

Long-term observation of air pollution-weather/climate interactions at the SORPES station: a review and outlook

Aijun Ding (✉)^{1,2,3}, Wei Nie^{1,2,3}, Xin Huang^{1,2,3}, Xuguang Chi^{1,2,3}, Jianning Sun^{1,2,3}, Veli-Matti Kerminen⁴, Zheng Xu^{1,2,3}, Weidong Guo^{1,2,3}, Tuukka Petäjä^{1,4}, Xiuqun Yang^{1,2,3}, Markku Kulmala⁴, Congbin Fu^{1,2,3}

1 Joint International Research Laboratory of Atmospheric and Earth System Sciences (JirLATEST), Nanjing University, Nanjing 210023, China

2 Institute for Climate and Global Change Research, School of Atmospheric Sciences, Nanjing University, Nanjing 210023, China

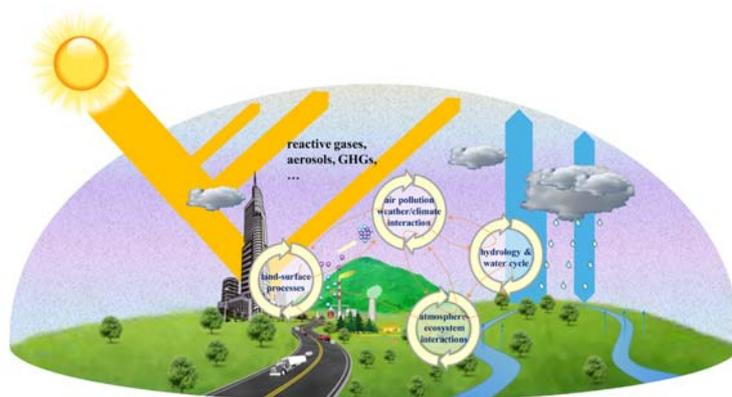
3 Collaborative Innovation Center of Climate Change, Jiangsu Province, Nanjing 210023, China

4 Department of Physics, University of Helsinki, Helsinki 00014, Finland

HIGHLIGHTS

- The concept design and detailed information of the SORPES station are introduced.
- Main scientific findings based 5-year measurements at the station are summarized.
- The future outlook of the development plan and its implications are discussed.
- The results improved understanding of interaction of physical and chemical processes.
- More SORPES-type stations are needed for different regions in China and the world.

GRAPHIC ABSTRACT



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ABSTRACT

This work presents an overall introduction to the Station for Observing Regional Processes of the Earth System – SORPES in Nanjing, East China, and gives an overview about main scientific findings in studies of air pollution-weather/climate interactions obtained since 2011. The main results summarized in this paper include overall characteristics of trace gases and aerosols, chemical transformation mechanisms for secondary pollutants like O_3 , HONO and secondary inorganic aerosols, and the air pollution – weather/climate interactions and feedbacks in mixed air pollution plumes from sources like fossil fuel combustion, biomass burning and dust storms. The future outlook of the development plan on instrumentation, networking and data-sharing for the SORPES station is also discussed.

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1 Introduction

China has been facing a great challenge in tackling air pollution issues in recent decades. Because of dramatically increasing energy consumption associated with rapid urbanization and industrialization, many regions of China, especially megacities and city clusters in the coastal eastern China, have experienced heavy primary

and secondary air pollution [1–7]. Some pollutants, such as nitrogen oxides (NO_x) and ozone (O_3), have shown increasing trends [8–13], with more frequent extreme pollution episodes [9,14–16]. Fine particulate matter, $PM_{2.5}$ (particles of 2.5 microns or less in aerodynamic diameter in the ambient air) often show extremely high concentrations in megacities during the winter haze period [4,5,16,17], although an overall decreasing trend has been observed during the recent years [18–20]. Such high concentrations of air pollutants will certainly cause negative impact on human health, plant growth and economy development [21–25].

✉ Corresponding author
E-mail: dingaj@nju.edu.cn

Even though many efforts have been made by scientists and the government during the recent decade, air pollution control in China is still facing some important challenges, especially understanding of the sources, chemical formation mechanisms, and transport and dispersion processes [2,4,5,26,27]. A relatively poor understanding of these processes/mechanisms makes it difficult to implement them into numerical models, which are the main tools for predicting air quality, for making adequate early warning or emergency control systems for long-lasting extreme pollution episodes, or for making long-term emission control policy [17,26,28]. Kulmala [24] pointed out the main challenges in improving current understanding of the complex air pollution situation in China, and Kulmala et al. [27] summarized the main knowledge gaps in our understanding of secondary new particle formation. Both of the above works strongly underlined the need of long-term measurements using ground-based stations in the quest for solving these grand challenges.

