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Postprint on Source Apportionment and Control of Atmospheric Haze

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Abstract

With the acceleration of economic development and urbanization, the situation of atmospheric compound pollution in China has become increasingly severe since 2000. Satellite observations indicate that approximately 30% of China's territory and nearly 500 million people are affected by atmospheric compound pollution. Atmospheric haze pollution dominated by fine particles is particularly severe in the Bohai Rim, Yangtze River Delta, Pearl River Delta, Guanzhong, and Chengdu-Chongqing regions. Despite years of unremitting efforts by governments at all levels, by the launch of the special project in 2012, concentrations of primary pollutants such as PM₁₀ and SO₂ had begun to decrease, whereas those of secondary pollutants like PM_{2.5} and O₃ showed no significant reduction. Prior to the project's initiation, domestic research, though abundant, lacked systematic planning, resulting in incomplete, fragmented, and unsystematic studies that hindered a comprehensive understanding of China's haze pollution and the development of scientifically sound control strategies. Consequently, systematic research on haze source apportionment and control is imperative for scientifically addressing haze pollution. In response to the "Innovation 2020" initiative and the Academy's Twelfth Five-Year Plan, the Chinese Academy of Sciences (CAS) launched the project establishment and feasibility studies for the "Atmospheric Haze Source Apportionment and Control" special project in 2010. Following over 20 seminars and evaluations, the special project was formally approved in October 2012.

Full Text

Strategic Priority Research Programs (Category B) of the Chinese Academy of Sciences

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1. Background and Significance

With accelerating economic development and urbanization, atmospheric compound pollution in China has become increasingly severe since 2000. Satellite observations indicate that approximately 30% of China's land area and nearly 500 million people are affected by atmospheric compound pollution. The Bohai Rim, Yangtze River Delta, Pearl River Delta, Guanzhong, and Chengdu-Chongqing regions suffer particularly serious haze pollution characterized by fine particles. Governments at all levels have made unremitting efforts for many years. By the time the program launched in 2012, concentrations of primary pollutants such as PM10 and SO₂ had begun to decline, but secondary pollutants like PM_{2.5} and O₃ showed no significant reduction. Prior to the program, although numerous domestic studies existed, the lack of overall coordination led to incomplete, unsystematic, and "fragmented" research that could not comprehensively understand China's haze pollution or propose scientifically effective control strategies. Therefore, systematic research on haze tracing and control was urgently needed for scientific haze management. In accordance with the "Innovation 2020" initiative and the Academy's 12th Five-Year Plan, the Chinese Academy of Sciences initiated the demonstration and approval process for the "Tracing and Controlling Atmospheric Haze" program in 2010. After more than 20 discussions and evaluations, the program was formally approved in October 2012. The program established five projects: haze tracing simulation, atmospheric haze source identification, numerical modeling and cooperative control schemes, key monitoring technologies and equipment development, and advanced control technologies for key haze pollutants.

2.2 Overall Progress

Since its launch, the program has achieved remarkable progress in all research areas. By the end of 2015, researchers had published 396 papers in journals including *Nature Geoscience* and *Proceedings of the National Academy of Sciences*, among which 305 were SCI-indexed (61 with impact factors greater than 5). The program filed 87 invention patent applications (29 granted), 5 international PCT patents, 21 utility model patents (17 granted), registered 4 software copyrights, and published 1 monograph.

2.2.1 Haze Tracing Simulation

Based on the budget closure principle, the program designed comprehensive field observations for direct and accurate radical measurements. By establishing a reaction-rate-based numerical closure experimental method, researchers discovered a “non-traditional radical regeneration mechanism” in both daytime and nighttime data analyses from the Pearl River Delta and Beijing, revealing a universal property of China’s tropospheric oxidation capacity: OH radical concentrations in low-NO_x and high-VOCs regions have reached peak levels predictable by current theory (Figure 1 [Figure 1: see original paper]). This finding is significant for understanding the removal of primary pollutants and formation of secondary pollutants in China. Through field observations and laboratory simulations, the program revealed the synergistic mechanism by which coal smoke pollutants promote secondary particle formation from vehicle emissions and proposed a theory for explosive secondary particle growth under compound pollution conditions. For example, mineral oxides can provide reaction interfaces for coexisting NO₂ and SO₂ in the atmosphere, facilitating rapid conversion of sulfite to sulfate through activated molecular oxygen. This mechanism was validated in heavy haze field observations during spring 2013, providing important insights for developing coordinated emission reduction measures for Beijing’s air pollution. Smog chamber simulations demonstrated that the combined effect of high-concentration SO₂ from coal combustion and vehicle emissions significantly increases atmospheric fine particle loading, representing a major cause of frequent winter haze episodes in northern China.

