

# **Coal Fire Sublimates: Are We Missing Something?**

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Abstract: Uncontrolled coal fires present a nearly unparalleled environmental and human health disaster. These fires can cause the destruction of the ecosystem, ignite forest fires, become a source of windblown dust and siltation of streams, and pollute surface water, ground water, and crops. They can cause significant disruption of families and communities resulting from physical hazards of collapse or explosion, excessive heat, visual blight and loss of potentially valuable acreage, deterioration of cultural infrastructure, personal and public property, and loss of a valuable energy resource. The emission of CO<sub>2</sub> and other greenhouse gases presents a significant health hazard due to respiration of dust and aerosols, and exposure to acidic gases, potentially toxic trace elements, and organic compounds. Numerous studies have described many dozens of phases that have condensed from the effluents of these fires. However, many of these studies may have overlooked the nano- and ultra-fine particles that exist beside, in, and under the brightly colored, often spectacular crystalline macro phases. Using a scanning electron microscope with an energy dispersive X-ray detector we examined a small (30 mm  $\times$  10 mm) piece of condensate from an uncontrolled coal fire in the Jharia region of India and found more than 30 different phases in this one small piece. The phases included ammonium, copper, iron, lead, bismuth chlorides; bismuth, lead, and ammonium silici-fluorides; ammonium and lead iodates; iron, barium, lead, copper, and zinc sulfides; iron and silicon oxides; and others. Broken fragments revealed multiple generations of phases. Though not strictly nanoparticles (smaller than 0.1  $\mu$ m), many of these particles are in the micrometer to sub-micron range and it is likely that there are phases present in the nanoparticle size range. Certainly, particulates in the nanoparticle and ultra-fine particle range are being released from the uncontrolled coal fires and may be impacting the environment and the health of the mine workers and nearby residents, an issue that should be investigated.

Keywords: microminerals; nanominerals; sublimates; coal fires

## 1. Introduction

Uncontrolled coal fires have likely occurred in every country where coal has been mined or is exposed at the surface. Nichols and Finkelman [1] list almost 40 countries where these fires have been reported. Hundreds to thousands of active coal fires, both large and small, are burning today in a number of countries including China, India, the USA, South Africa, Australia, Brazil, and Russia, as well as several countries in Europe. The fires present an unparalleled environmental disaster. Finkelman [2] noted that these fires cause visual blight, the destruction of the ecosystem, and pollution of soils, surface water, ground water, and crops; they also initiate forest fires, and are a source of windblown dust and siltation of streams. There is also the risk of deterioration of cultural infrastructure and personal and public property, loss of a valuable energy resource, loss of potentially valuable acreage, disruption of families and communities, physical hazards from collapse or explosion, and safety hazards from excessive heat. The emission of  $CO_2$  and other greenhouse gases also presents a significant health hazard due to respiration of dust and aerosols, and exposure to acid gases and to potentially toxic trace elements and organic compounds.



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Although the fires have caused several accidental deaths in the USA, India, and Zimbabwe, little has been reported on their long- and short-term health impacts. Dhar et al. [3] demonstrated that in the Jharia region of India, people living within one mile of an active uncontrolled coal fire were twice as likely to self-report health issues as people living in a similar village five miles from the fires. Nichols and Finkelman [2] noted that relatively few studies have addressed the many potential health problems that can be caused by the uncontrolled coal fires. They state that the reasons for this dearth of reporting include the following: (1) The vast majority of the people living near the uncontrolled coal fires are generally poor, live in rural areas and lack the financial and political wherewithal to demand attention. (2) The people impacted by these fires are up against the interests of the coal companies and government agencies who want the mines to continue generating income rather than invest in mitigation practices. (3) Uncontrolled coal fires are generally, though not exclusively, a problem of developing countries with limited resources and numerous other important issues to address. (4) The coal fires present difficult, often dangerous, nonglamorous working conditions that do not attract many researchers. (5) The health issues caused by long-term, low-level exposure to pollutants are not as dramatic as exotic zoonotic diseases. In addition, the collection and characterization of the emissions from these uncontrolled fires are challenging and the analyses of the gases are often not quantitative.