Ground-based field measurement is a widely-used means for understanding the characteristics and mechanisms of air pollution, in addition to which they provide observational data for model evaluation. However, the observed variations of air pollutant concentrations are always a combined result of complex physical and chemical processes [29], making it difficult to understand the exact mechanisms with a limited number of measurement components. For example, chemical processes, including those taking place in the gas and aqueous phases, as well as heterogeneous reactions, play key roles in the transformation of secondary pollutants along the transport pathways [30,31]. Furthermore, multi-scale meteorological processes such as advection, convection and turbulent dispersion in the planetary boundary layer (PBL) affect the formation and fate of haze episodes [29,32,33]. Synoptic weather contributes mainly to the multi-day (~5–7 days) cycle of air pollution [5,32,33], whereas PBL dynamics controls the diurnal cycle of air pollutants and constrains the vertical mixing and ventilation of air pollutants emitted from the ground surface [17,34,35]. In China, because of high concentration of absorbing aerosols, such as black carbon (BC) and dust [17,36–39], more and more evidences have been found that the interaction between the air pollution and PBL meteorology plays an important role in enhancing air pollution and in modifying weather and regional climate [16,17,26,28,35,40–42]. Therefore, to gain a comprehensive understanding on the physical and chemical processes/mechanisms of different air pollution cases in a specific region, synthesis and concurrent comprehensive measurements of air quality and boundary layer meteorology are needed.

In this paper, we present an overview of and introduction to the first stage of an integrated measurement platform, Station for Observing Regional Processes of the Earth System – (SORPES), as an example for long-term

measurements of air quality and boundary layer meteorology, in the Yangtze River Delta (YRD) of East China, one of the most polluted regions in China with complex pollution sources, strong solar radiation and humid climate. We introduce the overall design concept, detailed measurement parameters and instruments, and summarize the main results observed so far at this station. Specially, we demonstrate how we comprehensively understand the physical and chemical mechanisms in this unique atmospheric environment based on integrated field measurements, various data analysis methods and numerical modeling techniques.

2 Description of the station and instruments

2.1 Overall concept of the SORPES station

The Station for Observing Regional Processes of the Earth System (SORPES) is a cross-disciplinary research and experiment platform developed by Nanjing University in collaboration with University of Helsinki [16]. It was designed to be developed into a SMEAR (Station for Measuring Ecosystem-Atmosphere Relations) type “flagship” station according to the concept of Hari et al. [43,44]. Compared to the boreal forest SMEAR II station in Finland [45], the SORPES station focuses more on the impact of human activities on the environmental and climate system in the rapidly urbanized and industrialized eastern China region under the influence of monsoon climate [46]. The main scientific themes of the SORPES include land surface-atmosphere interactions, air pollution-weather/climate interactions, ecosystem-atmosphere interactions and hydrological cycle, as well as linkages between these associated processes (see Fig. 1). SORPES has also been considered as a part of the PEEX (Pan–Eurasian Experiment) infrastructure [47].

The geographical location of Nanjing makes it an ideal place to study the unique atmospheric chemistry and its interactions with physical processes for a complex mixture of different air masses from a regional scale. Nanjing sits on the southern tip of the most polluted East and North China Plain (see Fig. 2(a)). It can capture regional plumes, carried by the continental winter monsoon, from intensive fossil fuel combustion sources, dust source areas and biomass burning activities in the north. Under the impact of summer monsoon in the warm season, the site is generally downwind from the sub-tropical forest region, with strong biogenic emissions, in the South China (Fig. 2(a)). However, from a synoptic-scale perspective, air masses arriving at Nanjing are alternatively originated from different source regions associated with moving cyclones or anti-cyclones [16].

Nanjing is one of the most western cities in the Yangtze River Delta (YRD) region, with many cities clustered



Fig. 1 Concept and key scientific themes for the SORPES station. Note: The yellow and blue arrows show the radiative transfer of shortwave and long-wave radiation in the atmosphere, respectively

along the Nanjing to Shanghai axis. Under the north-east and south-east prevailing winds in winter and summer, Nanjing is generally downwind from the regional and sub-regional scale anthropogenic sources (Figs. 2 and 3). So, it is an ideal location for measuring regional air masses and for studying the chemical and physical processes of air pollutants after they have experienced sufficient evolution, mixing and chemical transformation in the atmosphere. To capture properly the region-scale characteristics, we set up the “flagship” central site of SORPES in Xianlin (118°57' 10"E, 32°07'14"N), suburban area of Nanjing with 20 km north-east from Nanjing downtown, and few outlying “satellite” sites in the city and the vicinity (Fig. 2(c)), including an urban air quality site, Gulou, an urban flux site, Dangxiao, and a countryside grassland flux site, Lishui [50]. The central “flagship” site is heavily instrumented, with measurements mainly focusing on air quality and boundary layer meteorology since 2011. Currently, the “satellite” sites have only rather limited numbers of instruments because they are aiming at specific purposes, such as a comparison of different land cover types etc. All these sites are undergoing further upgrade and integration to establish a network. However, in this review paper we mainly focus on results obtained at the SORPES Xianlin “flagship” site. We call the Xianlin site as the SORPES station in the following text.

