 ^c (0.6)	0.3 ^c (0.3)
Br	18 (2.1)	3.5 (0.7)	4.7 (0.9)	1.8 (0.3)
Pb	110 (11)	6.5 (2.1)	39 (4.4)	3.8 ^c (1.3)

^a Estimated analytical uncertainty shown in parentheses. ^b na, measurements not available. ^c Detected at the analytical uncertainty in fewer than half the samples. ^d Detected at 3 times the analytical uncertainty in fewer than half the samples.

channel to collect coarse-particle samples suitable for SEM analyses. Coarse-particle mass (CPM, defined here as particle mass between 2.5 and 8 μm) was obtained by gravimetric analysis of deposits on the Nuclepore filter after correcting for the presence of fine particles. The VAPS system was installed in a container that also houses Czech Hydrometeorological Institute (CHMI) instruments for measuring CO, O₃, NO_x, SO₂, and TSP concentrations. The container is located near the main square in the center of Teplice. VAPS samples were collected for 24-h periods except during episodes of high aerosol concentrations, when the sampling period was shortened to 12 h. Samples were collected daily during periods when elevated levels were expected. The sampling frequency decreased to once every sixth day during the summer.

Manual scanning electron microscopy (SEM) coupled with energy-dispersive X-ray fluorescence analysis (EDX) was performed on selected aerosol samples collected in Teplice and on airborne samples collected in the plumes of industrial and power plant sources in northwestern Bohemia. Prior to manual SEM analysis, nondestructive XRF analysis of the bulk sample was conducted. The results of XRF analyses were used to screen filters for SEM analysis. SEM/EDX analysis provided information on the size, morphology, and chemistry of individual particles. Approximately 100 particles were characterized for each selected sample, and the results were tabulated by size and chemical class.

Source Sampling. Samples to characterize emissions from various sources for use in receptor modeling were obtained from the Ledvice, Tušimice, Počerady, and Pruneřov power plants in the northwestern CR; the Řetenice glass factory and a hospital incinerator in Teplice; chimneys of

home heating furnaces in Teplice; and motor vehicle traffic. Samples from the major point sources were collected with a dilution source probe (DSP), shown in Figure 3. The apparatus used is similar in design to those used in the past (9). Inorganic pollutants (SO₂, metals, and SO₄²⁻) and organic pollutants (elemental and organic carbon, PAHs) were collected with this sampling system. Air from the stack was first drawn into an isokinetic sampling probe and was then diluted with ambient air, in a ratio of 10:1, in a Teflon-coated, 0.4-L quartz sampling manifold. The dilution airstream was passed through an absorbent to remove water vapor and through Teflon and quartz filters to remove ambient particles prior to mixing of stack gases with ambient air in the sampling manifold. Separate sampling trains were used to collect inorganic and organic pollutant samples. The first sampling train consisted of a cyclone inlet (*d*₅₀ cut point of 2.5 μm) followed by an annular denuder to collect SO₂ and then by a Teflon filter pack to collect fine particles. Carbonaceous species were collected on the second sampling train consisting of a cyclone (*d*₅₀ cut point of 2.5 μm) followed by a quartz filter and a PUF trap.

Two principal concerns regarding the use of the DSP were condensation of stack water and condensation of metal vapors in the sampling manifold. To minimize condensation of water and hence sampling losses, the ratio of desiccated ambient air to stack air was adjusted to maintain a relative humidity of less than 80% in the sampling manifold. The sampling manifold was carefully monitored throughout the sampling period for the presence of liquid water. No condensation of water vapor was seen on the surfaces of the sampling manifold during these studies. The second concern relates to the equilibration of metal vapors with particles in

TABLE 3. Mean Aerosol Composition in Teplice, Winter 1994

species	November 1993–February 1994	
SO ₂ (μg/m ³)	na	
species	November 1993–February 1994	
	fine	coarse
Particulate Matter (μg/m ³)		
total mass	51.1 (2.8) ^a	14.4 (1.1)
organic carbon	na ^b	na
elemental carbon	na	na
metal oxides	3.0 (0.2)	8.8 (1.2)
sulfate	15.3 (1.9)	0.7 ^c (0.4)
sum of PAHs	0.15 (0.006)	na
benzo[a]pyrene	5.5 × 10 ⁻³ (3 × 10 ⁻³)	na
no. of samples	117	112
Trace Elements (ng/m ³)		
Al	240 (58)	1100 (330)
Si	430 (66)	1950 (490)
S	3700 (305)	175 ^c (98.7)
Cl	120 (12)	53 ^c (9.2)
K	250 (21)	130 (17)
Ca	87 (7.7)	400 (35)
Ti	27 (4.7)	97 (14.6)
V	6.8 (1.6)	2.9 ^c (1.2)
Cr	2.2 ^d (0.7)	1.5 ^c (0.6)
Mn	11 (1.3)	9.2 (1.2)
Fe	200 (21)	550 (60)
Ni	2.3 (0.9)	0.6 ^c (0.5)
Cu	6.6 (1.2)	6.1 (1.0)
Zn	100 (10)	11 (2.7)
As	13 (2.4)	1.8 ^c (0.8)
Se	3.5 (0.7)	0.3 ^c (0.3)
Br	9.7 (1.3)	3.0 (0.6)
Pb	85 (9.0)	6.6 (1.9)

^a Estimated analytical uncertainty shown in parentheses. ^b na, measurements not available. ^c Detected at the analytical uncertainty in fewer than half the samples. ^d Detected at 3 times the analytical uncertainty in fewer than half the samples.

the sampling manifold. For efficient condensation of supersaturated metal vapors onto existing particles, the time constant for collisions with particles should be shorter than the residence time of air in the sampling manifold. The residence time of air in the sampling manifold is about 0.5 s, which is roughly the same time scale for collisions of metal vapors with particles in the sampling manifold for conditions found in the stack of the Ledvice power plant. This condition may not be achieved in the stacks of power plants that have more efficient pollution control devices than those used in the Ledvice power plant, resulting in the lack of equilibration of some metals (e.g., As and Se).

