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Source contributions to airborne particle deposition at the Yungang Grottoes, China

Lynn G. Salmon^a, Christos S. Christoforou^a, Timothy J. Gerk^a, Glen R. Cass^{* a}, Gary S. Casuccio^b, Gary A. Cooke^b, Michael Leger^b, Ilhan Olmez^c

 ^a Environmental Engineering Science Department and Environmental Quality Laboratory, California Institute of Technology, Pasadena, CA 91125, USA
^bR.J. Lee Group, Inc., 350 Hochberg Road, Monroeville, PA 15146, USA
^cNuclear Reactor Laboratory, Massachusetts Institute of Technology, Cambridge, MA 02139, USA

Abstract

The Buddhist cave temple complex at Yungang in northern China is affected by a rapid accumulation of airborne particles that settle onto the thousands of statues contained within those caves. Experiments have been conducted to identify the most important air pollution sources that contribute to the dust deposition problem. The spatial distribution of the deposition rate of airborne particles within a 2 km \times 2 km area surrounding the grottoes was measured during a 2-day period in April, 1991. Peak particle deposition rates of > 60 μ g m⁻² s⁻¹ were found at locations within the village of Yungang itself and along the adjacent coal-haul highway. Moving away from the village and coal-haul highway, deposition rates decline to much lower values, indicating that the village and highway are significant sources of airborne particles. A comparison of the mineralogical composition of the dust deposits in the caves with the composition of local soil dust, paved road dust from the coal-haul highway and deteriorated cave ceiling rock material indicates that the dust deposits in the caves are a combination of the above sources, with the paved road dust from the coal-haul highway providing the closest match to the largest quantity of the material deposited in the caves.

Keywords: Airborne particles; Emission sources; Dust deposition; Deposition fluxes; Coal-haul highway

1. Introduction

In the 5th century A.D., excavation of the Buddhist cave temples at Yungang began under the patronage of the emperors of the Northern Wei Dynasty. Over a period of ~ 50 years, 20 major caverns and numerous smaller caves were carved out of a sandstone cliff at Yungang near the present industrial city of Datong in northern Shanxi Province, China (Mizuno, 1950; Knauer, 1983; Caswell, 1988). The largest grottoes typically contain a monumental statue of the Buddha, carved from the native sandstone. The walls and ceilings of these caves are covered by thousands

^{*}Corresponding author.

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of carved, and in some cases painted, sculptures that are objects of reverence to the Buddhist faithful.

These grottoes and the village of Yungang at their base are now located in a major coal mining region of northern central China. Dust and smoke are generated by operations at the coal mines and by coal trucks on a highway that runs within a few 100 m of the grottoes. Coal combustion occurs for the purposes of cooking and heating in the village of Yungang, and coal-fired locomotives run on nearby railroad tracks. Fugitive soil dust is generated by traffic on unpaved roads and sidewalks in the village of Yungang and by visitors to the caves, as well as by dust storms from the Gobi desert. As a result of such sources, outdoor airborne coarse particle (diameter > 2.1 μ m) concentrations immediately outside the entrance to the caves at Yungang averaged 378 μ g m⁻³, and outdoor fine particle (diameter $\leq 2.1 \ \mu$ m) concentrations averaged 130 μ g m⁻³ during 1991-1992 (Salmon et al., 1994). Peak 24-h average outdoor particle concentrations exceeded 1200 $\mu g m^{-3}$.

Air containing these high outdoor particle concentrations enters the caves, whereupon particles are deposited onto the sculptures within. The deposits of dust obscure features of the statues and degrade the painted surfaces. Over the period 1986–1991, since the last cleaning, a dust loading of up to 5 kg m⁻² has accumulated to depths of as much as 0.8 cm on the horizontal surfaces of the statues (Christoforou et al., 1994).

A series of experiments and measurements was conducted at Yungang in 1991, in order to examine the character and origin of this dust deposition problem. During these experiments a monitoring network was established to measure the spatial distribution of particle deposition rates over a 2 km \times 2 km geographic area surrounding the grottoes. By mapping the spatial distribution of the particle flux in the countryside surrounding the grottoes, areas of the highest deposition flux can be found that indicate close proximity to a major particle source, thus identifying the source itself. In addition, dust samples were taken from the surfaces of the statues in the caves, and

authentic source samples were taken from likely major contributing particle sources, including local coal dust, soil dust and paved road dust from the large coal-haul highway located near the grottoes. By matching the mineralogical composition of the dust deposits in the caves to the best linear combination of the mineralogical composition of the likely major sources, it is also possible to test hypotheses about the most important sources contributing to the soiling problem in the caves. This information on relationships between local dust sources and the soiling of the statues in the caves can be used to help guide remediation efforts directed at reducing the soiling problem.

2. Materials and methods

2.1. Measurement of the spatial distribution of coarse particle deposition at Yungang

The map shown in Fig. 1 depicts a $2km \times 2km$ square area surrounding the Yungang Grottoes. The cave temples are carved into a cliff located near the center of the map with the entrances to the caves facing south. Immediately in front of the caves is a walled park area that serves the visitors to the grottoes. There is a terrace in front of the caves that, at the time of our experiments, was largely an open expanse of dirt having small brick pathways in some areas and trees at its perimeter. The small village of Yungang, with its unpaved roads, is located to the south of the park. A major paved highway that carries thousands of coal trucks daily is located to the south and west of the village. To the south and west of that highway is a river bed. Railroad tracks carrying coal-fired locomotives are located on the far side of the river bed. A few small coal mines are located near the edges of the map, and very large coal mines are found outside the area mapped along the river bed to the west and east.

The map in Fig. 1 was subdivided into a grid system of 0.5 km by 0.5 km squares. Over the period April 19–21, 1991, glass microscope slides (75 mm by 25 mm by 1 mm thick) used as particle deposition plates were placed close to the center of each grid square at locations shown by the + symbols in Fig. 1. Additional deposition plates

were placed within the village of Yungang and along the terrace just outside of the entrance to the caves at locations shown by the solid dots in Fig. 1. The deposition plates were oriented horizontally and were whenever possible located just above eye level on top of walls where they would not be noticed or disturbed. The glass deposition plates were covered after use with a thin microscope cover slip (24 mm by 50 mm by 0.15 mm thick) to prevent collected particles from becoming detached.

The particle deposition flux to these microscope slides was determined by the micro-video imaging procedure described by Christoforou et al. (1994). A light microscope equipped with a video camera is connected to a frame-grabbing board (Quick Capture, part no. DT2255, Data Translation, Inc., Marlboro, MA) that is located within a Macintosh IIci computer. A microscope slide containing deposited particles is placed under the microscope. A digital picture of the sample is acquired with the assistance of the freely available computer program IMAGE¹, and the particles are counted and sized by the computer. Thirty video frames were captured and saved at $85 \times$ magnification and 30 more were taken at the same locations at $170 \times$ magnification on each glass slide. This allows the collection of data on particles in the size range $\geq 2 \ \mu m$. The frames need to be non-overlapping and should be spread evenly over the slide's exposed surface. We made use of verniers on the mobile microscope stage to take one frame at each magnification at each cell in a 3 \times 10 array. The spacing between frames was 3 mm. A total of 2000 particles were counted and sized whenever possible on each microscope slide, a value that has been shown to vield accurate results (Christoforou et al., 1994). Particle projected areas were converted into equivalent spherical volumes, which were then multiplied by an assumed particle mass density of 2.2 g cm⁻³ to determine the particle mass deposition flux in μg m^{-2} s⁻¹ relative to the exposed collector surface

¹The author is Wayne Rasband and the program is available by anonymous ftp from alw.nih.gov area. The above mass density was confirmed to be close to the mass density of a soil dust sample taken outside of the caves.