One approach to determine the composition and thus the potential environmental and health impacts of coal fire emissions is to collect and analyze the gaseous effluent. Pone et al. [4] found high concentrations of hazardous gases including benzene, toluene, ethylbenzene, xylene, methane, carbon monoxide, and carbon dioxide emitted from the Witbank and Sasolburg coalfields of South Africa. In addition to these gases, O'Keefe et al. [5] found mercury, hydrogen disulfide, carbonyl sulfide, carbon disulfide, numerous aliphatic hydrocarbons and tar in two coal fires in Kentucky, USA. Kruszewski [6], using Fourier-transform infrared (FTIR) spectroscopy on gases emanating from a burning coal waste heap in Poland, reported finding formaldoxime isocyanate, HON=CHNCO, ethenol, C<sub>2</sub>H<sub>3</sub>OH, hydrogen isocyanide, HNC, tetrafluoro-pxylylene, C<sub>6</sub>(CH<sub>3</sub>)<sub>2</sub>F<sub>4</sub>, 1-fluorocyclohexadienyl radical, C<sub>6</sub>H<sub>6</sub>F-perfluorinated p-benzyne,  $C_6F_4$ , 1,2,4-trioxolane,  $C_2H_3O_3$ , thioacetaldehyde, CH<sub>3</sub>CHS, thiocarbonyl fluoride, dithiop-benzoquinone, c-cyanomethanimine, bromomethane, peroxyethyl nitrate, triflic radical, CF<sub>3</sub>OSO<sub>3</sub>, and possibly a cyclic C10 molecule. The derivatives of freons include CHClF, HCFBr, CF<sub>2</sub> I+. Organo(semi)metallics include monomethylsilane, titanacyclopropene,  $CH_3MoH$  and  $CH_2MoH_2$ , and an indium–acetylene complex. It is safe to say that exposure to any of these compounds would not be beneficial. Collecting and analyzing the gaseous effluents is difficult and dangerous due to the risk of burns, collapse, and exposure to toxic gases, and requires special equipment.

Another approach to determine emissions from these fires is to collect and characterize the sublimates and condensates that have formed around the vent from which the gases escaped. Numerous publications have documented a wide range of chemical elements, minerals and other phases precipitating from gases released from uncontrolled coal fires. Publications describing the mineralogy and geochemistry of sublimates and condensates from global coal fires include those of Rost [7], Lapham et al. [8], Chessnokov and Shcherbakova [9], Matýsek and Jiráse [10], Zacek and Skaia, [11], Masalehadani et al. [12], Skaia et al. [13], Ciesielczuk et al. [14], and Witzke et al. [15]. Additional references can be found in Stracher et al.'s work [16]. Among the elements reported being mobilized from these fires are boron, fluorine, sulfur, chromium, manganese, nickel, copper, zinc, germanium, arsenic, selenium, bromine, chlorine, molybdenum, cadmium, tin, antimony, iodine, barium, mercury, lead, and bismuth. Most studies focused on the visible, brightly colored phases. Despite the many studies of the coal fire sublimates and condensates (see compilations by Stracher, [17,18]; and Stracher et al. [19]), we may be missing something. These valuable studies may be overlooking an important aspect of these uncontrolled coal fires. The obvious colorful, bizarre, spectacular phases illustrated in these publications may have distracted researchers from the true complexity of the process.

This complexity may only be found on close inspection on the micro scale but only a few studies have considered the role of nanoparticles (those measuring less  $0.1 \mu$ m) and ultra-fine particles. In their analysis of coal fires, Silva et al. [20] discussed nanominerals and ultra-fine particles in sublimates from the Ruth Mullins coal fire, Perry County, Eastern Kentucky, USA. A few years later, in their review of the mineralogical and chemical composition of nanoparticles associated with spontaneous coal fires, Silva et al. [21] stated that these particles are an integral part of coal fire emissions and recommended methods for their sampling and analysis. They noted that studying the influence of nanoparticles in areas of spontaneous coal combustion requires a multidisciplinary approach.