Emissions from the chimney of a hand-fired coal stove were sampled using two fine particle (<2.5 μm aero. diameter) sampling systems placed inside a 2 m × 2 m × 2 m sampling chamber placed over the chimney. One of the sampling systems consisted of a cyclone inlet ($d_{50} = 2.5 \mu\text{m}$) followed by annular denuders coated with Na₂CO₃ (for collection of SO₂) and a Teflon filter pack (for collection of fine particles); the other consisted of a cyclone inlet ($d_{50} = 2.5 \mu\text{m}$) followed by a quartz filter pack (for collection of OC and EC and semivolatile PAHs) and a PUF trap (for collection of volatilized PAHs). Sampling was conducted during smoldering low-temperature conditions and active high-temperature conditions in the stove. Particulate emissions from motor vehicles were sampled using a VAPS system placed in the Letensky Tunnel in Prague. A wide variety of conditions were encountered in the tunnel, ranging from stop-and-go to steady traffic flow. The sampler was placed about 30 m from one end of the tunnel. Light-duty gasoline vehicles accounted

for about 90% of the traffic in the tunnel during sampling, and diesels accounted for the remainder.

Ambient Sampling Results

Composition of the Aerosol in Teplice and Prachatic. The mass and elemental composition of aerosol samples collected in Teplice during 1992, 1993, and 1994 are shown in Tables 1, 2, and 3, respectively. The tables show the average composition of aerosols measured for winter, spring, and summer sampling periods. Organic carbon concentrations were multiplied by 1.4 to account for the presence of oxidized species, trace elements were expressed as their stable oxides, and sulfates were expressed as (NH₄)₂SO₄ in the tables. Nitrates were found to be present in extremely low concentrations in the subset of samples analyzed by IC. A variable fraction (roughly one-third) of the FPM could not be accounted for by the above species. We conclude that the missing mass is water of hydration associated with the sulfates. Coarse-particle mass typically ranged from 10 to 30% of the fine-particle mass.

Mean FPM concentrations for the winter of 1993 (122 μg/m³) were significantly higher than for the winters of 1992 (68 μg/m³) and 1994 (50 μg/m³). Part of these differences can be attributed to two air pollution episodes that occurred during the winter of 1993. During the first episode, lasting from January 29 to February 6, 1993, the peak 24-h average FPM concentration was 803 μg/m³, and the average FPM concentration for this episode was 574 μg/m³. Peak 24-h average concentrations of SO₄²⁻ and organic carbon during this episode were 400 and 140 μg/m³, respectively. pH measurements were made on two filter samples collected on February 3 and 4, 1993. A pH of 3.7 was found for the first sample, and a pH of 4.1 was found for the second sample. An extraction volume of 10 mL was used. These pH levels indicate the presence of acidic aerosols, consistent with NH₄-HSO₄. The second episode was less severe than the first and lasted from February 8 to February 14, 1993. The peak 24-h FPM concentration was 204 μg/m³, and the average FPM concentration was 127 μg/m³ during this episode. Peak 24-h mean concentrations of trace elements were also very high during these episodes (e.g., V, 33; Ni, 18; Cu, 47; Zn, 616; As, 275; Se, 34; and Pb, 456 ng/m³). These concentrations are all much larger than those typically found at urban sites in the United States (10).

The difference between the average FPM concentrations observed during the first episode of January 29–February 5 (574 μg/m³) and during the second episode of February 8–14 (127 μg/m³) was due in large part to changes in meteorological conditions. During the first episode, there was a very strong thermal inversion extending from the valley floor to an elevation of about 1500 m above it. As a result, vertical and horizontal mixing was severely inhibited, allowing the trapping of emissions close to the surface and close to their sources. Daytime temperatures were as low as -10 °C, the atmosphere was foggy (visibility was < 200 m), and local wind speeds were very low (<0.5 m/s). On February 7, a cold front had broken up this inversion allowing for some ventilation of pollutants. However, the next day another inversion formed aloft. The base of this inversion layer varied from 500 to 900 m above sea level (roughly 250–650 m above the valley floor), and the inversion extended upward to about 1250 m above the valley floor. Visibility was better, and wind speeds were higher at the surface. Thus, the extent of mixing within the valleys was probably greater than during the first episode. However, emissions were still likely to be trapped between the mountains, thus leading to the high pollutant levels that were observed.

Both of these air stagnation episodes affected the whole Czech Republic with high pollutant levels observed elsewhere.

TABLE 4. Mean Aerosol Composition in Prachatice, Winter and Summer 1993

species	January–March		May–August	
SO ₂ (μg/m ³)	29.0 (4.4) ^a		4.4 (0.7)	
species	January–March		May–August	
	fine	coarse	fine	coarse
Particulate Matter (μg/m ³)				
total mass	44.0 (0.8)	8.0 (0.3)	17.9 (0.4)	5.7 (0.2)
organic carbon	na ^b	na	na	na
elemental carbon	na	na	na	na
metal oxides	1.8 (0.09)	4.3 (0.52)	1.1 (0.07)	2.8 (0.35)
sulfate	9.5 (1.2)	1.0 (0.13)	6.7 (0.9)	0.5 ^c (0.2)
sum of PAHs	0.163 (0.008)	na	0.024 (0.001)	na
benzo[a]pyrene	4.7 × 10 ⁻³ (2.4 × 10 ⁻⁴)	na	1.4 × 10 ⁻⁴ (1 × 10 ⁻⁵)	na
no. of samples	32	28	29	27
Trace Elements (ng/m ³)				
Al	120 ^d (33)	510 (137)	82 (20)	260 (75)
Si	290 (36)	860 (210)	165 (24)	630 (150)
S	2460 (160)	250 (70)	1630 (110)	110 ^c (47.3)
Cl	200 (14)	7.8 ^c (8.3)	20 (2.2)	6.5 (1.4)
K	240 (14)	84 (11.0)	87 (5.9)	82 (8.5)
Ca	76 (4.9)	280 (18)	48 (3.5)	165 (12)
Ti	18 (2.7)	58 (7.6)	8.8 (1.6)	25 (3.7)
V	4.2 (0.9)	0.5 ^c (0.8)	1.3 (0.5)	0.5 ^c (0.4)
Cr	0.8 ^c (0.4)	–0.6 ^c (0.3)	0.6 (0.2)	0.5 ^c (0.2)
Mn	5.4 (0.6)	5.4 (0.6)	3.4 (0.4)	3.8 (0.4)
Fe	120 (9.5)	240 (22)	83 (7.5)	170 (16)
Ni	1.7 ^d (0.8)	0.3 ^c (0.5)	2.2 ^d (0.6)	–0.1 ^c (0.4)
Cu	17 (1.6)	5.3 (0.8)	21 (2.0)	6.7 (1.0)
Zn	81 (6.2)	11 (1.8)	23 (2.1)	5.7 (0.9)
As	30 (2.6)	4.2 (0.7)	1.9 (0.6)	0.5 ^c (0.2)
Se	2.1 (0.4)	0.1 ^c (0.1)	0.5 ^c (0.2)	<0.1 ^c (<0.1)
Br	11 (1.1)	2.7 (0.4)	4.2 (0.5)	1.6 (0.2)
Pb	54 (4.6)	6.3 (1.1)	23 (2.3)	3.1 (0.6)