2.2.2 Atmospheric Haze Source Tracing

Leveraging CERN (China Ecosystem Research Network) observation stations, the program established the “China Atmosphere Aerosol Observation Network (CARE-China)” (Figure 2 [Figure 2: see original paper]). Using unified instruments, experimental standards, and data protocols, this network obtained continuous mass concentration spectra of PM_{2.5}, PM₁₀, and TSP, as well as spatiotemporal distributions and variation patterns of gaseous pollutants (SO₂, NO_x, O₃, VOCs, and CO) and aerosol optical depth. For the first time, the program acquired a spatiotemporal dataset of aerosol chemical composition spectra (7 categories/100+ species across 9 size fractions) for different regions of China, filling a gap in Asian regional data. Source apportionment revealed significant differences in PM_{2.5} sources between heavily polluted and clean periods in Beijing. During clean periods, major sources were coal combustion, biomass burning, dust, and vehicle emissions. During heavy pollution episodes, the peak size of particle mass concentration and major chemical components shifted from 0.43–0.65 nm to 0.65–2.1 nm, with PM_{2.5} primarily originating from vehicles, coal combustion, industry, and dust. Local secondary pollutants were the main cause of elevated atmospheric particles. Therefore, reducing regional emissions during the early pollution stage and cutting local precursor emissions during peak pollution periods represent effective means for controlling Beijing’s air

pollution.

2.2.3 Numerical Modeling and Cooperative Control Schemes

Integrating field observations and laboratory simulations, the program developed a full-scale nested-grid air quality forecasting model system (NAQPMS). Using two-way nesting technology, the system achieved nested coupling calculations from global, transcontinental, regional, urban agglomeration, city, and street scales within a unified modeling framework, effectively improving forecast accuracy. Centered on NAQPMS, the program developed a multi-model ensemble forecasting system for atmospheric haze, achieving key breakthroughs: (1) overcoming challenges in model error estimation and multivariate collaborative assimilation of gaseous and aerosol pollutants, establishing an urban-scale atmospheric pollution source inversion system and atmospheric haze data assimilation system that reduced PM_{2.5} forecast errors by over 30% during heavy pollution periods; (2) addressing difficulties in estimating model input parameters and model uncertainties, combining multi-model simulation technology with Monte Carlo multivariate random ensemble perturbation techniques to achieve statistically integrated deterministic forecasting and stochastic probabilistic forecasting (Figure 3 [Figure 3: see original paper]). Based on multi-model ensemble forecasting technology, the program organized and launched the third phase of the Asian Air Quality Model Intercomparison Study (MICS-ASIA III), with over 20 models from Europe, America, China, Japan, and Korea participating, earning positive international recognition. As core technical support, the program established national and regional operational air pollution forecasting and early warning systems, which have been adopted as core forecasting technologies by the Ministry of Environmental Protection's National Environmental Quality Forecasting and Early Warning Center, regional centers for the Beijing-Tianjin-Hebei, Yangtze River Delta, and Pearl River Delta regions, and provincial-level centers in Jiangsu, Guangdong, Hebei, Beijing, Tianjin, Shanghai, Heilongjiang, Liaoning, Inner Mongolia, and other provinces and municipalities. The system supported air quality assurance for major events including the Beijing APEC Summit, the 70th Anniversary of Victory in the Anti-Japanese War military parade, and the Hangzhou G20 Summit, as well as heavy pollution emergency response, receiving public commendation from the Ministry of Environmental Protection and the China National Environmental Monitoring Centre.

2.2.4 Key Monitoring Technologies and Equipment Development

Based on field observation and laboratory research needs, the program focused on developing online measurement technologies and equipment for atmospheric oxidants (HO_x and NO₃ radicals), solving problems such as sampling losses and water vapor absorption nonlinear interference in spectral inversion, with radical detection indicators reaching international advanced levels. The program independently developed atmospheric integrating nephelometers, cavity

ring-down spectrometers, hygroscopic tandem differential mobility analyzers, and vacuum ultraviolet photoionization VOC mass spectrometers, constructing a comprehensive characterization and research platform for atmospheric fine particle physicochemical properties. The program developed light-scattering time-of-flight (TOF) dynamic particle sizing and scanning differential mobility (SMPS+CPC) technologies, enabling online measurement of particle size distributions across a wide range of 5 nm–20 μ m. An integrated method combining thermal decomposition with reflectance and transmittance measurements was proposed for selecting organic carbon/elemental carbon (OC/EC) split points, with key technical indicators reaching international advanced levels. Multiple key technologies for lidar detection were breakthrough, including independently developed atmospheric fine particle detection lidar. An innovative complete set of high-sensitivity detection technology engineering solutions for atmospheric fine particles was designed, constructing an online monitoring system for atmospheric fine particles (Figure 4 [Figure 4: see original paper]), providing important technical support for national environmental monitoring capacity building.