Though not strictly nanoparticles, many coal fire sublimates and condensate particles in the micrometer to sub-micron range are being released from the uncontrolled coal fires and present environmental and human health hazards [22]. In this study, we elected to analyze in depth a single small piece of a condensate from an active underground coal fire in the Jharia region of India. In this one particle, we found an extraordinary array of microminerals and other phases as well as potentially toxic elements that rival what other researchers have found in much more extensive collections.

These extremely small nanoparticles may present a health concern. Xia et al. [23] state that although mankind stands to obtain great benefit from nanotechnology, it is important to consider their potential health impacts. Although no human ailments have been ascribed to nanoparticles thus far, experimental studies indicate that nanoparticles could initiate adverse biological responses that can lead to toxicological outcomes. One of the principal mechanisms is the generation of reactive oxygen species and oxidant injury. Telianu et al. [24] state that although nanoparticles possess unique physicochemical properties that justify their broad use in applications, they can also manifest neurotoxic effects including oxidative stress, resulting in cell apoptosis and autophagy, immune responses, and neuroinflammation, which will affect the blood-brain barrier function. Warheit [25] stated that some evidence suggests that inhaled ultra-fine or nanoparticles, following deposition in the lung, largely escape alveolar macrophage surveillance and gain access to the pulmonary connective tissues, a potentially vulnerable anatomical compartment in relation to respiratory disease. Clearly, it is not only important to characterize the gases being emitted from the uncontrolled coal fires and the relatively large sublimates and condensates, but it is also equally important to determine the chemistry of the micro- and nanoparticles being emitted that may be impacting the health of the coal mine workers and nearby villagers.

We bring attention to this potentially important but often overlooked aspect of uncontrolled coal fires in the hope that future researchers will incorporate the characterization of the micro- and nanoparticles produced by this widespread and dangerous phenomenon.

## 2. Discussion

In 2019, a series of samples were collected from the vicinity of active uncontrolled coal fires in the Jharia region of India. Several samples were collected from Leuripathra, a village near Dhanbad in the state of Jharkhand, India. The samples included sublimates and condensates from active vents spewing gases from the active underground coal fires, coal, soil, surface water, and plants. An initial characterization of the samples revealed exceptional amounts of fluorine in the sublimates and condensates, as well as in the soil, water, and plants in the vicinity of the fires. Fluorine is both an essential element required for strong bones and teeth and an element that can lead to health problems if taken in excess of the bodies' requirements. Dental fluorosis is reported to affect hundreds of millions of people worldwide, with China and India having the largest number of people affected [26].

Following the initial survey of a dozen or so samples from four active coal fire locations in the region, we focused on condensate samples from Leuripathra. The samples were collected only meters away from occupied homes (Figure 1). We then focused our attention on a small (30 mm  $\times$  10 mm) piece of the sublimates and condensates (Figure 2) and were astounded to find more than 30 different phases in this one small piece (Figure 3, Table 1). The phases were identified using an JEOL JSM-IT100 scanning electron microscope with an energy dispersive X-ray detector. The 'Likely Mineral' identification was based on a composition similar to that of phases reported in other uncontrolled coal fire publications. Tentative mineral identifications (Likely Mineral in Table 1) were based on phases of similar composition reported from uncontrolled coal fires and/or from volcano fumaroles. In addition, the examination of broken fragments from this particle revealed multiple generations of sublimates and condensates. This small sample contained minerals composed of various combinations of fluorine, chlorine, iodine, selenium, lead, mercury, tin, bromine, cadmium, copper, boron, molybdenum, bismuth, iron, silicon, aluminum, magnesium, calcium, potassium, titanium sulfur, and ammonia. Many of these phases are no more than a few microns in size. It is entirely feasible that wind could dislodge and transport these micro- and nanosized particles, exposing mine workers, local villagers and people living downwind from the fires to this potentially harmful miasma.