^a Estimated analytical uncertainty shown in parentheses. ^b na, measurements not available. ^c Detected at the analytical uncertainty in fewer than half the samples. ^d Detected at 3 times the analytical uncertainty in fewer than half the samples.

For instance, mean FPM concentrations measured in Prachatice were 106 and 76 μg/m³ during the first and second episodes of February 1993. The mean aerosol composition measured in Prachatice during the winter of 1993 is shown in Table 4. Average FPM concentrations measured in Prachatice during the winter of 1993 (44 μg/m³) were approximately one-third those measured in Teplice during the same time period. The high levels of As and some other metals observed in Teplice and Prachatice may result from coal burning as it is the only major source that is common to both towns.

As can be seen from Tables 1–3, FPM concentrations were substantially lower during spring and summer sampling periods than during winter sampling periods in Teplice. These differences reflect the absence of home heating emissions and enhanced ventilation of the planetary boundary layer during the warmer seasons. Mean FPM concentrations observed in Prachatice during the summer of 1993 (18 μg/m³) were higher than those measured in relatively clean continental air (11), but were still lower than those measured in Teplice during the warmer seasons (36 μg/m³ in 1992; 29 μg/m³ in 1993).

Continuous Monitoring Data. Time series of TSP concentrations measured by the β-gauge technique and SO₂ concentrations measured by pulsed fluorescence in Teplice during 1993 are shown in Figure 4, panels a and b. The 30-min concentrations reported by the continuous monitors were averaged over 24 h. TSP concentrations obtained by the β-gauge continuous monitor and PM-2.5 concentrations measured by gravimetric analysis of Teflon filters collected by the VAPS were highly correlated ($r = 0.92$). β-gauge data (y) were related to VAPS data (x) (in μg/m³) by the expression,

$y = (1.15 \pm 0.33)x + (14.7 \pm 69.5)$. As expected, the β-gauge readings were generally higher than the VAPS data because they also include the mass of coarse particles larger than 2.5 μm in diameter. However, there is a rather large spread between the two data sets, which may partly be the effect of high humidity on the β-gauge measurements. Breakthrough of SO₂ frequently occurred in annular denuder measurements made during the air pollution episodes, as the denuder collection sites became saturated. As a result, SO₂ concentrations measured by IC analysis of denuder extracts tended to underestimate ambient SO₂ levels during these periods (4).

Polynuclear Aromatic Hydrocarbon Measurements. Samples collected during 1993 and the winter of 1994 at the Teplice and Prachatice monitoring sites were analyzed for nine PAHs as shown in Table 5. The sampling method (i.e., a quartz filter followed by a PUF trap) is expected to quantitatively collect the heavier PAHs—benz[a]anthracene, benzo[k]fluoranthene, benzo[a]pyrene (B[a]P), indeno[1,2,3-cd]pyrene, and benzo[ghi]perylene—while the lighter PAHs (the so-called semivolatile PAHs) would not be collected quantitatively. Total PAHs (sum of quartz filter and PUF trap) were several times higher during the winter than during the summer in both Teplice and Prachatice. Total PAHs measured during the winters of 1993 and 1994 in Teplice averaged about 60% higher than those measured in Prachatice over the same time periods. PAH concentrations were relatively low and were about equal at both sites during the summer of 1993.

The time series of the ratio of B[a]P to Pb in Teplice during 1993 is shown in Figure 5. During the winter, the ratio of B[a]P to Pb ranges from 0.05 to 0.15, and during the summer

TABLE 5. Average Polynuclear Aromatic Hydrocarbons Measured in Teplice and Prachatice, January 1993–March 1994 (ng/m³)

species	January–March 1993		May–August 1993		November–March 1994	
	Teplice	Prachatice	Teplice	Prachatice	Teplice	Prachatice ^a
phenanthrene	61.1	36.6	10.7	11.7	47.4	30.8
anthracene	80.1	75.3	5.6	4.2	7.4 ^b	nr ^c
fluoranthene	38.6	25.1	3.4	4.0	27.7	18.3
pyrene	37.9	21.6	2.5	2.6	24.6	15.3
benz[a]anthracene	11.6	8.7	0.5	0.4	7.1	5.8
benzo[k]fluoranthene	7.2	5.7	0.3	0.1	4.2	3.6
benzo[a]pyrene	8.0	4.7	0.5	0.1	5.5	3.4
indeno[1,2,3-cd]pyrene	7.7	5.9	0.5	0.4	6.6	4.5
benzo[ghi]perylene	5.4	3.5	0.4	0.3	4.4	2.4
chrysene	nr	nr	nr	nr	6.6 ^b	nr
benzo[b]fluoranthene	nr	nr	nr	nr	6.7 ^b	nr
dibenz[a,h]anthracene	nr	nr	nr	nr	1.4 ^d	nr
sum	278	187	27	24	149 ^e	84 ^e

^a Data available only through 1/8/94. ^b Average of 2/4/94 to 3/17/94. ^c nr, not reported. ^d Average of 1/1/94 to 2/3/94. ^e Some data are missing.