2.2. Chemical composition of dust deposits in the caves

Bulk samples of particulate matter were collected at Yungang from horizontal surfaces in caves 6 and 9 at locations shown in Fig. 2. The entrance to each cave faces south, and the design of these two caves is similar to many others at Yungang. An antechamber is connected to the main interior chamber of each grotto by a doorway at ground level and also by a window cut into the rock wall between the outer and inner chambers, between ~ 6 and 8.5 m above ground level. The interior chamber is hollowed out around a large central stone pillar. The pillar in cave 6 is elaborately carved with numerous smaller figures adorning all four sides. The pillar in cave 9 is carved into a single large statue of the Buddha seated in the center of the cave facing towards the window. In antiquity, the outer chamber of each cave was sheltered by a wooden temple front building several stories high and one room deep that extended up the front face of the cliff. Cave 6 retains its wooden temple front structure. The wooden shelter in front of cave 9 has fallen away leaving the rock entrances to that cave directly open to the outdoors. Outdoor air enters both caves at a high rate due to a natural convection flow that is driven by the temperature difference between the rock walls of the cave and the outdoor air.

In 1986, caves 6 and 9 were cleaned and documentary photographs were taken (Great Treasury of Chinese Fine Arts, 1986). In all but one case, the dust deposits chosen for our analysis were removed in April 1991 from locations that are known from those documentary photographs to have been cleaned in 1986. With the exception of dust sample 8, these samples reflect the composition of recent deposition from the atmosphere at Yungang.

The bulk dust samples from the caves were characterized using a battery of chemical and physical analyses. The samples were weighed



Fig. 1. Map showing a $2km \times 2km$ area surrounding the Yungang Grottoes. Numerical values shown next to the symbols + and \bullet indicate particle deposition fluxes to horizontal surfaces over the period April 19–21, 1991, in units of $\mu g m^{-2} s^{-1}$.

gravimetrically to determine the mass loading per unit surface area in the caves. Polarized light microscopy (PLM) was used to classify each sample semi-quantitatively into its component parts according to established methods (McCrone et al., 1967). Particular attention was paid to determine the extent to which coal dust was present in the deposits. Weight loss-on-ignition was used as a further method to assess coal dust content. X-ray diffraction analysis (XRD) was used to determine the mineralogy of the dust deposits.

For XRD analysis, a portion of each sample

was ground, mixed with calcium fluoride (CaF_2) as an internal standard, ground further, and sifted onto a vaseline-coated glass slide. Each slide was scanned from 4° to 64° 2Theta at 1°/min on a Philips XRG 3100 XRD unit. Graphite monochromatized copper radiation was generated at 40 kV and 35 ma. Peak positions and intensities were tabulated and were used to identify and quantify crystalline phases. Quantitative analysis was performed using calibration coefficients derived from pure mineral samples mixed with CaF₂.

The bulk powder samples also were subjected



Fig. 2. Locations of dust deposit samples taken in caves 6 and 9.

to neutron activation analysis to determine the concentrations of the elements Al, As, Au, Ba, Br, Cd, Ce, Co, Cr, Cs, Eu, Fe, Ga, Hf, Hg, In, K, La, Lu, Mg, Mn, Mo, Na, Nd, Rb, Sb, Sc, Se, Sm, Sr, Ta, Tb, Th, Ti, U, V, Yb and Zn (Olmez, 1989). Samples were extracted in de-ionized water and the concentrations of water-soluble sulfate, nitrate and chloride were determined by ion chromatography (Weiss, 1986). The aqueous extracts also were analyzed for ammonium ion concentration by colorimetry (Bolleter et al., 1961) and for water-soluble sodium and magnesium ions by atomic absorption spectroscopy (AA). Additional dry deposit samples were acid digested and analyzed for certain elements including Al, Fe, K, Mn, Ti and Zn by inductively coupled plasma mass spectrometry (ICP-MS).

2.3. Source samples

Some sources that contribute airborne particles to the atmosphere at Yungang are readily observed. These include fugitive dust entrained

by coal trucks traveling on the coal-haul highway, and dirt kicked up by traffic on the dirt roads in the village of Yungang and by activity on the dirt terrace in front of the caves (e.g. dust generated by visitors and by daily broom sweeping of the terrace). In addition, coal is piled throughout the community for future use, and these coal piles may shed coal dust particles. Coal smoke arises from coal combustion by households, industry and by railroad locomotives. An additional source of material that could contribute to the deposits in the caves will be described as cave roof-fall debris. The stone walls in some caves, including cave 6 in particular, have deteriorated due to prolonged exposure to moisture and to the atmosphere. Previous investigations by the Scientific and Technological Institute for Conservation of Cultural Relics (1988) have studied the partial conversion of the original rock walls of the cave to kaolin over time. During our experiments at the caves, it became evident that small particles of soft rock were falling from the cave ceilings

that could accumulate within deposits on the statues below. This too will be considered as a possible source contributing to the dust deposits.

Bulk samples of material from the dust sources described above were collected in annealed glass jars with solvent washed Teflon lid liners. Road dust was obtained from the very deep, black deposits on the edge of the coal-haul highway as it runs past the village of Yungang. Loose dirt was collected from the surface of the terrace located in front of cave 9. Lump coal was collected from a pile serving a kitchen and boiler in the village of Yungang. The lump coal was ground into a powder using a mortar and pestle. Cave ceiling debris was collected by sedimentation onto an aluminum collection plate placed below the ceiling in cave 6 for a period of 14 days. In addition, a sample of the native cave rock was obtained by sampling from a pile of rock removed by workmen who were renovating the interior floor of cave 5, which is immediately adjacent to cave 6. The cave rock sample was broken such that any surfaces that had been exposed to the atmosphere first were discarded. Then, the remaining native rock sample was ground into powdered form with a mortar and pestle. The bulk source powder samples were then subjected to the same set of chemical and physical analyses as described for the dust deposits removed from the statues in caves 6 and 9. Source sample material was also collected on Teflon filters and analyzed by X-ray fluorescence (XRF) for trace elements.