**Figure 1.** Smoke being emitted from an underground coal fire in Levripathra, a village in the state of Jharkhand, India (near Jharia Town 23.7426° N, 86.4111° E).

The characterization of sublimates and condensates from the coal fires, taken from several locations in the Jharia region of India, revealed an abundance of fluorine-bearing minerals, nowhere greater than in the samples from Leuripathra. Fluorine in coal is generally around 100 ppm, but the fluorine in the coal samples collected from the region was about 450 ppm. The sublimates and condensates from one site averaged about 70,000 ppm F, with one sample from Leuripathra having 38 wt.% F. The plants in that region had as

much as 7000 ppm F in the leaves, while the soil samples had more than 300 ppm F, and the water samples had about 140 ppm F, all exceptionally high concentrations. Scanning electron microscopy/energy dispersive X-ray analysis (SEM/EDS) of the sublimates and condensates revealed abundant crystals of bararite and cryptohalite, both ammonium fluorosilicates [(NH<sub>4</sub>)<sub>4</sub>SiF<sub>6</sub>)], as well as unidentified iodine minerals (Figure 4), many containing fluorine. The ultimate source of the fluorine may be the relatively common fluorapatites in the coal (Figure 5). Also found in the coal was a zircon containing a nanometer-sized grain of platinum (Figure 6).



**Figure 2.** Photograph of coal fire condensates on the edges of a rock above a fissure emitting smoke from an underground coal fire adjacent to the village of Levripathra (near Jharia Town 23.7426° N, 86.4111° E).



Figure 3. Carbon-coated sample used for the scanning electron microscopy/energy dispersive X-ray analysis.

Chemistry	Likely Mineral	Geologic Setting of Mineral (C—Coal Fire Condensates) (F—Volcanic Fumaroles)
	Halides	
NH <sub>4</sub> Cl	Sal Ammoniac	C, F
NH <sub>4</sub> I		
NH <sub>4</sub> F		
BNH <sub>4</sub> Cl		
(NH <sub>4</sub> ) <sub>2</sub> SiF <sub>6</sub>	Cryptohalite/Bararite	C, F
NH <sub>4</sub> SiCl		
CuCl	Melanothallite/Eriochalcite	C, F
FeCl	Molysite	C, F
PbCl	Cotunnite	C, F
BiCl	Bismoclite/Daubreeite	
BiF	Gananite	
PbI(F,Cl)		
PbF(I)	Matlockite	F
CuCl(I)	Atacamite	C, F
AlF	Óskarssonite/Rosenergite	F
BiSiF		
BiSiCl		
CuF(Cl)	Claringbullite	
MoCdI		
	Sulfides/Sulfates/Selenides	
S	Sulfur	C, F
ISe		
AlSO <sub>4</sub>	Alunogen	C, F
BiMoSe		
Mo(Cd,Pb)S		
PbS(Cd)	Galena	C, F
CuS	Hydrocyanite/Dolerophanite	C, F
FeSO <sub>4</sub>	Guanajuatite	
BaSO <sub>4</sub>	Barite	C, F
ZnS	Sphalerite	C, F
NH <sub>4</sub> AsBi		
	Oxides	
FeO	Hematite	C, F
SiO	Tridymite/opal	C, F

 Table 1. Partial list of phases observed (those in bold are illustrated in the figures).



Figure 4. Bulbous bararite/cryptohalite covered by an unidentified iodine phase (bright coatings).



**Figure 5.** A relatively large fluorapatite crystal in a coal sample from the vicinity of the underground coal fire in Leuripathra.

Although uncontrolled coal fires in the Jharia region of India have been known for more than a century, we did not find any compilations of coal fire mineralogy from the region. The only published description of coal fire minerals that we found was a report [27] describing the initial discovery of cryptohalite and bararite on the surface of the ground above a burning coal seam in the Bararee colliery, Barari, Jharia, India.