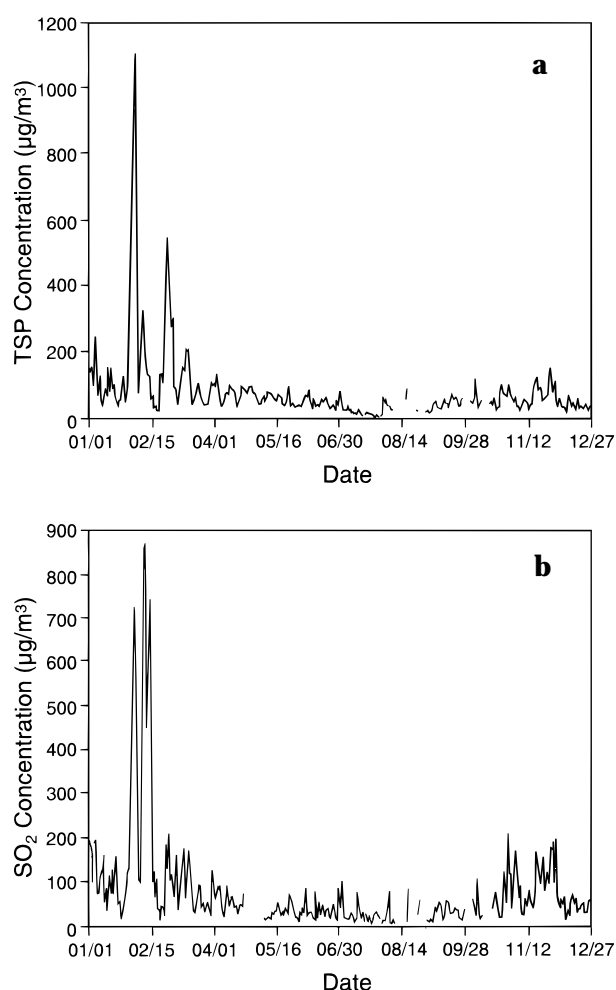


FIGURE 4. (a) Time series of TSP concentrations measured by β -gauge in Teplice in 1993. (b) Time series of SO₂ concentrations measured by pulsed fluorescence in Teplice in 1993.

the ratio of B[a]P to Pb is about 0.01. The seasonal variation in the ratio of B[a]P to Pb indicates the presence of at least two major sources of PAHs, if we assume that the ratio of B[a]P to Pb in motor vehicle emissions is similar in winter and summer. Emissions from inefficient combustion of lignite in home heating systems are the most likely source of the increase in B[a]P and PAH concentrations during the winter in Teplice. Wood burning may be an additional source

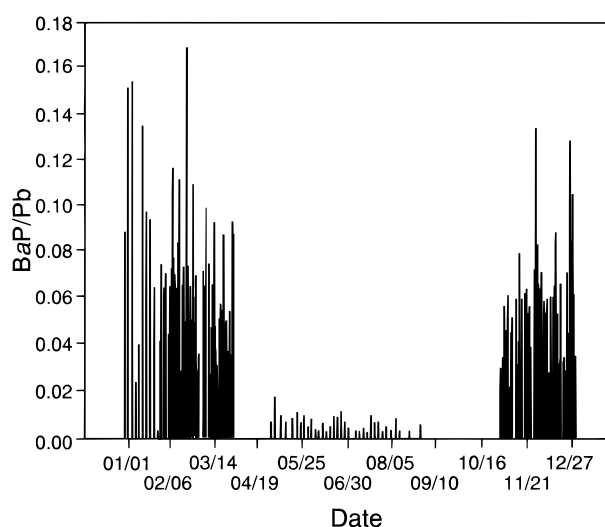


FIGURE 5. Time series of the ratio of benzo[a]pyrene (B[a]P) to Pb in Teplice in 1993.

of PAHs in Prachatice, as evidenced by the higher ratio of K to FPM in winter samples collected in Prachatice as compared to those obtained in Teplice (12).

Scanning Electron Microscopy Results. A subset of ambient (winter 1992, 1993) and source samples (home heating, power plant, traffic tunnel) collected for the Czech study were analyzed manually by SEM/EDX. The SEM/EDX results supported the bulk XRF analyses of the samples. During winter temperature inversions, however, the ambient filters were often too heavily loaded for quantitative SEM analysis. Major elements detected in individual coarse particles included Al, Si, K, Ca, Ti, and Fe. These elements are typically associated with soil minerals. During inversion episodes, unusually high fractions of the coarse mass were associated with aluminosilicate fly ash particles from lignite or coal-fired boilers.

SEM photomicrographs of coarse and fine fraction particles collected on February 8, 1993, in Teplice are shown in Figure 6, panels a and b. An extremely high concentration of fly ash particles was observed in the coarse fraction sample. Most of the remaining particles were aluminosilicate minerals or gypsum. Some of the gypsum may have formed by the reaction of calcite with anthropogenic sulfate. Some organic particles were also observed in the coarse fraction, but no bioaerosols (fungal spores, pollen) were identified. Fine particles observed by SEM (Figure 6b) were dominated by