In addition, a sample of smoke from the combustion of coal in a local residential cooking stove was taken. Three 47-mm diameter filter samples were collected simultaneously by immersion in the plume as it exited from the rooftop chimney of a small house in the village of Yungang. Smoke was drawn at a flow rate of 10 l min⁻¹ for a duration of 8 min through each of two parallel Teflon membrane filters (Gelman, Teflo, 2.0 μ m pore size) and a third parallel quartz fiber filter (Pallflex 2500 QAO). These filter substrates were selected to be compatible with particular chemical analyses. The Teflon filters were weighed before and after sampling for gravimetric determination of smoke particle mass concentration. One Teflon filter sample from each set was analyzed

by X-ray fluorescence (XRF) for 34 elements. The other Teflon filter sample of each set was analyzed by ion chromatography (IC) for nitrate, sulfate and chloride ion concentration, for ammonium ion concentration by colorimetry and for water-soluble sodium and magnesium by atomic absorption spectroscopy (AA). Following sample collection, the quartz fiber filters were analyzed to determine the organic carbon, elemental carbon and carbonate carbon particle concentration by a thermal evolution and combustion technique (Huntzicker et al., 1982). Detailed descriptions of the filter preparation and analytical methods have been reported previously (Nazaroff et al., 1992; Ligocki et al., 1993).

3. Results

3.1. Spatial distribution of particle deposition in the vicinity of the Yungang Grottoes.

Measurements made of the spatial distribution of particle deposition rates in the vicinity of the Yungang Grottoes are shown adjacent to the + signs and solid dots in Fig. 1 in units of $\mu g m^{-2}$ s⁻¹. The highest deposition rates of ~ 60 μ g m⁻² s^{-1} are observed in the center of the map along the coal-haul highway and within the village of Yungang. Much lower values in the range 5-12 $\mu g m^{-2} s^{-1}$ occur in the countryside and near the edges of the map. The entrances to the cave temples are located between the village and the open countryside behind the grottoes and consequently are exposed to intermediate levels of deposition from the outdoor air averaging ~ 16 $\mu g m^{-2} s^{-1}$ along the terrace in front of the caves.

The existence of a high flux area within the village and along the coal-haul highway, surrounded by generally lower deposition fluxes elsewhere, strongly suggests that the coal-haul highway and activities within the village are major contributors to the local particle deposition problems. At the same time, particle fluxes at locations distant from the caves having values of $5-12 \mu g m^{-2} s^{-1}$ indicate that the regional background deposition rate is fairly high. That regional background deposition rate probably places a

lower limit on the degree of improvement of the problem at the caves that could be achieved by control of the very local sources in the village and along the coal-haul highway.

3.2. Characteristics of dust deposits in the caves

The mineralogy of the dust deposits removed from the surfaces of sculptures within the caves is described in Table 1 for samples taken at the locations shown in Fig. 2. The coal dust content given in Table 1 is estimated by weight loss-onignition while the mineral content is determined by X-ray diffraction. A comparison of the coal dust content of these dust samples as estimated by a skilled light microscopist is compared with the weight loss-on-ignition data in Table 2, and indicates, qualitatively, that much coal dust is indeed present in the deposits and that the weight loss-on-ignition data do not overestimate the amount of coal present. On average, the data of Table 1 account for 91.5% of the mass of the deposits; the occasional total in excess of 100% is still probably indistinguishable from 100% reconstitution of the sample within the error bounds of the methods used.

The dust deposits are seen to consist of a mixture of $\sim 75\%$ mineral matter and 25% coal dust. The mineral dust is largely quartz and feldspar accompanied by 3-6% calcite. This composition is consistent with its origin from the weathering of the local sandstone which contains a carbonate binder between its grains as determined by reacting freshly exposed rock surfaces with HCl. The clay content of the mineral matter in the dust deposits, which totals from 10% to as much as 22% of the dust, results from weathering of the parent rock via the process of kaolinization as mentioned earlier. Kaolin was the only clay mineral positively identified in the samples. Amorphous aluminosilicates, such as allophane or imogolite, are almost certainly present because the kaolinite peak intensities are too low to account for the 4.48 Å generic clay reflection.

Table 1 Composition of dust deposits accumulated within caves 6 and 9 at Yungang over the period 1986–1991

| Site | Location | ation Deposit depth ^a (cm) | t Mass loading ^a (g m ⁻²) | Composition ^b (% by mass) | | | | | | | | |
|---------------------|----------|---|--|--------------------------------------|-------------------------|-----------------------|---------|----------|------|--------|--------------|-------------------------|
| | | | | Quartz | Plagioclase feldspar | Potassium feldspar | Calcite | Dolomite | Clay | Gypsum | Coal dust | Total identified (%) |
| Anteroom | | | | | | | | | | | | |
| Cave 9 ^c | 1 | 0.76 | 6658 | 36 | 16 | 10 | 6 | 0 | 11 | 0 | 24 | 103 |
| Cave 9 | 2 | 0.25 | 2695 | 35 | 20 | 9 | 4 | 0 | 12 | 0 | 29 | 109 |
| Cave 9 | 3 | 0.06 | 16 ^d | 15 | 9 | 5 | 4 | 0 | 16 | 2 | 22 | 73 |
| Cave 9 | 4 | 0.13 | 572 | 38 | 19 | 10 | 5 | 2 | 10 | 0 | 23 | 107 |
| Cave 9 | 5 | 0.8 | 4981 | 44 | 6 | 13 | 3 | 1 | 22 | 0 | 22 | 114 |
| Cave 6 | 6 | 0.13 | 201 | 20 | 16 | 7 | 4 | 0 | 14 | 2 | 27 | 90 |
| Cave 6 | 7 | 0.06 | 113 | 14 | 8 | 8 | 4 | 1 | 18 | 3 | 23 | 79 |
| Cave 6 | 8 | 1.0 | 2655 ^e | 32 | 10 | 5 | 4 | 0 | 17 | 1 | 25 | 94 |
| Cave 6 | 9 | 0.5 | 156 | 18 | 16 | 14 | 5 | 1 | 11 | 3 | 24 | 92 |
| Cave 6 | 10 | 0.4 | 171 | 17 | 7 | 7 | 4 | 0 | 14 | 2 | 32 | 83 |
| Cave 6 | 11 | 0.5 | 298 | 16 | 7 | 10 | 3 | 1 | 15 | 1 | 27 | 80 |
| Cave 6 | 12 | 0.1 | 370 | 19 | 7 | 10 | 4 | 0 | 14 | 2 | 18 | 74 |

^a From Christoforou et al. (1994). Deposits are on nearly horizontal surfaces cleaned in 1986 except for locations 3 and 8.

^bQuartz, plagioclase feldspar, potassium feldspar, calcite, dolomite, clay and gypsum are determined by X-ray diffraction analysis; coal dust is determined by weight loss-on-ignition. Precision for X-ray diffraction analysis is $\pm 10\%$ relative to the reported value for values > 10\%, $\pm 20\%$ for values from 2–10% and $\pm 50\%$ for values $\leq 2\%$.

^cLocation is essentially outdoors given the lack of a wooden temple structure in front of cave 9.

^dDeposit on a nearly vertical surface.

