

The role of atmospheric aerosol composition in climate change

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Abstract— The chemical composition of atmospheric aerosols has been investigated. Contributions of sulfate and soot in aerosols to the atmospheric extinction are studied. Discussions are made on the problems of aerosol emitted from volcano, forest fires in northern China, 1987 and oil field fires in Kuwait, 1991. It is indicated that the changes in concentration, particle size, and chemical composition of aerosol after those events could have impacts on the climate change either regionally or globally and that the impact of aerosol particles on climate change could compensate for some temperature increase caused by greenhouse gases and the increase of surface intensity of ultraviolet radiation due to ozone layer depletion.

Keywords: atmospheric aerosol; chemical composition; climate change.

In the atmosphere, besides huge amounts of oxygen and nitrogen, exist many kinds of other gases and particles in very low concentrations. All of those gases and particles are called as atmospheric trace component which includes atmospheric trace gases such as carbon dioxide and so on and atmospheric aerosol particles, or briefly aerosol. The climate of the Earth has the potential to be changed on all timescales by the way in which short-wave radiation from the Sun is scattered and absorbed, and thermal infrared radiation is absorbed and emitted by the Earth-atmospheric system. If the climate system is in equilibrium, then the absorbed solar energy is exactly balanced by radiation emitted to the space from the Earth and its atmosphere. But it has been well known that of particular relevance to concerns about climate at present are the changes of atmospheric trace components. The greenhouse effects of carbon dioxide, atmospheric trace gases and ozone depletion caused by nitrous oxide, CFCs and so on have drawn enormous attention and the effect of atmospheric concentration and chemical composition of aerosol on climate change is one of the hot points of discussions (Houghton, 1990).

Both solar and earth radiation energy pass through atmosphere and take it as the media of transport. While transporting through the atmosphere, the energy of different wavelength is hindered by different compounds in the atmosphere trace

component. This is characterized by the atmospheric extinction, which affects the solar energy distribution among the earth surface, near surface, and high altitude, and consequently, leads to the climate change.

The atmospheric extinction can be characterized by extinction coefficient (b_{ext}), which is a sum of light scattering coefficient (b_{scat}) and light absorption coefficient (b_{abs}) of the gases (g) and particulates (p) in the atmosphere.

$$b_{\text{ext}} = b_{\text{scat,g}} + b_{\text{abs,g}} + b_{\text{scat,p}} + b_{\text{abs,p}}.$$

The atmospheric extinction coefficient is determined by the light character of the individual atmospheric trace gases and aerosol of different chemical composition. It is also a sum of the light character of all individual compounds A_1, A_2, \dots, A_n in the atmosphere (Su, 1986):

$$b_{\text{ext}} = \sum_{i=1}^n a_i [A_i],$$

$[A_i]$ and a_i indicate concentration of a compound A_i in the atmosphere and the coefficient, respectively.

It has been proved that in the polluted atmosphere, the contribution of gases to atmospheric extinction is much less than that of aerosol. Here we mainly discuss the role of aerosol in atmospheric extinction. The chemical composition of atmospheric aerosol is very complicated and characterized mostly by the sources of aerosol and meteorological factors. Most of the chemical elements on the earth can be found in the aerosol according to the results of X-ray fluorescence analysis and proton induced X-ray emission analysis for typical aerosol. However, different chemical elements and compound in aerosol have their unique extinction character and, consequently, their effects on climate change are different, too. Field experiment and investigation in Beijing-Tianjin area show that sulfate, soot, water (relative humidity) and so on in the aerosol contribute to the increase of atmospheric extinction predominately (Su, 1986; Fig. 1). Aerosol nitrate originated from NO_x -rich sources may also be a compound increasing extinction.

Most of atmospheric sulfate in aerosol are the product of transformation of SO_2 , COS, $(\text{CH}_3)_2\text{S}$ and so on through homogeneous and heterogeneous reactions in the atmosphere. Sulfate in the aerosol and especially in cloud condensation nuclei is characterized by very high scattering effect in the atmosphere and, especially, in clouds. The extinction in clouds is mainly caused by light scattering of sulfate in condensation nuclei. Friedlander (Friedlander, 1977) indicated that sulfate contributed more to the extinction than its mass concentration alone would be. Based on the scattering-mass ratio (b_{scat}/p) for sulfate compounds and the concentrations measured

in a field experiment, sulfate contributed 17% of the aerosol mass and 33% of the scattering measured during the sampling intervals analyzed. That means the contribution of sulfate to the whole extinction effect of aerosol is about two times as much as the sulfate mass content in aerosol. Water helps the sulfate and other salts of aerosol in increases of scattering effect. Hence, high relative humidity increases the light scattering and gives a higher extinction coefficient than that of the same quantity of dry sulfate. Nitrate also have high light scattering effect but less than sulfate.