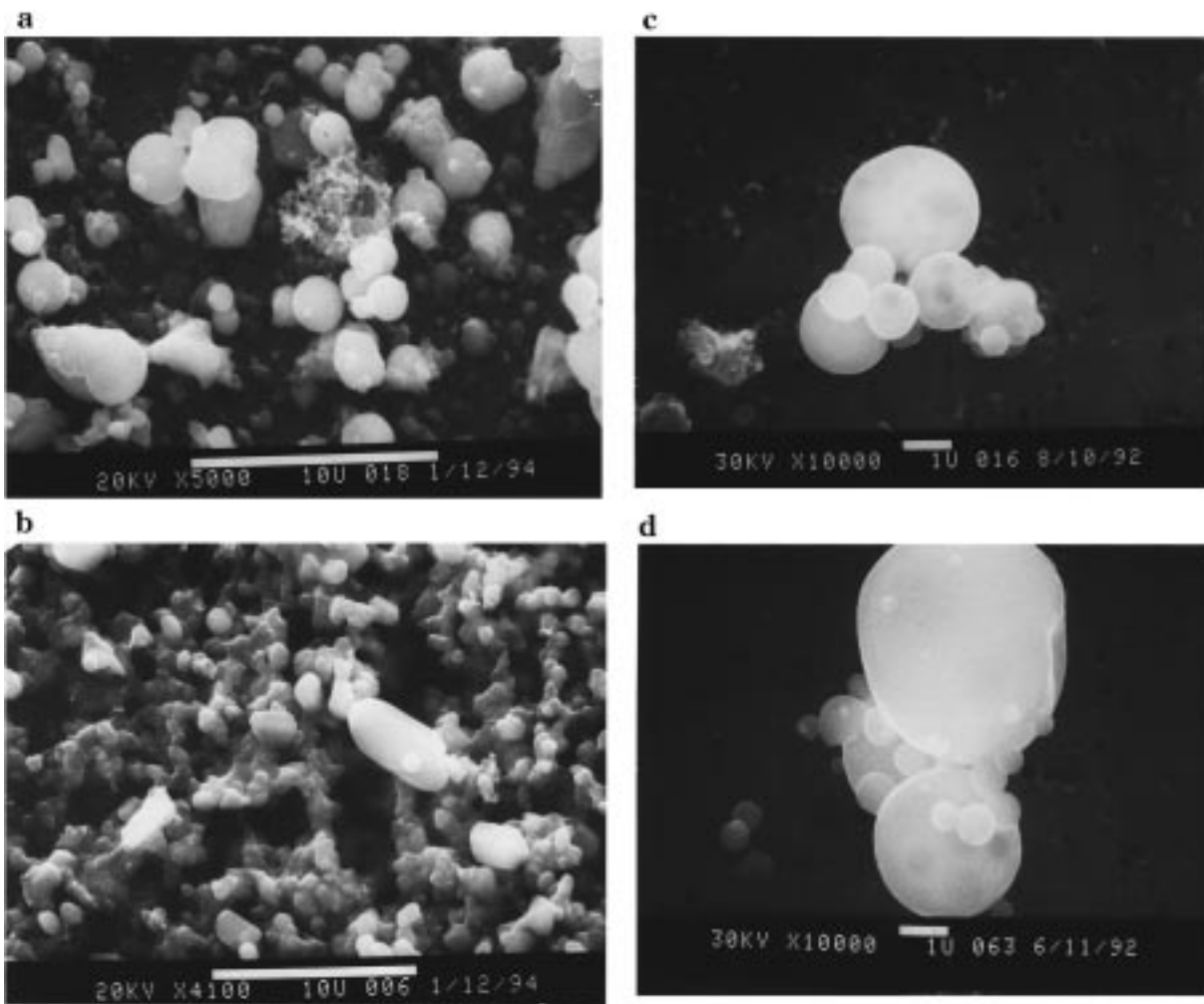


FIGURE 6. (a) SEM photomicrographs of coarse particles collected during February 1993 air pollution episode. (b) Same as panel a for fine particles. (c) SEM photomicrograph of fly ash particles collected in the plume of Ledvice power plant. (d) SEM photomicrograph of ambient air sample collected in Teplice on March 2, 1992.

sulfates and carbonaceous particles. Because of the very high loading, particles piled up on the filter rendered the sample unsuitable for counting and sizing. Fly ash particles were also observed in large number. SEM analysis of other samples collected during the winter of 1992 and the pollution episodes in February 1993 revealed similar results: the fine fraction was dominated mainly by sulfates and carbonaceous particles (80–90% by particle count) with the remainder composed largely of spherical fly ash.

Figure 6, panels c and d are close-up photomicrographs of fly ash aggregates collected in the Ledvice power plant plume by aircraft and in ambient air in Teplice (sample collected on March 2, 1992; 133 of the 290 coarse particles analyzed were fly ash). The similarity between the two photomicrographs suggests that power plants were the source of these particles in the sample. Not all the fly ash particles were spheres or cenospheres, as are found in coal-fired power plant emissions (13). Some Teplice fly ash particles appeared rounded but not strictly spherical and were bonded with organic matter to form aggregates. The latter may result from combustion at lower temperatures, which would produce more condensable organic particles and nonspherical fly ash. Nonspherical fly ash particles rich in organic material were found in the home heating emissions samples. Additional sources contributing to the ambient samples were indicated by the presence of coal particles rich in pyrite, iron spheres, aluminum chips, oil fly ash, and silicon-rich spheres.

Modeling Results

Receptor Modeling. Estimates of the contributions of emissions from power plants, incinerators, automobiles, and home heating to primary PM-2.5 concentrations measured at the main monitoring site in Teplice were made using the chemical mass balance (CMB) receptor model (14). The source composition profiles used in the receptor modeling calculations are listed in Table 6. Sulfate only represents a few percent, at most, of the fine-particle emissions from all the sources considered in this study, while it constitutes a much larger fraction of the mass of the ambient samples. Therefore, most of the sulfate seen in the study was of secondary origin. Unfortunately, the CMB method is not suitable for apportioning sources of secondary sulfate.