2.2 Measurement of atmospheric compositions at the SORPES station

The SORPES station started in 2011 with three measurement modules of atmospheric chemistry-related compounds (aerosol and trace gases measurements) in a

2-floor building, measurements of meteorological variables in a 50 m × 50 m observation field, and flux measurements with a 75-m-high tower on a hill about 40 m above the ground level.

All the instruments of trace gas and aerosol concentrations are housed on the top floor of a laboratory building. The instrument details are listed in Table S1 in the Supporting Materials. Briefly, the routinely-measurable trace gases like O₃, sulfur dioxide (SO₂), carbon monoxide (CO), NO_x=NO + NO₂, carbon dioxide (CO₂), total reactive nitrogen oxides NO_y (= NO + NO₂ + PAN + HNO₃ + NO₃⁻ + organonitrates + HONO + 2N₂O₅...) are measured by the online analyzers with the time resolution of 1–5 min (Thermo Fisher Scientific 49i, 43i, 48i, 42i, 420i, 42i-Y, respectively, USA) [16]. The HONO concentration is measured by a long absorption photometer (QUMA, LOPAP-03, Germany) [51]. The isotope δ(¹⁸O) and δ (D) of water vapor is measured by a Cavity ring-down spectroscopy (Picarro, L2120-i, USA) [52]. Meanwhile, another Cavity ring-down spectroscopy is used for the measurement of greenhouse gas including CO₂ and CH₄ (Picarro, G2301, USA).

The PM_{2.5} mass concentration is measured by the online analyzer based on the light scattering and beta ray absorption method (Thermo Fisher Scientific, 5030SHARP, USA). The chemical components of PM_{2.5}, including the water-soluble inorganic ions, organic carbon and element carbon (OC/EC), and heavy metal elements are detected by the instrument for Measuring AeRosols and GAses (MARGA, Metrohm, Switzerland) [53], semi-continuous OC/EC analyzer (Sunset, RT-4, USA) and an online X-ray fluorescence heavy metal analyzer (Skyray, EHM-X200, China), respectively. As the primary