Soot, a carbon-containing black mixture in the atmosphere, is the other important extinction component of aerosol, which emits from incomplete combustion of mineral fuel and biomass burning. The light absorption effect of soot has been studied by oxidizing combustible components of aerosol sample and investigating its light transparency with a laboratory apparatus (Novakov, 1981; Su, 1987). The scheme of the thermal analysis is shown in Fig.2. The particulates sample, collected on a pre-heated quartz filter, is placed in the quartz combustion tube, in which the filter is perpendicular to the tube axis. To determine the relative transparency of aerosol on the filter we use a He-Ne laser beam which passes through the combustion tube and accordingly through the filter. The intensity of laser beam after the filter is monitored. The tube is supplied with purified oxygen/helium for oxidizing the carbonaceous material into CO_2 during the rise of temperature in the tube. Flue gas from the tube passes through a nondispersive infrared analyzer at a constant rate for determining the CO_2 concentration. The actual measurement consists of simultaneous monitoring both the CO_2 concentration in flue gas and the relative transparency of aerosol on the filter as a function of the temperature in the tube. A typical measurement is shown in Fig.3. The lower curve represents the CO_2 concentration, while the upper one corresponds to the relative transparency. Inspection of these curves shows that a sudden change in the relative transparency occurs concomitantly with the evolution of CO_2 peak at about 500°C . The light intensity, transparency, after the 500°C peak has evolved, corresponds to that of a blank filter. This demonstrates that the light-absorbing species in the aerosol sample are combustible carbonaceous, and are named graphitic carbon or black carbon. It contributes to the aerosol extinction by its light absorption

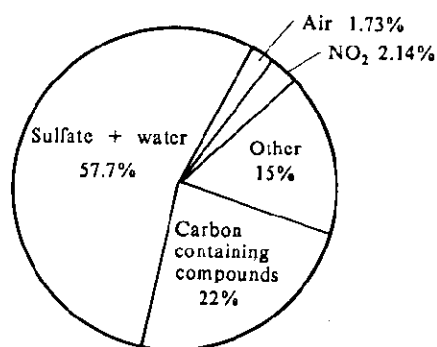


Fig. 1 The contribution of composition of aerosol to light extinction

property and makes soot an important role in atmospheric extinction and even cause regional climate change after a big fire.

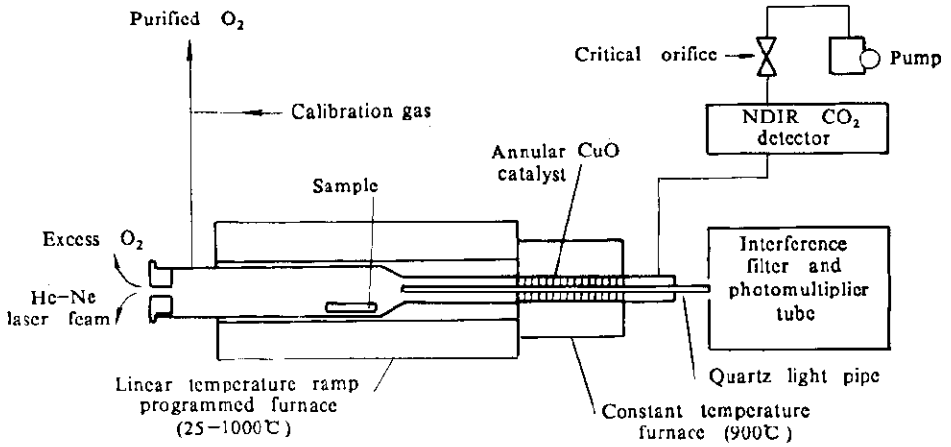


Fig.2 Schematic representation of thermal analysis apparatus

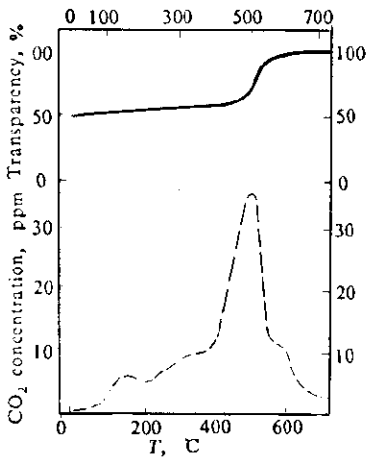


Fig. 3 Change of concentration of CO_2 in flue gas and relative transparency with temperature of aerosol oxidation

W/m^2 , indicating that they can have a significant climate impact on decadal time-scales.

Atmospheric aerosol comes mainly from different emission sources, such as volcanic eruption, forest fires, fossil fuel burning, and atmospheric reaction processes.