Lignite-fueled power plants and home heating in the northwestern CR are probably the dominant sources of SO₂ in the region. Table 7 shows annual emissions of SO₂ and particulate matter from power plants and residential space heating for 1992 (2). As can be seen from Table 7, total annual power plant and home heating emissions of SO₂ in the Teplice region were 574 600 and 8400 T, respectively, for 1992 (2). Because we are mainly concerned with wintertime values, the power plant emissions are scaled down by a factor of 4, to about 144 000 T. The SO₂ emissions from home heating are used as reported because most home heating occurs during the winter. Thus 94% of the SO₂ emissions in the

TABLE 6. Source Profiles Used in Receptor Modeling Calculations^a

species	Ledvice power plant	home heating—S	home heating—A	mobile	hospital incinerator
OC	7.5	68	62	54	56
EC		2.0	6.2	10	18
Al	16				0.77
Si	20	0.082		0.29	1.7
S	1.4	0.83	1.5	2.7	0.5
Cl		0.12	0.21	0.38	4.3
K	1.8	0.067	0.26	0.10	2.2
Ca	0.43	0.20		0.10	0.87
Ti	0.98	0.030			0.21
V	0.034				
Cr	0.029				
Mn	0.019			0.021	0.097
Fe	4.2	0.084	0.044	0.39	0.55
Ni	0.010			0.0073	
Cu	0.012			0.021	
Zn	0.067	0.015	0.19	0.095	0.98
As	0.022	0.0062	0.023		
Se	0.056	0.0025	0.0027		
Br		0.0067	0.0059	0.26	0.028
Pb	0.019	0.0060	0.021	0.91	0.11

^a All values given as percent of total mass. Abbreviations: home heating—S, smoldering phase; home heating—A, active (high-temperature) phase; OC, organic carbon; EC, elemental carbon.

TABLE 7. Emissions of Particulate Matter and SO₂ from Major Point Sources and Residential Space Heating in the Northwest Czech Republic

source	SO ₂ (ton/year)	particulate matter (ton/year)	stack height (m)	distance from Teplice (km)
power plant				
Ledvice	38200	11600	200	3
Tuřimice I	48200	3856	195	46
Tuřimice II	88200	2732	300	46
Pruneřov I	59700	5788	200	46
Pruneřov II	163100	3984	300	46
Litvinov	55344	23055	100	15
(Chemopetrol Z.) Počerady	110700	9448	200	29
residential space heating (home heating)				
District Usti n.L.	2378	1952		1
District Teplice	2220	1828		1
District Most	1494	1228		19
District Chomutov	2306	1890		35

region during the winter are due to power plants and 6% are due to residential space heating.

Without adequate knowledge of the rate of oxidation of SO₂ to SO₄²⁻, only the sum of ambient SO₂ and SO₄²⁻ or total S (TS) could be apportioned to its sources. The power plant (*S_{PP}*) and home heating (*S_{HH}*) contributions to total S were scaled to receptor modeling results for nonsulfate or primary FPM (*NSP_{HH}*, *NSP_{PP}*, i.e., trace metals and organic carbon) by using the following relation

$$(S_{HH})/(S_{PP}) = (NSP_{HH} \times X_{HH})/(NSP_{PP} \times X_{PP}) \quad (1)$$

subject to the constraint

$$(S_{HH}) + (S_{PP}) = TS \quad (2)$$

where *X* is the ratio of SO₂ emissions to fine particle mass emissions and the subscripts HH and PP refer to home heating and power plants, respectively.

TABLE 8. Relative Contributions of Different Source Types to Primary FPM Concentrations Predicted by the CMB Method and Estimates of Power Plant and Home Heating Contributions to Total S

	1st episode (%)	2nd episode (%)	winter 1993 av (%)
SO ₄ ²⁻	48 ± 6.0	50 ± 6.2	40 ± 4.8
home heating—S	17.5 ± 4.7	18.8 ± 7.0	24.0 ± 7.4
home heating—A	1.6 ± 0.4	4.3 ± 3.5	<1.0
power plants	1.8 ± 0.3	11.4 ± 0.9	3.3 ± 0.5
mobile	1.6 ± 0.4	2.5 ± 0.6	2.8 ± 0.6
incinerator	3.4 ± 1.1	4.5 ± 1.8	6.9 ± 1.8
unknown	25 ± 17.2	8.5 ± 24	23
χ ²	1.8	1.3	0.5
Total S			
home heating	50	24	40
power plants	50	76	60

Emissions inventories (cf. Table 7) were used to determine the ratios, *X_{HH}* and *X_{PP}*. Emissions-weighted mean values of *X_{HH}* and *X_{PP}* were calculated by multiplying their values in individual sources by SO₂ emissions and by *R₁/R₂*, where *R₁* is the distance of the closest (Ledvice) power plant from Teplice and *R₂* is the distance of the source from Teplice. Simple inverse scaling based on distance was chosen because power plant emissions are channeled between the mountain ranges, which results in a larger contribution from upwind sources than if uniform dispersion in all directions is present (i.e., 1/*R*²). The elevation of the mountain peaks above the valley floor ranges from about 600 m to 1000 m, while the stack heights of the point sources in the valley are at most 300 m (cf. Table 7). Uncertainties for this approach are based on the combined uncertainties for the CMB results and for the ratios, *X_{HH}* and *X_{PP}*. The method given above could be applied to secondary sulfate aerosol, if the assumption that the same fraction of SO₂ emitted by each source is converted to SO₄²⁻ were made.

CMB model results for primary constituents and SO₂ + SO₄²⁻ apportionments for the winter of 1993 are shown in Table 8. Also shown are measures of the CMB model accuracy, given by χ² at the bottom of Table 8. It can be seen that sulfates constitute approximately one-half of the FPM. Home heating emissions, which are mainly composed of organic carbon (cf. Table 6), were found to be the major source of primary FPM. Power plants, motor vehicles, and incinerators were less important sources of primary FPM. A variable percentage of FPM could not be assigned to any specific source and is assumed to be water of hydration associated with sulfates. The contributions of power plants to total S were found to be much larger than to primary FPM. There is a marked increase in the power plant contributions to both primary FPM and to total S from the first episode to the second episode of February 1993, which may be related to the change in meteorological conditions outlined earlier.