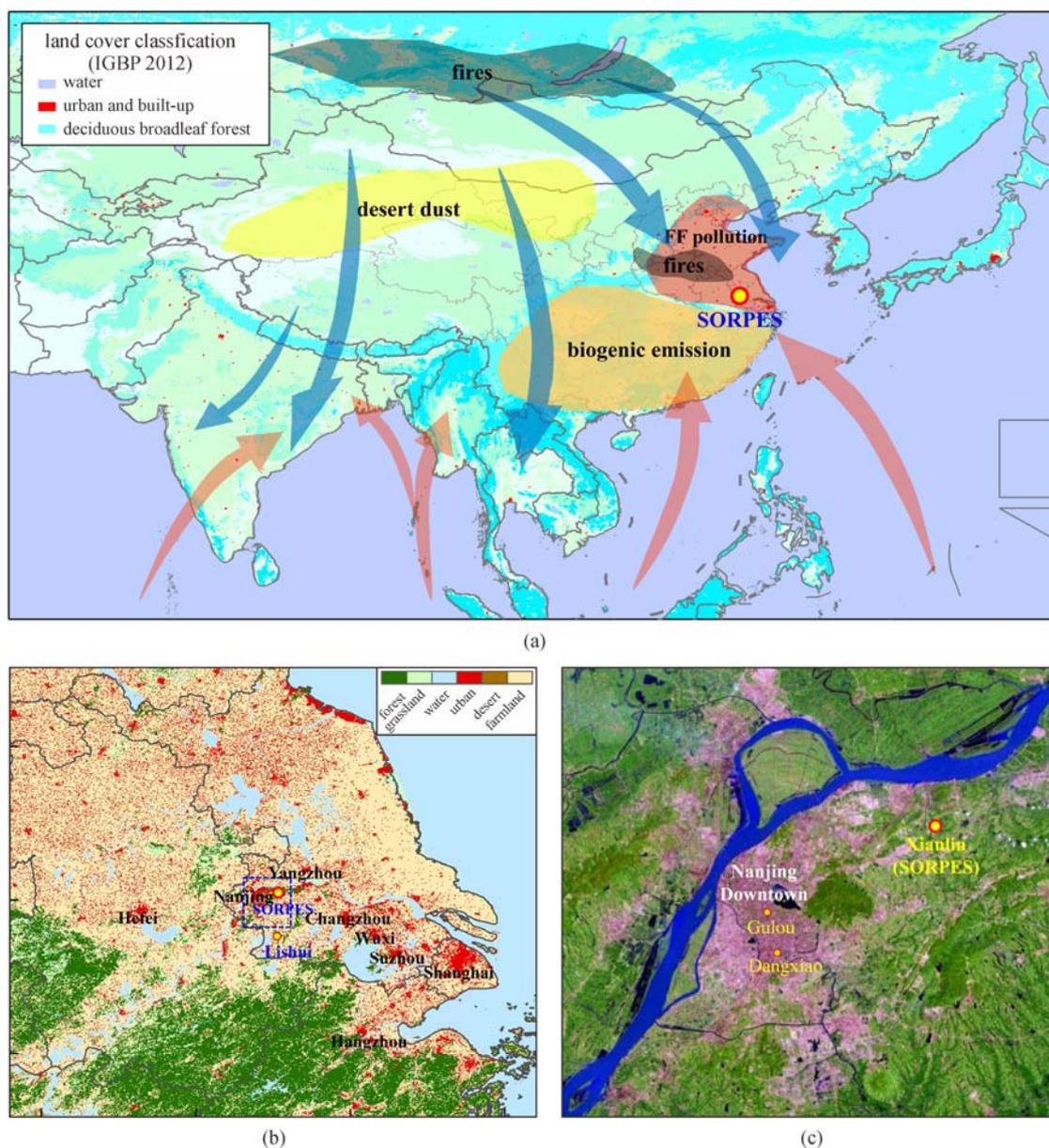


Fig. 2 Maps showing the location and geographical representatives of the SORPES station from different scales: (a) the East Asian monsoon region; (b) Yangtze River Delta region and surrounding regions, (c) Nanjing downtown and suburban areas

parameter of aerosol optical properties, the absorption and scattering efficiency of aerosols are measured by the Photoacoustic Extinctionmeter (DMT, PAX870, USA) and nephelometer (Ecotech, Aurora-3000, Australia), respectively. The size distribution of large particles ($0.5\text{--}20\ \mu\text{m}$) is measured with Aerodynamic Particle Sizer (TSI, APS-3321, USA) and that of submicron particles ($D_{p_{6-800\ \text{nm}}}$, $D_{p_{1-3\ \text{nm}}}$) with a DMPS (differential mobility particle sizer [54]) and PSM (particle size magnifier [55]) constructed at the University of Helsinki [56]. The air ion mobility distribution is measured by the Air Ion Spectrometer (AIS) constructed by the University of Tartu, Estonia [57,58].

2.3 Measurement of PBL meteorology and vertical profiles of air pollutants

The SORPES meteorological observation field is equipped with a suite of standard meteorological sensors (Table S2 in the Supporting Materials) to continuously measure soil heat flux (Soil Heat Flux Plate, HFP01, Campbell, USA), soil water content (Water Content Reflectometer, CS616, Campbell, USA), incoming and outgoing short- and long-wave radiation (4 component Net radiometer, CNR4, Campbell, USA), and basic weather-related variables such as the rain fall, relative humidity (RH), air temperature,

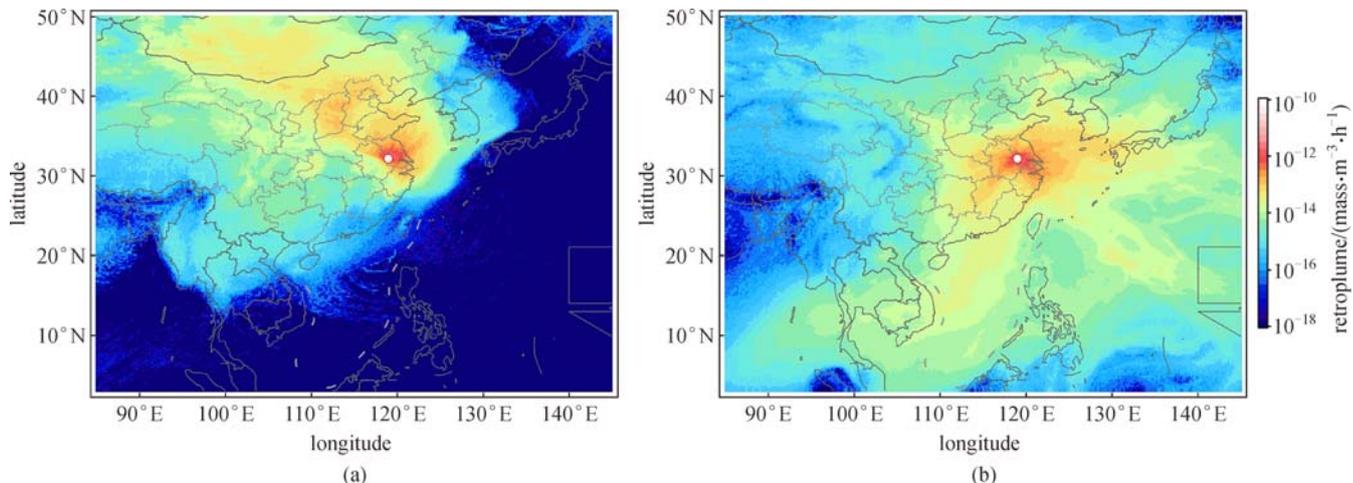


Fig. 3 Averaged Lagrangian retroplume of the SORPES station for (a) winter and (b) summer. Note: “Retroplume” represents the distribution of probability or residence time of a simulated air mass. These results were calculated using HYSPLIT [48] based on the method developed by Ding et al. [49]. (Modified from Ding et al. [16])

atmospheric pressure, and wind speed and direction (weather station; GRWS100, Campbell, USA).