2.2.5 Advanced Control Technologies for Key Haze Pollutants

The program established pilot-scale flow and cleaning platforms for electrostatic-fabric integrated precipitators, completed studies on electric field and flue gas characteristics' effects on electrocoagulation, optimized internal component structures and processes for demonstration projects, achieving electrostatic-fabric precipitator efficiency >99.98%. An industrial exhaust catalytic purification demonstration unit was built with 22,000 m³/h airflow and 450 mg/m³ VOCs concentration, passing third-party testing standards. A research platform for VOCs regenerative (catalytic) combustion was established, preparing monolithic catalysts with commercial promotion value and completing pilot production line construction with operational performance meeting design requirements. A novel domestic coal co-combustion technology using bituminous coal as fuel was developed (Figure 5 [Figure 5: see original paper]), avoiding reductive flue gas formation, with emission factors for PM_{2.5}, CO, and VOCs more than one order of magnitude lower than traditional coal stoves and combustion efficiency (based on CO₂ emission factors) improved by over 35%.

3. Originality

The program adopted two technical routes—"tracing causes" and "tracing sources"—addressing two core issues of regional haze formation and control in China. Using PM_{2.5} as a bridge connecting haze pollution phenomena and pollutant emissions, the program conducted bidirectional closed-loop studies on cause tracing and source identification. This research approach features distinctive characteristics while simultaneously addressing key scientific questions and meeting national strategic needs.

3.2 Innovation of Research Results

For the first time, smog chamber experiments were used to evaluate the secondary particle formation potential of gasoline and diesel vehicle emissions in China, estimating the combined primary and secondary contributions of on-road vehicles to PM_{2.5}. The synergistic effect of SO₂ and vehicle exhaust in secondary particle formation was investigated, contributing to understanding the explosive growth mechanism of secondary fine particles under China's atmospheric compound pollution conditions.

4. Significance for Industry

Independently developed detection equipment such as atmospheric fine particle lidar and aerosol mass spectrometers have been partially industrialized. For example, atmospheric fine particle mass concentration monitoring equipment has been installed at over 2,000 environmental and meteorological monitoring stations in more than 20 provinces and cities including Anhui and Jiangsu over the past five years, with batch applications in operational PM_{2.5} monitoring. Atmospheric fine particle lidar has sold over 60 units, capturing more than 70% market share. Combined sales revenue for these two products exceeds 500 million RMB. The developed aerosol mass spectrometer sold 19 units in 2015 alone, with cumulative sales revenue exceeding 100 million RMB, and has entered European and American markets. These achievements have changed the situation where China's environmental and meteorological monitoring equipment heavily relied on imports, driving related industrial development and generating significant economic and social benefits.

The developed electrostatic-fabric integrated precipitator technology has been demonstrated at the China Power Investment Henan Pingdingshan Branch's 1,030 MW supercritical generating unit, processing 9.8×10^6 m³/h of flue gas with output value of 90 million RMB. Various novel catalytic materials for VOCs degradation were developed, successfully establishing a production line with annual capacity of 100,000 liters of monolithic catalysts and completing over a dozen VOCs treatment demonstration projects including Tianjin Toyo Ink Co., Ltd. (20,000 Nm³/h airflow) and Dongguan Aozhong Electronic Materials Co., Ltd. (25,000 Nm³/h airflow), generating 30 million RMB in output value. These results have effectively promoted the development of industrial flue gas and other key haze pollution source control industries in China.

5. Recommendations for Future Development in Disciplines, Industry, and Talent Cultivation

The program has achieved a series of important results on key challenges including atmospheric oxidant capacity, secondary particle formation, and haze forecasting and early warning, leading domestic and international research on China's haze pollution. These accomplishments have laid the foundation for the National Natural Science Foundation of China's major research plan "Causes

and Response Mechanisms of Atmospheric Compound Pollution in China” and the Ministry of Science and Technology’s “Key Special Project for Air Pollution Prevention and Control Science and Technology.” Given the complexity of haze formation and the urgent need for scientifically sound coordinated emission reduction strategies, we recommend continued strengthening of air pollution prevention and control capacity building, particularly accelerating preparatory work for large-scale infrastructure such as the “Atmospheric Environmental Simulation System,” “High Tower System,” and “Urban Agglomeration Atmospheric Environmental Optical Monitoring System.”

While some advanced instruments and equipment developed by the program have been industrialized, further efforts are needed to promote industrial applications and increase the market share of domestic instruments. We recommend strengthening top-level design to facilitate integration of the program’s air pollution control technologies with government and enterprise pollution control practices, and promptly launching the national “Clean Diesel Engine Action Plan” to gradually implement ultra-low emissions in non-power industries, thereby controlling air pollution while driving related industrial development.

The program has achieved remarkable results in talent cultivation and research team building. Future efforts should continue to strengthen the cultivation of high-level innovative talents and young backbone personnel in air pollution prevention and control, implementing the national innovation talent promotion plan.

(Supporting Institution: Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences)

Note: Figure translations are in progress. See original paper for figures.

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