^eDeposit on a horizontal surface not cleaned in 1986.

| Site | Location | Loss-on-ignition ^a | Polarized light microscopy ^b |
|---------------------|---------------------------------------|-------------------------------|--|
| Dust deposits | · · · · · · · · · · · · · · · · · · · | | |
| Anteroom cave 9 | 1 ^c | 24 | 15 |
| Cave 9 | 2 | 29 | 15 |
| Cave 9 | 3ª | 22 ^e | 50 |
| Cave 9 | 4 | 23 | 20 |
| Cave 9 | 5 | 22 | 15 |
| Cave 6 | 6 | 27 | 40 |
| Cave 6 | 7 | 23 | 60 |
| Cave 6 | 8^{f} | 25 | 20 |
| Cave 6 | 9 | 24 | 30 |
| Cave 6 | 10 | 32 | 20 |
| Cave 6 | 11 | 27 | 20 |
| Cave 6 | 12 | 18 | 20 |
| Source samples | | | |
| Soil dust | | 11 ^e | 10 |
| Coal road dust | | 35 | 50 |
| Coal dust | | 97 | 100 |
| Cave ceiling debris | | 4 ^e | 3 |
| Cliff rock | | 6 | 0 |

| Table 2 | - |
|---|------------|
| Comparison of coal dust content by weight loss-on-ignition versus polarized light | microscopy |

^a The estimated precision for the weight loss-on-ignition method is $\pm 10\%$ relative to the reported value for values > 10\%, $\pm 20\%$ for values from 5-10% and $\pm 50\%$ for values from 1-5%.

^bThe estimated precision for the polarized light microscopy method is $\pm 20\%$ relative to the reported value for values > 25%, $\pm 50\%$ for values from 5-25% and $\pm 100\%$ for values $\leq 5\%$.

^cLocation is essentially outdoors given the lack of a wooden temple structure in front of cave 9.

^dDeposit on a nearly vertical surface.

^cAverage of two samples.

^fDeposit on a horizontal surface not cleaned in 1986.

Coal burning activities near Yungang produce SO_2 gas which could react with the binder in local sandstone to produce gypsum. Gypsum is identified at a concentration of 1-3% by mass in the dust deposits, and magnesium sulfate is seen on the surface of a native rock sample that was exposed historically to the atmosphere at Yungang. Gypsum is not seen in the interior of the crushed 'source rock sample' that will be discussed shortly. The implication is that SO_2 in the atmosphere at Yungang may lead to some gypsum formation in places where calcareous minerals are exposed to the atmosphere.

Tables 3a and b show the concentrations of selected chemical elements and ionic substances present in the dust deposits as determined by neutron activation analysis, ion chromatography and colorimetry. Elements characteristic of soil and rock dust including Al, Fe, Mn and Ti are present as expected. Potassium is found as would be expected from the potassium feldspar detected by X-ray diffraction. Mercury and arsenic are found in the cave dust deposits at surprisingly high concentrations that are not characteristic of the native rock or soil. We believe that this mercury and arsenic is probably derived from mercury sulfide and arsenic sulfide minerals (e.g. vermilion and orpiment) that have been used as pigments to decorate the statues in the caves (Pique, 1993).

3.3. Characteristics of source samples

Tables 4 and 5 provide a mineralogical and elemental characterization of the source samples

| Table 3a | | | | | | |
|--------------------------|-------|---------|----------|--------|-----------|---------|
| Chemical characteristics | (% by | / mass) | measured | in dus | t deposit | samples |