Ireson et al. (15) modeled the contribution of major point sources (mainly power plants and glass factories) to SO₂ and particulate matter in northern Bohemia with a Gaussian plume dispersion model (ISC2). They found good agreement between model predictions and observations when they only used emissions from major point sources except during air stagnation episodes. During these periods, they consistently underestimated measured SO₂ concentrations in the region. In addition, they found an inverse relation between surface wind speed and SO₂ concentrations. As a result, they concluded that area sources, such as residential space heating, may be major contributors to ambient concentrations at the Teplice site during air stagnation episodes, even though their emissions are much smaller than those from

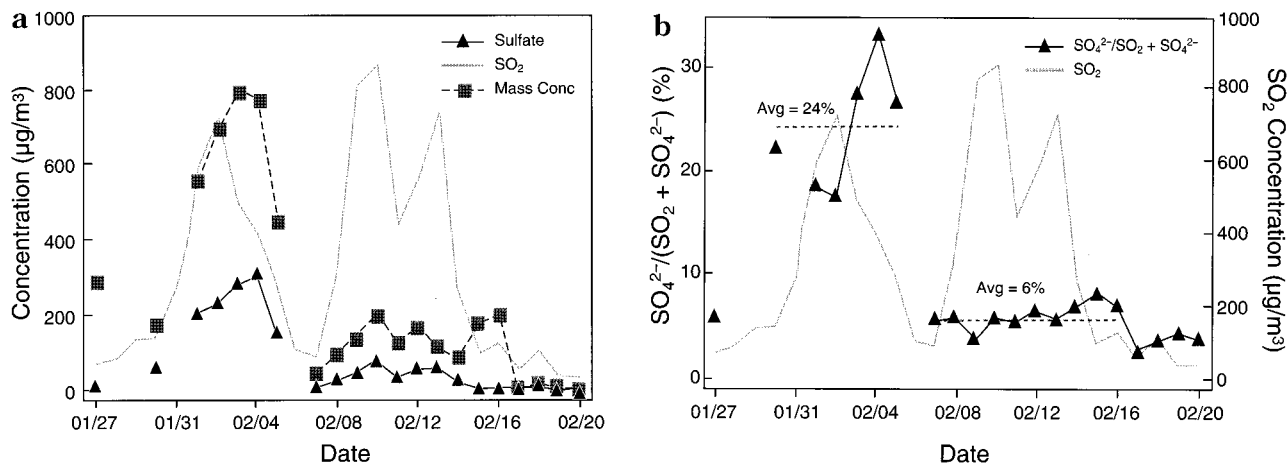


FIGURE 7. (a) Sulfate and sulfur dioxide concentrations in Teplice during February 1993 episodes. (b) $\text{SO}_4^{2-}/(\text{SO}_2 + \text{SO}_4^{2-})$ ratio for data shown in panel a. Average values of this ratio for the two episodes are also shown.

power plants in the region. This finding is consistent with the receptor modeling calculations given above.

Sulfur Dioxide to Sulfate Conversion. Significant features of the two air pollution episodes in February 1993 were the differences between SO_2 and fine-particle mass concentrations, as seen in Figure 7a, and the ratios of $\text{SO}_4^{2-}/(\text{SO}_2 + \text{SO}_4^{2-})$, shown in Figure 7b. The ratio $\text{SO}_4^{2-}/(\text{SO}_2 + \text{SO}_4^{2-})$, provides an indication of the conversion of SO_2 to SO_4^{2-} since air was effectively trapped within the valleys during the air pollution episodes. Although this database is insufficient to calculate the rate of conversion of SO_2 to SO_4^{2-} , it can be used to provide some information on the mechanism of SO_4^{2-} formation. The high solar zenith angles and high O_3 columns during winter at the latitude of northern Bohemia strongly limit the solar UV flux reaching the surface and hence the production rate of OH radicals and subsequent rate of homogeneous oxidation of SO_2 to SO_4^{2-} .

SEM analysis of the fine fraction collected during the 1993 winter episode showed that the fine particles containing sulfur were typically in the size range of $0.5\text{--}1\text{ }\mu\text{m}$ in diameter. IC analysis provided evidence that the fine particle sulfur was in the form of SO_4^{2-} . Fe (16) and Mn (17) in fog droplets have been shown to enhance the conversion of SO_2 to SO_4^{2-} . Meng and Seinfeld (18) theorized that SO_4^{2-} particles formed from heterogeneous aqueous reactions would result in particles from 0.7 to $1.5\text{ }\mu\text{m}$ in diameter, consistent with SEM results from this study. This particle size information and the $\text{SO}_4^{2-}/(\text{SO}_2 + \text{SO}_4^{2-})$ ratios suggest SO_2 to SO_4^{2-} conversion by a heterogeneous mechanism. Results of studies in Ljubljana, Slovenia (19, 20) are also consistent with the heterogeneous oxidation of SO_2 on carbonaceous particles. During the first episode, the concentrations of fine particles, carbon, Fe, and Mn were substantially higher than during the second episode. These factors may help account for the differences in the apparent efficiency of conversion of SO_2 to SO_4^{2-} for the two winter episodes in 1993.

Comparison with the 1952 London Episode. During the January 29–February 5, 1993, air pollution episode in Teplice, the meteorological conditions were characterized by low wind speed ($<0.5\text{ m/s}$), low temperatures (daily max $T < 0\text{ }^\circ\text{C}$), and hazy fog-like conditions (visibility often less than 100 m). These conditions were similar to those in London, December 5–9, 1952 (21). The maximum particulate matter (measured by the blackness of the filter) and SO_2 (measured by the hydrogen peroxide method) 24-h average concentrations in London were 1600 and $1800\text{ }\mu\text{g}/\text{m}^3$ (0.7 ppm), respectively. The sampling and analyses methods used at that time may have suffered from artifacts such as SO_2 reacting

on the filters to form sulfates; therefore, it is probable that the reported levels of SO_2 for London were overestimates of the actual values.