Fluxes of surface sensible/latent heat and $\text{H}_2\text{O}/\text{CO}_2$ fluxes are measured using the eddy covariance (EC) technique [59]. Three-dimensional wind and temperature fluctuations are measured by CSAT3A sonic anemometers at the altitude of 2.3 m above ground level (a.g.l.) as well as at 25 and 50-m altitudes on the 75-m tower (see Table S2 in the Supporting Materials). CO_2 and H_2O fluctuations are detected using two fast-response, open-path, mid-infrared $\text{CO}_2/\text{H}_2\text{O}$ Gas Analyzers (EC150, Campbell, USA) installed close to the sonic path. The high frequency signal is recorded by a CR3000 micro-logger. In addition to the eddy-covariance systems, the wind speed, wind direction, temperature and RH are measured by sensors installed at the altitudes of 4 m, 9 m, 18 m, 36 m and 72 m a.g.l on the 75 m tower. The wind speed and direction are measured by five anemometers, each including a 010C wind speed sensor (Campbell, USA) and a 020C wind direction sensor (Campbell, USA). The temperature and RH are measured by a HMP155A Temperature and Relative Humidity Probe (Campbell, USA), which are capable of monitoring RH over the range of 0 to 100% and temperature over the range of -80°C to $+60^\circ\text{C}$.

A Micro-Pulse Lidar (MPL) system (Sigmaspace, MPL-4B-527, USA) is used to measure the vertical structure of aerosol and/or cloud backscatter coefficients during intensive campaigns. The laser light source is a diode-pumped frequency doubled solid-state laser (Nd-YLF at 527 nm, USA), yielding pulsed visible green light, with the vertical resolution of 15 m and the lowest sounding height of 255 m. In addition to the fixed ground base sensing instruments, a vehicle equipped with a boundary-layer radar wind profiler (Airda 3000M, China) is used for mobile measurement of real-time 3D wind field

information in the boundary layer. The profiler can compute averaged wind profiles for periods ranging from 6 min to an hour. The maximum detected height of this radar is 3 km.

To measure the vertical structure of trace gases, aerosols and boundary-layer meteorological parameters, tagged balloon measurements are conducted for specific synoptic processes during intensive campaign periods. The tagged balloon (a volume of 10 m^3) has a maximum loading capacity of 2–3 kg, when inflated with helium gas, capable of carrying instruments up to a 2.3-km altitude at an ascending speed of a few meters per second under the atmospheric conditions with wind speeds lower than $5 \text{ m} \cdot \text{s}^{-1}$. It usually takes about 30 min for the ascending or descending profile measurement. The instruments mounted on the tagged balloon include a portable Aethalometer (Aethlabs, AE51, USA) for BC, a personal aerosol monitor (TSI, AM510, USA) for $\text{PM}_{2.5}$ mass concentration, and a personal ozone monitor (2B Technology, POM, USA) for O_3 .

3 Overall results and key findings obtained at the SORPES station

Main trace gases and aerosol instruments have been continuously run at the SORPES station since July 2011 and some other advanced instruments have been step-by-step added in the following few years. So far, more than 5 years of high-quality data sets have been recorded. Based on these data sets, a series of studies have been conducted to understand the general characteristics of main chemical and physical parameters, key chemical formation mechanisms, and interactions between physical and chemical processes. Table 1 summarizes the main research topics,

methods and data period as well as the main findings and highlights of these studies. Below we give a summary about the main results and key findings:

3.1 Overall characteristics of trace gases and aerosols

With the continuous measurement data, several works have been conducted to investigate the overall characteristics of trace gases, aerosols and boundary layer meteorology in this region. Ding et al. [16] published the first paper to briefly introduce the SORPES station and presented an overview of 1-year measurements of O_3 , $PM_{2.5}$ and related trace gases at the SORPES station. That study found that O_3 and $PM_{2.5}$ showed strong seasonal cycles and peaked in warm and cold seasons, respectively, and that pollution episodes of both the pollutants were generally associated with an air mass transport pathway over the city clusters in the YRD under specific synoptic weather conditions. That work also reported that agricultural straw burning contributes to the intense $PM_{2.5}$ and O_3 pollution from the end of May to Mid-June, the main harvest season of

wheat in East China. For ozone formation, based on a correlation analysis of CO - NO_x - O_3 (Figs. 4(a) and 4(b)), that study pointed out a VOCs (volatile organic compounds) - sensitive regime for this region and suggested the need of stronger reduction of VOCs compared with NO_x for the mitigation of O_3 pollution and secondary PM formation in the YRD region in warm seasons [16]. That study also suggested the cross-regional efforts in controlling O_3 and $PM_{2.5}$ based on Lagrangian backward dispersion modeling for episode days of exceedance of the two pollutants recorded at the SORPES station (Figs. 4(c) and 4(d)).