| Species ^a | Cave 9 Dust 1 | Cave 9 Dust 2 | Cave 9 Dust 3 | Cave 9 Dust 4 | Cave 9 Dust 5 | Cave 6 Dust 6 |
|--|---|---|---|---|---|---|
| Na Mg Al K | $\begin{array}{c} 1.28 \pm 0.08 \\ 1.49 \pm 0.26 \\ 5.57 \pm 0.23 \\ 1.81 \pm 0.16 \end{array}$ | $1.48 \pm 0.1 \\ 1.16 \pm 0.2 \\ 6.4 \pm 0.27 \\ 2.09 \pm 0.18$ | $\begin{array}{c} 0.743 \pm 0.049 \\ 1.4 \pm 0.23 \\ 6.4 \pm 0.26 \\ 1.6 \pm 0.16 \end{array}$ | $\begin{array}{c} 1.19 \pm 0.08 \\ 1.11 \pm 0.21 \\ 5.89 \pm 0.25 \\ 1.69 \pm 0.17 \end{array}$ | $\begin{array}{c} 0.508 \pm 0.033 \\ 1.5 \pm 0.25 \\ 6.97 \pm 0.28 \\ 2.75 \pm 0.21 \end{array}$ | $\begin{array}{c} 1.01 \pm 0.07 \\ 1.98 \pm 0.33 \\ 5.67 \pm 0.23 \\ 1.53 \pm 0.13 \end{array}$ |
| Ti V Cr Mn | $\begin{array}{c} 0.393 \pm 0.039 \\ 0.0058 \pm 0.0004 \\ 0.0062 \pm 0.0003 \\ 0.049 \pm 0.001 \end{array}$ | $\begin{array}{c} 0.543 \pm 0.053 \\ 0.0071 \pm 0.0004 \\ 0.0079 \pm 0.0003 \\ 0.054 \pm 0.001 \end{array}$ | $\begin{array}{c} 0.377 \pm 0.038 \\ 0.0069 \pm 0.0004 \\ 0.0061 \pm 0.0002 \\ 0.048 \pm 0.001 \end{array}$ | $\begin{array}{c} 0.383 \pm 0.041 \\ 0.0065 \pm 0.0005 \\ 0.0054 \pm 0.0003 \\ 0.049 \pm 0.001 \end{array}$ | $\begin{array}{c} 0.34 \pm 0.03 \\ 0.0062 \pm 0.0004 \\ 0.0051 \pm 0.0002 \\ 0.038 \pm 0.001 \end{array}$ | $\begin{array}{c} 0.383 \pm 0.034 \\ 0.0064 \pm 0.0004 \\ 0.0059 \pm 0.0002 \\ 0.055 \pm 0.001 \end{array}$ |
| Fe Zn Ga As | $\begin{array}{c} 2.46 \pm 0.12 \\ 0.0079 \pm 0.0044 \\ 0.0010 \pm 0.0005 \\ 0.0012 \pm 0.0001 \end{array}$ | $\begin{array}{c} 2.79 \pm 0.12 \\ 0.0057 \pm 0.0022 \\ 0.0017 \pm 0.0011 \\ 0.0013 \pm 0.0001 \end{array}$ | $\begin{array}{c} 2.74 \pm 0.11 \\ 0.0116 \pm 0.0028 \\ 0.0006 \pm 0.0006 \\ 0.011 \pm 0.001 \end{array}$ | $\begin{array}{c} 2.69 \pm 0.12 \\ 0.0028 \pm 0.0018 \\ 0.0011 \pm 0.0007 \\ 0.0018 \pm 0.0002 \end{array}$ | $\begin{array}{c} 2.03 \pm 0.08 \\ 0.0066 \pm 0.002 \\ 0.0007 \pm 0.0005 \\ 0.0036 \pm 0.0004 \end{array}$ | $\begin{array}{c} 2.72 \pm 0.11 \\ 0.011 \pm 0.002 \\ 0.0011 \pm 0.0006 \\ 0.0026 \pm 0.0003 \end{array}$ |
| Rb Sr Ba La | $\begin{array}{c} 0.0066 \pm 0.0018 \\ 0.053 \pm 0.007 \\ 0.052 \pm 0.005 \\ 0.0036 \pm 0.0002 \end{array}$ | $\begin{array}{c} 0.0058 \pm 0.0012 \\ 0.031 \pm 0.008 \\ 0.067 \pm 0.006 \\ 0.0036 \pm 0.0002 \end{array}$ | $\begin{array}{c} 0.0080 \pm 0.0014 \\ 0.016 \pm 0.005 \\ 0.051 \pm 0.005 \\ 0.0033 \pm 0.0002 \end{array}$ | $\begin{array}{c} 0.0078 \pm 0.0017 \\ 0.027 \pm 0.008 \\ 0.053 \pm 0.005 \\ 0.0043 \pm 0.0002 \end{array}$ | $\begin{array}{c} 0.0089 \pm 0.0014 \\ 0.034 \pm 0.005 \\ 0.096 \pm 0.006 \\ 0.0036 \pm 0.0002 \end{array}$ | $\begin{array}{c} 0.0080 \pm 0.0012 \\ 0.018 \pm 0.005 \\ 0.057 \pm 0.005 \\ 0.0032 \pm 0.0002 \end{array}$ |
| Ce Nd Hg | $\begin{array}{c} 0.0071 \pm 0.0004 \\ 0.0027 \pm 0.0004 \\ 0.000023 \pm 0.000012 \end{array}$ | $\begin{array}{c} 0.0066 \pm 0.0003 \\ 0.0024 \pm 0.0003 \\ 0.00005 \pm 0.000009 \end{array}$ | $\begin{array}{c} 0.0063 \pm 0.0003 \\ 0.0025 \pm 0.0003 \\ 0.0011 \pm 0.0001 \end{array}$ | $\begin{array}{c} 0.0079 \pm 0.0004 \\ 0.0032 \pm 0.0004 \\ 0.000063 \pm 0.000018 \end{array}$ | $\begin{array}{c} 0.0059 \pm 0.0003 \\ 0.0024 \pm 0.0003 \\ 0.0002 \pm 0.00001 \end{array}$ | $\begin{array}{c} 0.0061 \pm 0.0003 \\ 0.0025 \pm 0.0003 \\ 0.00093 \pm 0.00003 \end{array}$ |
| Th Cl ^{-b} SO ₄ ^{=b} NH ₄ ^{+c} | $\begin{array}{c} 0.0012 \pm 0.0001 \\ 0.063 \pm 0.015 \\ 0.60 \pm 0.04 \\ 0.009 \pm 0.001 \end{array}$ | $\begin{array}{c} 0.0011 \pm 0.0001 \\ 0.055 \pm 0.002 \\ 1.15 \pm 0.02 \\ 0.012 \pm 0.002 \end{array}$ | $\begin{array}{c} 0.0010 \pm 0.00006 \\ 0.034 \pm 0.011 \\ 2.25 \pm 0.06 \\ 0.050 \pm 0.011 \end{array}$ | $\begin{array}{c} 0.0012 \pm 0.0001 \\ 0.002 \pm 0.002 \\ 0.24 \pm 0.01 \\ 0.0 \pm 0.001 \end{array}$ | $\begin{array}{c} 0.0006 \pm 0.00004 \\ 0.030 \pm 0.001 \\ 1.10 \pm 0.01 \\ 0.005 \pm 0.001 \end{array}$ | $\begin{array}{c} 0.0009 \pm 0.00005 \\ 0.017 \pm 0.009 \\ 1.17 \pm 0.04 \\ 0.0 \pm 0.001 \end{array}$ |

Note: the following species were also analyzed by neutron activation analysis and found to be < 0.001% by mass in all cases; Sc, Co, Se, Br, Mo, Cd, In, Sb, Cs, Sm, Eu, Tb, Yb, Lu, Hf, Ta, Au and U.

^aDetermined by neutron activation analysis unless otherwise noted.

^bDetermined by ion chromatography.

^cDetermined by colorimetry.

by the same methods used to characterize the dust deposit samples taken from the caves, with the exception of native rock dust and coal smoke which were analyzed by other methods noted in the table. Not surprisingly, coal dust tests as 97% 'coal' with 3-4% mineral matter (ash). The local soil dust sample contains quartz, feldspar and calcite. The clay content of local soil dust is ~ 6%, which is less than the clay content of the dust deposits in the caves. The likely source of the elevated clay levels in the cave deposits is seen to be cave ceiling fall debris which has a high clay level as would be expected from the previously reported kaolinization of the cave surfaces.

The paved road dust from the coal-haul highway contains ~ 35% coal dust by mass. This high coal content is completely consistent with our observations of dust generation along that road. Coal trucks piled with uncovered coal loads to well above the top of the sides of the truck beds were observed to travel the road at a separation of approximately one to two truck lengths during most of the day. Lumps of coal were observed to fall from the trucks routinely to be crushed by the wheels of the trucks that follow. As a result, the dust deposits along the shoulders of this road in 1991 were several centimeters deep and very black.