**Figure 6.** The coal sample also contained a zircon with a nanometer-sized platinum grain (bright spot in the upper right).

Relatively large (~100 microns) crystals of salammoniac (NH<sub>4</sub>Cl) were also prevalent (Figure 7). Among the micrometer-sized sublimates were lead sulfide crystals (Figure 8), copper chloride, probably melanothallite or eriochalcite (Figure 9), and a bismuth chloride grain (Figure 10). Nichols et al. [1] reported that phases containing selenium, lead, mercury, tin, and bromine were also observed in the samples. Figures 11–13 illustrate other micronsized phases found in the Leuripathra samples.



Figure 7. Crystals of sal ammoniac on bararite/cryptohalite.



**Figure 8.** Micrometer-size lead sulfide crystals. The energy dispersive spectrum to the right indicates the presence of lead (Pb) and tin (Sn). The sulfur (S) peak is partially masked by the lead peak in the spot indicated in the lower image. The peaks for carbon (C), oxygen (O), fluorine (F), and chlorine (Cl) are due to the excitation of the adjacent phases.



**Figure 9.** A micrometer-sized copper chloride iodide sublimate. The energy dispersive spectrum to the right indicates the presence of copper (Cu), chlorine (Cl) and iodine (I). The peaks for carbon (C), oxygen (O), aluminum (Al), silicon (Si) and sulfur (S) are due to excitation of adjacent phases.



**Figure 10.** Bismuth chloride particle. The energy dispersive spectrum to the right indicates the presence of bismuth (Bi) and chlorine (Cl) in the spot indicated in the lower image. The peaks for silicon (Si), carbon (C), and oxygen (O) are due to excitation of adjacent phases. Fluorine (F) could be a minor component of the bismuth chloride phase or a component in an adjacent phase.



Figure 11. Silica overgrowth on bararite/cryptohalite.



**Figure 12.** Micron-sized barite crystal. The energy dispersive spectrum to the right indicates the presence of barium (Ba) and sulfur (S) in the spot indicated in the image. The peaks for carbon (C), oxygen (O), nitrogen (N), fluorine (F), aluminum (Al), and chlorine (Cl) are due to excitation of adjacent phases.



**Figure 13.** Micron-sized iron oxides. The energy dispersive spectrum to the right indicates the presence of major iron (Fe) in the spot indicated in the image. The peaks for carbon (C), oxygen (O), and silicon (Si) are due to excitation of the adjacent phases.

The small size of the particles, and their intimate intergrowths and volatility made further characterization difficult. Several attempts at X-ray diffraction resulted in a tentative identification of sal ammoniac, one of the more common and larger phases, in addition to a number of unidentified peaks. Attempts at Mossbauer spectroscopy were unsuccessful. While monitoring the energy dispersive X-ray analysis of the major iodine phase, perhaps ammonium iodide (bright coatings in Figure 4), the iodine concentration rapidly decreased as the electron beam volatilized the material. Several other phases were encountered, but, likely because of intimate intergrowths, we were unable to resolve the phases. These included the following phases: bismuth-molybdenum-cadmium-seleniumfluorine-chlorine; bismuth-cadmium-chlorine-iodine; molybdenum-cadmium-iodine; and arsenic-bismuth-nitrogen.

#### 3. Formation of the Sublimates: Thermodynamic Aspects

When a gas is vented to the atmosphere and expands against atmospheric pressure, it loses internal energy as its heat energy is transferred to heat sinks such as the atmosphere, rocks, sediment, and vegetation [28,29]. During this expansion, one or more minerals or amorphous solids may nucleate directly from the gas if the gas cools below the gas–liquid transformation temperature. This cooling process is called sublimation and the solids

that form are sublimates. As a sublimate cools and loses internal energy to a heat sink, it may transform into a chemically and stoichiometrically equivalent solid phase with a different crystal structure. Such polymorphs are called allotropes. An example of allotropes is the transformation of monoclinic ( $\beta$ ) to orthorhombic ( $\alpha$ ) sulfur (S<sub> $\beta$ </sub> -> S $\alpha$ ) as monoclinic sulfur cools.