Herrmann et al. [60] reported the first air ion and aerosol size distribution measurement at the SORPES station. Based on 4-month measurement data, that work established a simplified empirical criterion for estimation of the NPF probability in the study region. They also reported high ion cluster concentrations during the nights before early morning of the NPF event days, and suggested that ion-induced nucleation plays only a marginal role [60]. Qi et al. [56] made a comprehensive analysis on the NPF and

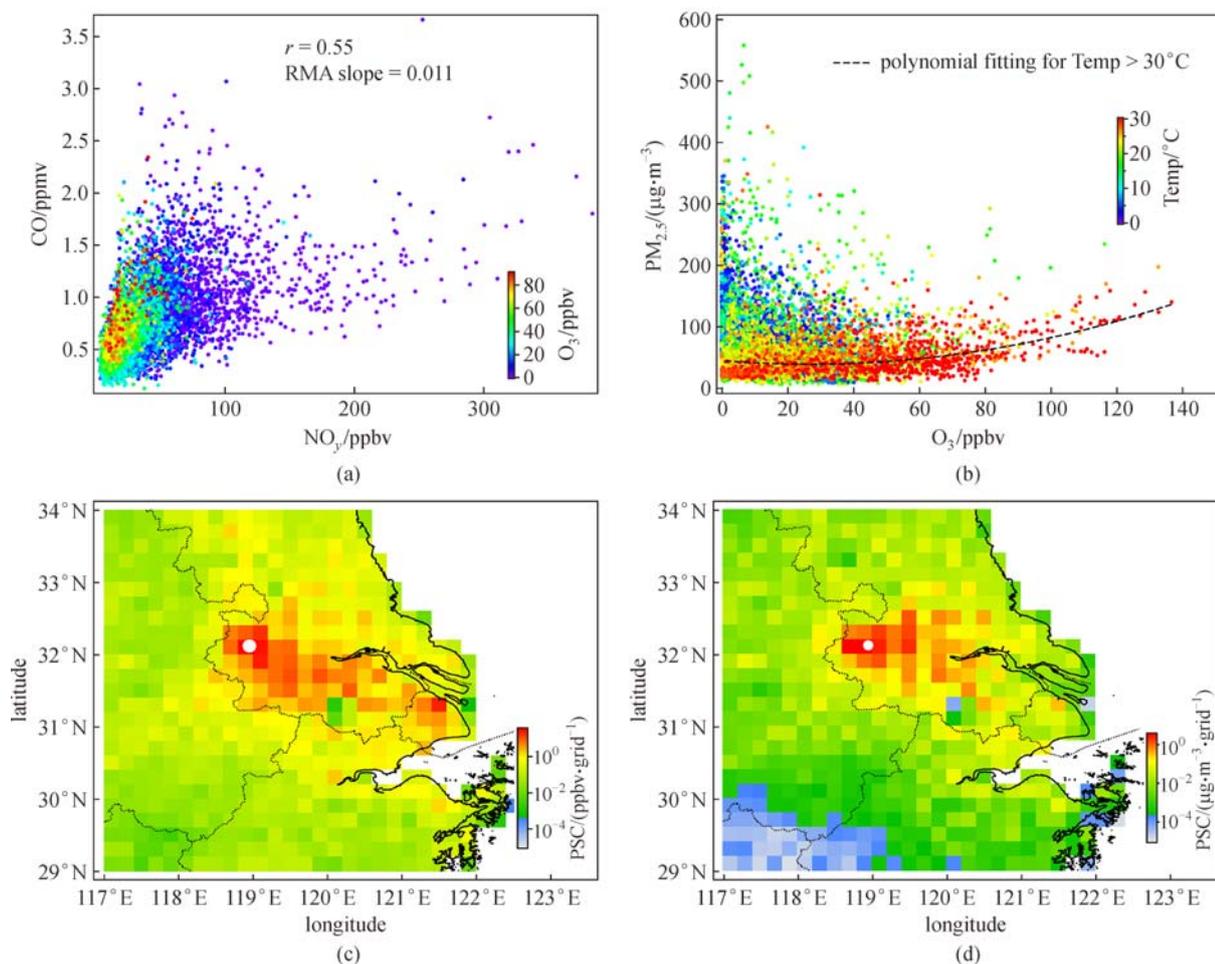


Fig. 4 (a) Scatter plots of CO - NO_y color-coded with O_3 concentration and (b) $PM_{2.5}$ - O_3 color-coded with air temperature; averaged distribution of potential source contribution (c) of CO for O_3 episode days and (d) for $PM_{2.5}$ episode days. (Modified from Ding et al. [16])