Volcanic eruption inject huge amount of aerosol and SO_2 to high altitude. SO_2 transforms to sulfate through chemical reaction in the air in 30–40 hours after eruption. Some studies (Mass, 1989) concluded that major volcanic events, of which there were only about five times during last century, may cause a global-mean cooling of 0.1 to 0.2 $^\circ\text{C}$ for an one to two year period after the event. A direct calculation of the radiative impact of major volcanic eruption (Ramanathan, 1988) shows that the decadal radiative forcing may be lower of 0.2–0.4

A big forest fire happened in the northeast of China (52° N, 122° E) from May 6 to June 2, 1987. This fire covered 5.2×10^4 km². Local visibility before the fire was about 20 km. It decreased very quickly during the fire, even down to 0.1 km. The photograph of smoke plume at 0:43 GMT May 8, 1987 taken from satellite is omitted. The altitude of plume was about 1000–1600 m along a distance of 50–140 km downwind to the fire and fluctuated between 700–1000 m after 160 km further (Xu, 1990). About a couple of days after the beginning of the fire at the tropopause above Mauna Loa (19.5° N, 155.6° E) and Laramie (41.3° N, 105.7° W), a new aerosol layer was detected (Sci. Event Alert Network Bulletin, 1987). On 29 May 1987 strong particle enrichment in troposphere above Laramie was detected, as shown in Fig.4. The number of aerosol particles ($d > 0.15 \mu$) at 10 km high increased to $10/\text{cm}^3$ compared with the normal average value of about $0.5/\text{cm}^3$. On 8 July, about 40 days after the fire, particle concentration in tropopause became smaller in about one-fifteenth of that on 29 May. This is an evidence of long distance transport of aerosol injecting to high altitude.

During the time of Kuwait raging oil fires in 1991 soot has been transported to and detected at the mountain top of Mauna Loa, Hawaii, where the concentration of soot in the atmosphere was about 20 times above normal levels (Tyson, 1991). Aerosol samples were collected at Kuwait during oil fires (March, 1991) and analyzed for determining their chemical composition (Su, 1987). The aerosol is characterized with very high soot concentration of more than 60% in aerosol and contains many kinds of organic compounds. Such black aerosol had the local visibility decreased to very low and moved well beyond the Middle East. It was predicted the heavy aerosol and thick clouds of smoke would trigger regional and even global climate change, as well as pollution. One model calculation shows that beneath the plume there is a severe reduction in daylight, and a daytime temperature drop of 10°C within 200 km of the source (Browning, 1991). Other model results show a decrease in surface air temperature of 4°C in the Gulf region (Bukan, 1991).

In the air above the industry developed regions, the aerosol concentration is

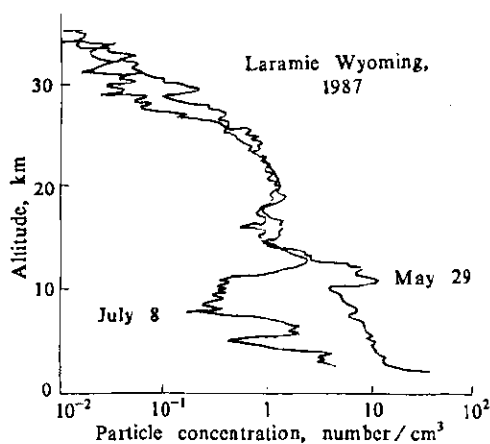


Fig.4 Plots of concentrations of particles with radius greater than 0.15 microns counted from balloons launched from Laramie, Wyoming on 29 May and 8 July 1987

relatively higher than that of other places and there exist more sulfate and soot in the air. For example, Ball and Robinson (Ball, 1982) have shown for the eastern U. S. an average annual depletion of solar irradiance of 7.5% at the surface. Some of this depleted radiation will, however, have been absorbed within the troposphere. If both the solar energy absorbed in the troposphere and that scattered to space are responded to one half of the average annual depletion of solar irradiance of 7.5% at the surface, for a daily mean surface irradiance of 200 W/m^2 , the change of it would be 7.5 W/m^2 and would have cooling effect. Measurement of total solar radiation in Beijing shows that radiation at the surface in the past decade decreased at the rate of 0.9% per year. Model calculation indicates that light extinction attributed to aerosol may account for the change trend (Wang 1992).

Increase in cloud condensation nuclei of industrial origin might explain why the Northern Hemisphere has not been warming as rapidly as the Southern Hemisphere over the last 50 years. Wigley (Wigley, 1989) estimated that each 0.1°C increase in twentieth century warming of Southern Hemisphere relative to the Northern Hemisphere correspond to a different of cloud condensation nuclei in about 10%, or a forcing differential of around -0.5 W/m^2 .

In summary, there is little doubt that major volcanic eruptions and raging forest or oil fires contribute to the interannual variability of global temperature record. There is no convincing evidence, however, of long time-scale effects. In the future such effects will continue to impose small year to year fluctuations on the global mean temperature. Furthermore, a period of sustained intense events could partially offset or delay the warming effect due to increased concentrations of greenhouse gases. It is also little doubt that increased of sulfate and soot in the atmosphere give some compensation to the warming effect by greenhouse gases. This compensation would not necessarily imply zero climate change. The sulfate and soot effects would tend to act only regionally, whilst the greenhouse effect is global. Hence, regional climate change would still be possible even if the global mean perturbation to the radiation balance were to be zero.

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