Chemical analysis of the coal smoke sample

| Species ^a | Cave 6 | Cave 6 | Cave 6 | Cave 6 | Cave 6 | Cave 6 |
|-------------------------------|-----------------------|-----------------------|-----------------------|-----------------------|----------------------|----------------------|
| | Dust 7 | Dust 8 | Dust 9 | Dust 10 | Dust 11 | Dust 12 |
| Na | 0.886 ± 0.058 | 1.16 ± 0.08 | 0.9 ± 0.059 | 0.86 ± 0.056 | 0.807 ± 0.053 | 1.00 ± 0.07 |
| Mg | 1.99 ± 0.33 | 1.38 ± 0.23 | 1.95 ± 0.33 | 1.55 ± 0.27 | 1.82 ± 0.32 | 2.11 ± 0.36 |
| Aľ | 5.63 ± 0.23 | 5.74 ± 0.23 | 5.62 ± 0.23 | 5.74 ± 0.24 | 6.37 ± 0.26 | 5.73 ± 0.24 |
| К | 1.57 ± 0.15 | 1.83 ± 0.16 | 1.37 ± 0.14 | 1.63 ± 0.16 | 1.6 ± 0.12 | 1.43 ± 0.12 |
| Ti | 0.351 ± 0.03 | 0.375 ± 0.032 | 0.325 ± 0.032 | 0.347 ± 0.042 | 0.387 ± 0.035 | 0.392 ± 0.037 |
| v | 0.0069 ± 0.0004 | 0.0070 ± 0.0004 | 0.0073 ± 0.0004 | 0.0071 ± 0.0005 | 0.0070 ± 0.0005 | 0.0067 ± 0.0005 |
| Cr | 0.0073 ± 0.0002 | 0.006 ± 0.0002 | 0.007 ± 0.0003 | 0.0063 ± 0.0002 | 0.0067 ± 0.0004 | 0.0067 ± 0.0002 |
| Mn | 0.055 ± 0.001 | 0.052 ± 0.001 | 0.054 ± 0.001 | 0.053 ± 0.001 | 0.056 ± 0.001 | 0.058 ± 0.001 |
| Fe | 3.17 ± 0.13 | 2.64 ± 0.11 | 2.97 ± 0.12 | 2.88 ± 0.12 | 2.94 ± 0.13 | 2.88 ± 0.12 |
| Zn | 0.017 ± 0.004 | 0.0089 ± 0.002 | 0.019 ± 0.004 | 0.016 ± 0.003 | 0.017 ± 0.004 | 0.013 ± 0.003 |
| Ga | 0.0010 ± 0.0006 | $0.0003 \pm d$ | 0.0012 ± 0.0006 | 0.0022 ± 0.0008 | 0.0013 ± 0.0008 | 0.0007 ± 0.0011 |
| As | 0.0032 ± 0.0003 | 0.0034 ± 0.0003 | 0.0022 ± 0.0002 | 0.0022 ± 0.0002 | 0.0024 ± 0.0002 | 0.003 ± 0.0003 |
| Rb | 0.0083 ± 0.0012 | 0.0080 ± 0.0012 | 0.0076 ± 0.0013 | 0.0075 ± 0.0012 | 0.0065 ± 0.0016 | 0.0081 ± 0.0011 |
| Sr | 0.027 ± 0.005 | 0.017 ± 0.005 | 0.024 ± 0.005 | 0.016 ± 0.007 | 0.032 ± 0.007 | 0.029 ± 0.006 |
| Ba | 0.056 ± 0.005 | 0.063 ± 0.005 | 0.069 ± 0.006 | 0.054 ± 0.005 | 0.067 ± 0.006 | 0.060 ± 0.005 |
| La | 0.0034 ± 0.0002 | 0.0031 ± 0.0002 | 0.0035 ± 0.0002 | 0.0031 ± 0.0002 | 0.0034 ± 0.0002 | 0.0037 ± 0.0002 |
| Ce | 0.0064 ± 0.0003 | 0.0062 ± 0.0003 | 0.0066 ± 0.0003 | 0.0062 ± 0.0003 | 0.0068 ± 0.0004 | 0.0071 ± 0.0003 |
| Nd | 0.0026 ± 0.0003 | 0.0023 ± 0.0003 | 0.0023 ± 0.0003 | 0.0023 ± 0.0003 | 0.0021 ± 0.0003 | 0.0026 ± 0.0003 |
| Hg | 0.0042 ± 0.0001 | 0.013 ± 0.0003 | 0.0038 ± 0.0001 | 0.0197 ± 0.0004 | 0.0024 ± 0.0001 | 0.0150 ± 0.0001 |
| Th | 0.00094 ± 0.00006 | 0.00092 ± 0.00006 | 0.00091 ± 0.00006 | 0.00093 ± 0.00006 | 0.00010 ± 0.0001 | 0.00010 ± 0.0001 |
| Cl-b | 0.035 ± 0.003 | 0.014 ± 0.003 | 0.057 ± 0.009 | 0.024 ± 0.011 | 0.012 ± 0.007 | 0.035 ± 0.003 |
| SO ₄ ^{=b} | 1.03 ± 0.02 | 0.83 ± 0.01 | 2.08 ± 0.05 | 1.23 ± 0.04 | 0.84 ± 0.03 | 1.32 ± 0.02 |
| NH ⁺ | 0.013 ± 0.003 | 0.010 ± 0.003 | 0.039 ± 0.009 | 0.059 ± 0.001 | 0.0 ± 0.001 | 0.015 ± 0.003 |

Table 3b Chemical characteristics (% by mass) measured in dust deposit samples

Note: the following species were also analyzed by neutron activation analysis and found to be < 0.001% by mass in all cases; Sc, Co, Se, Br, Mo, Cd, In, Sb, Cs, Sm, Eu, Tb, Yb, Lu, Hf, Ta, Au and U.

^aDetermined by neutron activation analysis unless otherwise noted.

^bDetermined by ion chromatography.

^cDetermined by colorimetry.

d, not determined.

shows that it consists of 39% organic carbon, 5.7% elemental (black) carbon, plus ~1% each Al, Ca, Cl⁻ and $SO_4^{=}$, with lesser amounts of crustal metals such as Fe, Ti, Mn, K and Mg as shown in Table 5.

3.4. Source-receptor reconciliation

The dust deposits within the caves undoubtedly reflect the combined effect of the deposition of a mixture of particles from several sources. Chemical mass balance models have been developed that seek to assist the estimation of the most probable linear combination of emission sources that contribute to ambient particulate matter samples (Miller et al., 1972; Friedlander, 1973). Such modeling procedures also can be applied to relate deposited airborne particles to their source.

In chemical mass balance models, the mass concentration of a chemical species at a receptor site (in this case a receptor dust deposit in the caves) is taken to be a linear combination of the emissions of that species from the various sources

$$C_{ik} = \sum_{j=1}^{m} a_{ij} f_{ijk} s_{jk}$$
(1)

where c_{ik} is the mass fraction of species *i* in

| Source type | Composition ^a (% by mass) | | | | | | | | |
|--|--------------------------------------|-------------------------|-----------------------|---------|----------|------|--------|--------------|-------------------------|
| | Quartz | Plagioclase feldspar | Potassium feldspar | Calcite | Dolomite | Clay | Gypsum | Coal dust | Total identified (%) |
| Soil dust ^b | 32 | 11 | 7 | 17 | 1 | 6 | 0 | 11 | 85 |
| Coal-haul highway ^b road dust | 29 | 19 | 6 | 3 | 2 | 8 | 0 | 35 | 102 |
| Coal dust | 1 | 0 | 3 | 0 | 0 | 0 | 0 | 97 | 101 |
| Cave ceiling debris | 53 | 0 | 40 | 0 | 2 | 16 | 0 | 4 | 115 |
| Native cliff rock ^c | 56 | 0 | 13 | 1 | 1 | 9 | 0 | 6 | 86 |

| Table 4 | | | | |
|----------------------------|---------------|----------|---------|----------|
| Composition of dust source | samples taken | near the | Yungang | Grottoes |

^a Quartz, plagioclase feldspar, potassium feldspar, calcite, dolomite, clay and gypsum are determined by X-ray diffraction analysis; coal dust is determined by weight loss-on-ignition. The estimated precision for the weight loss-on-ignition method is $\pm 10\%$ relative to the reported value for values > 10%, $\pm 20\%$ for values from 5–10% and $\pm 50\%$ for values from 1–5%. Precision for X-ray diffraction analysis is $\pm 10\%$ relative to the reported value for values s > 10%, $\pm 20\%$ for values s > 10%, $\pm 20\%$ for values s > 10%, $\pm 20\%$ for values from 2–10% and $\pm 50\%$ for values $\leq 2\%$.