By using the molar heat capacities of the allotropes of a sublimate and the molar enthalpies and entropies of transformation of those sublimates, it is possible to calculate and graph the pressures and temperatures (P-T) at which the component of a gas, such as one vented to the atmosphere, will transform to a sublimate and, conversely, the conditions of equilibrium. Stracher [30] applied the thermodynamic equivalent of Kirchhoff's Law to a closed thermodynamic cycle [31] in a four-step analytical procedure, called Thermodynamic Loop Analysis (TL Analysis), to derive the P-T equilibrium equation for the sublimation of sulfur from anthracite gas at the Centralia, Pennsylvania mine fire. The equation derived via TL Analysis is as follows:

ln [Ps<sub>2</sub>(g)] = 29.21–3.12 ×  $10^{-3}$ T–6686T <sup>-1</sup>–4750.8T<sup>-2</sup>–2.58 In(T), and the graph is illustrated in Figure 14. Graphs such as this are called Pressure–Temperature (P-T) stability diagrams because they indicate the partial gas pressure and temperature at which the vented gas component and the nucleated sublimate are stable, barring any other reactions involving the gas component or the sublimate with phases with which they come into contact. These graphs are useful for environmental studies because they indicate the pressures and temperatures at which a gas component and sublimate are stable.



**Figure 14.** Pressure–temperature stability diagram for orthorhombic sulfur and an  $S_2(g)$  component in coalfire gas. The partial pressure  $Ps_2(g) < 1$  atm. This diagram is applicable to all sulfur sublimating gas mixtures within this pressure range. Higher partial gas pressure corrections are possible. Modified from [32].

#### 4. Concluding Remarks

In a recent review of global coal use, the International Energy Agency [33] reported that global coal consumption was set at an all-time high in 2022 and that coal consumption is expected to stay at record levels despite declines in coal use in the United States and Europe. They note that, in 2022, coal consumption rose by more than 3 percent to 8.3 billion tons, with China, India, and Southeast Asian countries accounting for 75% of the coal that will be consumed worldwide in 2023. The increase in coal use will undoubtedly result in

additional uncontrolled coal fires with increased emissions of carbon dioxide, particulates, toxic trace elements, and nanoparticles, threatening the health of millions of the most vulnerable people in these regions.

It is evident that the health of the villagers living in proximity to the uncontrolled fires and the miners working in mines where the coal is burning is at risk from the release of noxious gases, toxic elements, and nanoparticles. The only solutions to this situation are to make a concerted effort to extinguish the fires and, if this is not feasible, relocate the villagers to a safer distance. We fully recognize that neither solution is ideal either economically or socially but allowing these situations to persist will likely lead to widespread, severe health problems.

It is obvious that uncontrolled coal fires release copious amounts of potentially toxic material, whether it is trace elements such as arsenic, selenium, mercury, and fluorine, gaseous compounds such as carbon monoxide, carbon dioxide, toluene, and benzene, or minute particles containing toxic elements or inducing oxidative stress. We urge researchers to look deeper into the condensates to identify the micro- and nanosized particles to help determine whether they might present an additional health threat to the communities living and working near the ubiquitous uncontrolled coal fires. There is no longer an excuse to miss the obvious and the subtle impacts of these uncontrolled coal fires.

Despite attempts to reduce carbon dioxide emissions, the mining and use of coal continues to increase, providing more opportunities for uncontrolled coal fires, the mobilization of micro- and nanoparticles, and their impacts on human health.

Sustainability requires that greater efforts be expended to prevent these wasteful and dangerous fires, as well as efforts to extinguish the coal fires that will inevitably occur.

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