There is a significant difference in topographical setting between Teplice and London. Teplice is located in a valley surrounded by land barriers that favor the accumulation of pollutants. Although London is located in the Thames Valley, no steep land barriers exist, and the maximum pollution was confined to the central part of the Thames Basin. In this regard, Teplice is similar to two other cities that are associated with historic smog episodes: Liege, Belgium, and Donora, PA, both located in valleys. Figure 8 shows the time series of TSP (measured by β -gauge) and SO_2 (measured by pulsed fluorescence) 24-h average concentrations measured in Teplice from January 28 to February 8, 1993, compared to data obtained in London from December 5 to December 9, 1952. In London (population 8.5 million) the 4-day smog episode was responsible for about 4000 excess deaths during the 2-week period from the onset of the pollution episode. At the peak of the London episode, approximately 900 deaths per day occurred as compared to 250 deaths per day before the episode (22). The Teplice episode is similar in character to the London smog episode: Both episodes were characterized by high SO_2 and PM concentrations; in both cities, home heating and power plants were the principal pollution sources; and both occurred during the winter months during strong surface inversions.

In addition, during the winter of 1993, PM-10 and SO_2 concentrations exceeded the maximum 24-h values of the health-based U.S. National Ambient Air Quality Standards (NAAQS) on 20 and 10 occasions, respectively. The 24-h air quality standard for SO_2 is 0.14 ppm , and for particulate matter it is $150\text{ }\mu\text{g}/\text{m}^3$. These exceedances in themselves indicate that there may have been health problems that occurred during these episodes that were associated with exposure to these high concentrations of SO_2 and particulate matter. Therefore, morbidity and mortality records should be examined to relate these exposures to health effects.

Discussion

Results of the study are summarized below.

(a) There were several air stagnation episodes in the valleys of the Ore Mountains during the course of this study. During one of these episodes, 24-h average FPM loadings approached $1\text{ mg}/\text{m}^3$ in Teplice. The major aerosol constituents during these periods were sulfate and organic carbon. Concentrations of V, Cr, Cu, Zn, As, Se, and Pb were much larger than those measured at other times in Teplice and in U.S. cities.

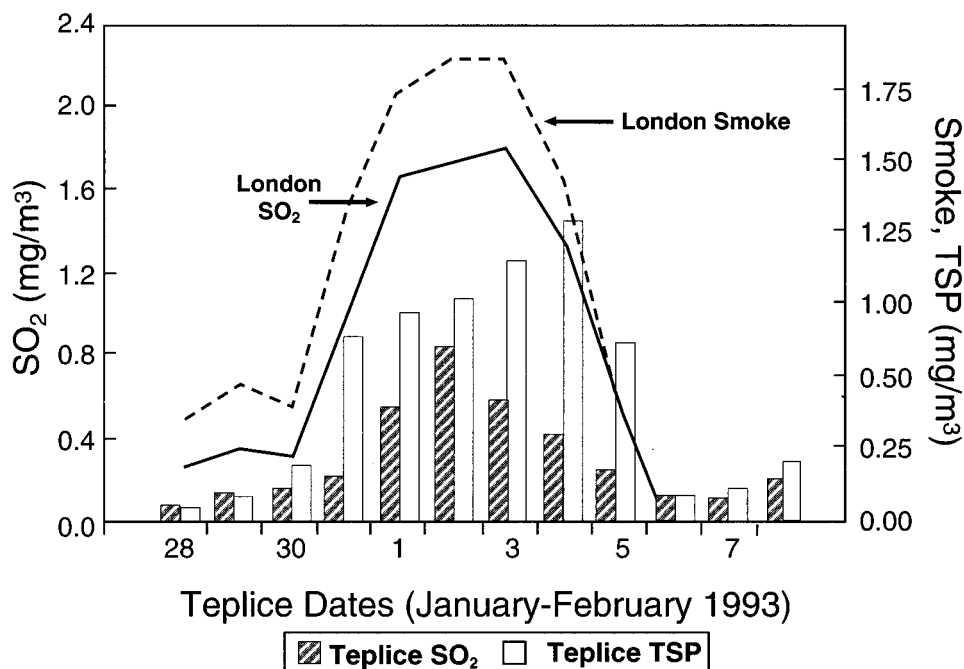


FIGURE 8. TSP and SO₂ concentrations measured during the January 29–February 5, 1993, air pollution episode in Teplice compared with black smoke and SO₂ concentrations measured during the December 5–9, 1952, air pollution episode in London.

(b) Between January 29 and February 6, 1993, concentrations of SO₂ and particulate matter in Teplice approached concentrations measured in the London smog episode of December 5–9, 1952. Both episodes were due to emissions from power plants and home heating systems using high-sulfur coal. During the London smog episode, there were substantial increases in mortality that have been associated with the high pollutant concentrations.

(c) About 85% of the fine particles analyzed by scanning electron microscopy (SEM) were sulfates or carbonaceous particles. The remaining particles were mostly fly ash associated with power plants or home heating. Coarse-particle mass typically ranged from 10 to 30% of the fine mass and was dominated by power plant fly ash and soil particles.

(d) The ratio of benzo[a]pyrene (B[a]P) to Pb was much higher during the winter than during the summer, indicating the presence of additional B[a]P sources such as residential space heating during the winter. Likewise, the higher ratios of As to FPM and PAHs to FPM during the winter compared to summer suggest the importance of home heating as a significant source of these pollutants.

(e) CMB model results indicate that residential space heating was the major source of primary FPM, while power plants, motor vehicles, and incinerators were less important sources. However, power plants were found to be a significant source of SO₂ and SO₄²⁻.

(f) Substantial oxidation of SO₂ to SO₄²⁻ occurred because primary SO₄²⁻ constitutes a much smaller fraction of the FPM in the source profiles than in the ambient data. Atmospheric conditions were not conducive to the homogeneous oxidation of SO₂. Thus, an efficient heterogeneous oxidation mechanism was hypothesized to explain the observed oxidation of SO₂.

(g) The extremely high concentrations of sulfur dioxide and particulate matter (especially fine particles) measured in the Czech Republic in these studies may pose a health risk to people living in the region. Exposure to high concentrations of SO₂ has been associated with respiratory problems (23), and exposure to high concentrations of PM-10 particles has been associated with increased morbidity and mortality (24).

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