^bAverage of two samples.

^cInterior of rock after having removed all surfaces previously exposed to the atmosphere.

receptor deposit k, a_{ij} is the mass fraction of species i in the total direct emissions from source j, f_{iik} is the fractionation coefficient for species i as it travels from source j to the particle deposit k and s_{ik} is the mass fraction of deposit k that is due to the combined presence of all the species contributed by source j. The unknowns to be determined are the relative source contributions, s_{ik} , to deposited particle mass accumulations due to the m different source types present in the community. The fractionation coefficient, f_{iik} , is used to adjust the relative composition of material from source *j* for distortion of its chemical composition profile which may occur due to preferential removal of some species but not others during atmospheric transport from the source to the receptor. In the present analysis, we will assume that the coarse particle species of interest here sediment from the atmosphere at equal rates, such that the relative composition of source material reaching the receptor deposits still resembles the relative mineralogical composition seen at the source (f_{iik} is taken to be 1.0 in all cases).

For a single receptor deposit, Eq. 1 is written n times, once for each distinct mineral substance emitted by the sources. An over-determined set of equations results, provided that the number of mineral phases studied exceeds the number of sources. The set of equations is then solved by

regression analysis to obtain the combination of the source contributions which explains the composition of the receptor deposits while leaving the minimum square residual error. The computer program used in the present paper to solve the system of Eq. 1 is the CMB7 computer program (Watson et al., 1990) distributed by the US Environmental Protection Agency.

The dust deposits in the caves were viewed as an unknown linear combination of the soil dust, coal-haul highway road dust, coal dust, cave ceiling debris and powdered native cliff rock source samples described earlier. The mineralogical compositions of the source material and receptor dust deposits are as shown in Tables 1 and 4. When this calculation is performed using all source types simultaneously, the estimated contributions to the cave dust deposits from some sources are insignificantly different from zero. Also, given the similarity of several of these source types, they often cannot be resolved separately. The computer-based fitting calculation was thus run repeatedly, at each step removing sources for which physically impossible contributions were generated (i.e. negative values arising from collinearity of one source with a different source) and then eliminating sources whose contributions were insignificantly different from zero. The result is an estimate of a combination of sources

| Species | Soil ^b dust | Highway ^b road dust | Coal ^b dust | Cave ^b ceiling | Native ^c rock | Coal ^d smoke |
|-------------------|---------------------------|-----------------------------------|---------------------------|------------------------------|-----------------------------|----------------------------|
| | | | | debris | ausi | |
| Na | 0.748 ± 0.049 | 0.708 ± 0.046 | 0.0096 ± 0.0009 | 0.178 ± 0.012 | 0.414 ± 0.021 | 0.174 ± 0.019 |
| Mg | 0.530 ± 0.103 | 0.590 ± 0.120 | 0.0098 ± 0.0062 | 1.69 ± 0.29 | 0.207 ± 0.011 | 0.164 ± 0.018 |
| Aľ | 4.45 ± 0.18 | 3.41 ± 0.14 | 0.167 ± 0.008 | 8.11 ± 0.33 | 8.43 ± 0.84 | 1.25 ± 0.16 |
| K | 1.75 ± 0.13 | 1.62 ± 0.14 | 0.025 ± 0.002 | 2.58 ± 0.17 | 2.62 ± 0.17 | 1.18 ± 0.13 |
| Ti | 0.287 ± 0.023 | 0.217 ± 0.021 | 0.0148 ± 0.0041 | 0.691 ± 0.047 | 0.105 ± 0.007 | 0.098 ± 0.006 |
| v | 0.0048 ± 0.0003 | 0.0042 ± 0.0003 | 0.0003 ± 0.00004 | 0.0082 ± 0.0004 | e | e |
| Cr | 0.0055 ± 0.0002 | 0.003 ± 0.0002 | 0.00027 ± 0.00003 | 0.0069 ± 0.0003 | 0.0000 ± 0.0002 | 0.0014 ± 0.0010 |
| Mn | 0.030 ± 0.0004 | 0.043 ± 0.001 | 0.0002 ± 0.00001 | 0.045 ± 0.001 | 0.024 ± 0.002 | 0.0073 ± 0.0012 |
| Fe | 1.74 ± 0.07 | 3.00 ± 0.13 | 0.025 ± 0.005 | 2.28 ± 0.10 | 1.39 ± 0.14 | 0.47 ± 0.026 |
| Zn | 0.0069 ± 0.0017 | 0.0053 ± 0.0028 | 0.0012 ± 0.0004 | 0.0034 ± 0.0009 | 0.0023 ± 0.0009 | 0.1453 ± 0.0083 |
| Ga | $0.0020 \pm e$ | 0.0010 ± 0.0005 | 0.00069 ± 0.00037 | 0.0030 ± 0.0014 | 0.0000 ± 0.0001 | 0.0000 ± 0.0009 |
| As | 0.00037 ± 0.00004 | 0.00092 ± 0.00009 | 0.000018 ± 0.000004 | 0.00013 ± 0.00002 | 0.0000 ± 0.0002 | 0.0056 ± 0.0034 |
| Rb | 0.0064 ± 0.0009 | 0.0041 ± 0.0012 | 0.00027 ± e | 0.0065 ± 0.0010 | 0.0000 ± 0.0001 | 0.0013 ± 0.0018 |
| Sr | 0.028 ± 0.005 | 0.019 ± 0.004 | 0.0015 ± 0.0003 | 0.029 ± 0.005 | 0.0016 ± 0.0002 | 0.0029 ± 0.0021 |
| Ba | 0.080 ± 0.006 | 0.074 ± 0.006 | 0.0019 ± 0.006 | 0.099 ± 0.007 | 0.000 ± 0.011 | 0.041 ± 0.100 |
| La | 0.0026 ± 0.0002 | 0.0015 ± 0.0001 | 0.00031 ± 0.00002 | 0.0037 ± 0.0002 | 0.0000 ± 0.0116 | 0.034 ± 0.151 |
| Ce | 0.0048 ± 0.0002 | 0.0029 ± 0.0002 | 0.00047 ± 0.00003 | 0.0063 ± 0.0003 | e | e |
| Nd | 0.0021 ± 0.0003 | 0.0012 ± 0.0002 | 0.00021 ± 0.00005 | 0.0021 ± 0.0003 | e | e |
| Hg | 0.000018 ± 0.000007 | 7 0.000024 ± 0.000008 | 0.000044 ± 0.000002 | 0.000015 ± 0.000007 | 0.0000 ± 0.0001 | 0.0000 ± 0.0024 |
| Th | 0.00051 ± 0.00003 | 0.00039 ± 0.00003 | 0.00025 ± 0.00001 | 0.00084 ± 0.00005 | e | e |
| Cl- | 0.229 ± 0.007 | 0.048 ± 0.004 | 0.019 ± 0.016 | 0.039 ± 0.004 | 0.017 ± 0.013 | 1.106 ± 0.131 |
| SO ₄ = | 1.869 ± 0.031 | 0.632 ± 0.013 | 0.568 ± 0.041 | 0.429 ± 0.012 | 0.718 ± 0.009 | 0.822 ± 0.032 |
| NH_4^+ | 0.027 ± 0.004 | 0.018 ± 0.003 | 0.029 ± 0.016 | 0.000 ± 0.001 | 0.006 ± 0.001 | 0.168 ± 0.012 |

Table 5 Chemical characteristics (% by mass) measured in source samples^a

^aNote: the following species were also analyzed by neutron activation analysis and found to be < 0.001% by mass in all cases; Sc, Co, Se, Br, Mo, Cd, In, Sb, Cs, Sm, Eu, Tb, Yb, Lu, Hf, Ta, Au and U.