particle growth based on two years of DMPS measurement at the station. The averaged number concentrations of particles in the nucleation, Aitken and accumulation modes were about 5300, 8000, 5800 cm^{-3} , respectively, which are comparable to those at urban/suburban sites in China but 10 times higher than in remote areas. During the 2 years of measurement (2011–2013), 44% of the whole sampling days were identified as NPF event days. NPF and growth rate also showed a strong year-to-year difference associated with different long-range transport pathways and solar radiation levels, but generally the NPF rate peaked in spring ($3.6 \text{ cm}^{-3} \cdot \text{s}^{-1}$) while the growth rate was the highest in summer ($12.8 \text{ nm} \cdot \text{h}^{-1}$).

With the aerosol number concentration and BC data at SORPES, Kulmala et al. [61] further investigated the contribution of primary and secondary contributions to mode-segregated particle number concentrations and compared the results obtained at the SMEAR station in Finland. They found that in both the remote forest station and SORPES station with a strong impact from human activities, the majority of particles were of secondary

origin. Secondary aerosols dominated the nucleation and Aitken modes, and were about half of accumulation mode particles.

3.2 Chemical formation mechanisms for secondary pollutants

One of the current focuses of SORPES is to understand the processes related to the formation of secondary aerosols, including sulfate, nitrate and newly formed particles. To understand the chemical transformation of secondary aerosols, their precursor gases and related oxidants, several data analysis and modeling studies were conducted based on data obtained at the SORPES station.

Nitrous acid (HONO), being an important source of hydroxyl radical (OH), is one of the trace gases influencing the chemical processes of secondary pollutants [65,66]. Based on a two-month intensive measurement, Nie et al. [62] found that the HONO concentration and HONO/NO₂ ratio were enhanced by a factor of about 2, with over 80% of the observed HONO being secondarily produced by a

Table 1 Main results obtained from SORPES station during 2011–2015

main research themes		study period	main results and key findings	references
overall characteristics	O ₃ and PM _{2.5}	2011.8–2012.7	VOC-limited regime for O ₃ production; elevated secondary aerosols in summer. Synoptic weather and human activities play a vital role in pollution episodes.	Ding et al. [16]
	new particle formation	2011.11–2012.3	Ion-induced nucleation plays a marginal role in NPF at SORPES. A simple empirical criterion was deduced to estimate NPF probability.	Herrmann et al. [60]
	NPF and growth	2011–2013	Particle formation rate peaks in spring while growth rate peaks in summer. Clean air masses favor NPF and polluted YRD air masses facilitate growth.	Qi et al. [56]
	identification of primary and secondary PM	2011–2014	The majority of particles are of secondary origin in both Nanjing and Hyytiälä. Secondary particles dominate particularly in the nucleation and Aitken modes.	Kulmala et al. [61]
chemical formation mechanisms	HONO formation	2012.3–2012.6	Biomass burning aerosols enhance the conversion of NO ₂ to HONO. Mixed anthropogenic and fire plumes further promote HONO formation.	Nie et al. [62]
	sulfate formation enhanced by NO ₂	2012.5–2012.6	NO ₂ promotes sulfate formation through catalytic and photochemical reactions. Aqueous-phase oxidation by NO ₂ elevates ambient sulfate and HONO level.	Xie et al. [53]
	NPF simulation	2013.6–2013.8	Regional and box model accomplish NPF simulations without VOC observation. Oxidation products of biogenic VOCs enhance growth of newly formed clusters.	Huang et al. [63]
air pollution–meteorology interactions	observational evidence for an extreme episode	2012.6	Air pollution modifies radiation transfer, temperature profile and precipitation. More stable stratification in turn enhances the accumulation of local pollution.	Ding et al. [29]
	theoretical analysis based on flux data	2013.5–2013.11	High PM enhances PBL stability, further increasing surface PM concentration. Feedback between PM and PBL gets more effective at high PM loadings.	Petäjä et al. [35]
	meteorology–chemistry online simulation	2012.6	Aerosol-induced energy reallocation adjusts thermal and humidity stratification. Modified convective activity and moisture transport redistributed precipitation.	Huang et al. [64]
	regional modeling and policy implication	2013.12	BC plays an important role in enhancing haze pollution in megacities. Reducing BC emission co-benefits mitigation of haze pollution and global warming.	Ding et al. [17]
land-atmosphere interaction	2013.3–2013.8	Surface type affects radiation balance, land-atmosphere exchanges and local climate. Urbanization and agricultural cultivation pose warming and cooling effects locally.	Guo et al. [50]	
measurement of aerosol optical properties and black carbon	2013.9–2015.1	Compensation parameter is backscatter fraction and SSA dependent. Backscatter fraction clearly affects aethalometer data and should be considered.	Virkkula et al. [37]	

heterogeneous conversion. It was suggested that the relatively high surface area in biomass burning aerosols, compared to the fossil fuel ones, was probably the main cause for the high conversion rate. Nie et al. [62] also reported that during a special “brown sky” episode in 10 June 2012, the mixed BB plumes with anthropogenic fossil fuel emissions caused even a higher HONO concentration and HONO/NO₂ ratio, together with enhanced sulfate formation.