^bAll elements determined by neutron activation analysis except for Cl^- and SO_4^- determined by ion chromatography and NH_4^+ determined by colorimetry.

^cNa and Mg determined by atomic absorption, Al, K, Ti, Mn, Fe and Zn determined by inductively coupled plasma mass spectrometry (ICP MS), other elements determined by X-ray fluorescence.

^dNa and Mg determined by atomic absorption, other elements determined by X-ray fluorescence.

e, no data available.

that would explain the receptor deposits in a way that each estimate of a source contribution is both significantly different from zero with > 95% confidence and positive.

Table 6 shows the calculated source contributions to the dust deposits in the caves. The deposits are estimated to consist largely of a combination of local soil dust (10-20%), deteriorated cave ceiling material (9-17%), plus a larger amount of road dust from the coal-haul highway (mean value 53%, range 27-92%). The dust deposition site most exposed to the outdoor activities along the dirt terrace in front of the caves is deposit sample 1, and it, not surprisingly, contains the highest estimated contribution from soil dust like that sampled from the terrace. Sample 5 taken from the rear of cave 9 is estimated to contain a large amount of native rock dust, which might be related to our observation that the surface of the ceiling at the rear of cave 9 had in places delaminated and fallen off in large patches, possibly due to water intrusion from behind the surface through the rock.

Samples 7, 10 and 11 show lower than average

| Site | Location | Soil dust | Highway road dust | Coal dust | Cave ceiling debris | Native rock dust | Total mass explained (%) | r ² |
|-----------------|----------------|----------------|-------------------------|----------------|---------------------------|------------------------|--------------------------------|----------------|
| Anteroom cave 9 | 1 ^a | 25.7 ± 7.7 | 63.4 ± 7.9 | | 13.4 ± 3.5 | | 103 | 0.96 |
| Cave 9 | 2 | | 92.4 ± 8.0 | _ | 10.6 ± 3.7 | | 103 | 0.94 |
| Cave 9 | 3 ^b | 14.8 ± 5.3 | 47.5 ± 5.7 | _ | | | 62 | 0.81 |
| Cave 9 | 4 | 19.3 ± 7.2 | 73.3 ± 8.3 | | 13.1 ± 3.6 | · | 106 | 0.98 |
| Cave 9 | 5 | | 41.7 ± 5.7 | _ | _ | 78.9 ± 9.0 | 121 | 0.90 |
| Cave 6 | 6 | | 82.7 ± 5.9 | | _ | _ | 83 | 0.88 |
| Cave 6 | 7 | 16.0 ± 5.8 | 27.9 ± 9.0 | 10.9 ± 3.9 | 9.3 ± 3.7 | _ | 64 | 0.79 |
| Cave 6 | 8° | - | 61.8 ± 6.2 | — | _ | 27.4 ± 6.6 | 89 | 0.86 |
| Cave 6 | 9 | 13.6 ± 6.7 | 57.0 ± 7.4 | | 14.6 ± 3.8 | | 85 | 0.90 |
| Cave 6 | 10 | 19.5 ± 6.4 | 27.4 ± 8.6 | 20.4 ± 4.8 | 10.9 ± 3.5 | _ | 78 | 0.87 |
| Cave 6 | 11 | 10.8 ± 4.5 | 29.4 ± 7.8 | 15.1 ± 4.1 | 17.4 ± 3.0 | | 73 | 0.87 |
| Cave 6 | 12 | 14.0 ± 5.4 | 36.6 ± 5.7 | | 17.0 ± 3.0 | | 68 | 0.89 |

Relative source contributions consistent with the mineralogical characterization of dust deposits and source samples reported in Tables 1 and 4

^aLocation is essentially outdoors given the lack of a wooden temple structure in front of cave 9.

^bDeposit on a nearly vertical surface.

Table 6

^cDeposit on a horizontal surface not cleaned in 1986.

road dust contributions plus a separate estimate for additional coal dust within the deposit. The dust from the coal-haul highway is a blend of mineral matter and coal. The extra coal could have originated from the road dust problem. We have systematically compared the dust deposits in the caves with combinations of the pure coal dust source plus the other non-highway mineral dust sources while excluding the separate road dust source. The model consistently fits the composition of the dust deposits more closely when the highway dust profile is used than when we attempt to synthesize the road dust from a combination of the coal and other mineral dust sources. We have also appended a selection of the major chemical species concentrations determined by neutron activation and ion chromatography to the source profile defined by the minerals present in each source and deposit sample, and then repeated the source attribution calculations. The road dust source is again identified as by far the largest contributor to the dust deposits.

4. Conclusions

A series of experiments have been conducted

to explore the relationship between airborne particle emission sources and dust deposition within the Buddhist cave temple complex at Yungang, China. The spatial distribution of particle deposition fluxes in the community near the caves has been measured to identify areas of high deposition flux that would indicate the location of major particle sources. The mineralogy of the dust deposits in the caves has been compared with the mineralogy of the most obvious particle sources in the community, including local soil dust from the terrace in front of the caves, road dust from the major coal-haul highway near the caves, deteriorated cave ceiling debris, pure coal dust and native cliff rock dust.

Measurements of the spatial distribution of dust deposition within a 2 km \times 2 km geographical area surrounding the grottoes show an area of high deposition flux, averaging ~ 60 μ g m⁻² s⁻¹ along the coal-haul highway and within the center of the village of Yungang next to the caves. This suggests that the coal truck traffic and activities on the dirt roads in the village are major local particle sources. The mineralogical composition of the dust deposits in the caves is consistent with the dust being largely a combination of 10-20% local soil dust, 9-17% deteriorated cave ceiling (or wall) debris and on average 53% coal-haul highway road dust (range 27-92%).

The similarity of several of the mineral sources examined, plus the fact that the road dust is a mix of minerals plus coal, makes it impossible to say with absolute certainty exactly which sources have created the dust deposits. Nevertheless, all available evidence suggests that dust from the coalhaul highway plus local soil dust sources in the village and along the terrace in front of the caves combine with particles shed from deteriorated rock surfaces interior to the caves to create the dust deposits on the sculptures. In addition, the presence of mercury and arsenic in the dust deposits suggests that pigments used to decorate the sculptures contain toxic trace metals that become incorporated into the dust deposits. Caution should be exercised when handling or disturbing the dust.

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