Xie et al. [53] further investigated the mechanism of secondary sulfate formation during the “brown sky” episode. They found that the aqueous oxidation of S(IV) by NO₂ under foggy/cloudy conditions together high NH₃ concentrations were probably the main contributors to the sulfate formation when O₃ was completely titrated by NO. The formed nitrite, as a by-product, enhanced HONO formation and gas-phase transformation of sulfate in the downwind area (Fig. 5). Xie et al. [53] also reported another unique case, showing the formation of sulfate by NO₂ associated with strong NPF and growth events. High sulfate formation rates were observed in mineral dust plumes mixed with anthropogenic pollutants. Dust-induced photochemical heterogeneous reactions of NO₂ promoted the formation of additional HONO and OH radicals, and further enhanced the new particle formation and growth, as well as the secondary sulfate formation. These heterogeneous processes connected aerosol particles

to the atmospheric oxidation capacity and in turn promoted the formation of condensing vapors, e.g. sulfuric acid or highly oxygenated molecules (HOMs). These results are consistent with what was reported by Nie et al. [39] based on data measured at a mountain top site, Mt. Heng, in South China. These findings provide an alternative explanation for the pathway of NO₂-induced sulfate formation to that proposed by He et al. [30]. The two different formation mechanisms of NO₂ promote sulfate formation in dust and biomass burning plumes mixed with fossil fuel pollutants are summarized in Fig. 5.

Comparison of numerical models with measurements provides a possibility to investigate whether the current theoretical understanding can explain the observational features in specific environments. Sulfuric acid has been supposed to be the key vapor to drive the particle nucleation, while low volatile organic vapors like some oxidation products of monoterpenes have been assumed to drive the condensation growth [67]. Huang et al. [63] conducted a comprehensive modeling study on the occurrence of NPF events observed at the SORPES station, by combining WRF-Chem (the Weather Research and Forecasting Model coupled with Chemistry) and the MALTE-BOX (the Model to predict new aerosol formation in the lower troposphere) sectional box model [68]. Based on these simulations, they found that biogenic organic compounds, particularly monoterpenes, play an

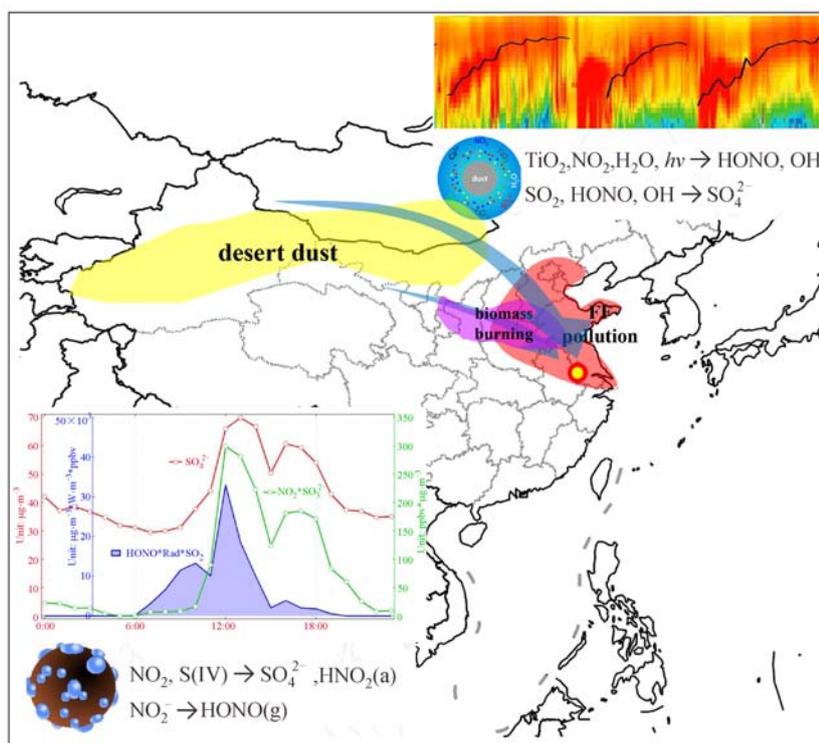


Fig. 5 A conceptual model for the NO₂ promoted sulfate formation via two different mechanisms: dust promoted photochemical heterogeneous reactions and aqueous-phase reactions in mixed plumes with biomass burning and fossil fuel sources

essential role in the initial condensational growth of newly formed clusters through their low-volatility oxidation products at the SORPES station.

3.3 Air pollution – meteorology interactions in mixed plumes

Meteorology, especially stagnant synoptic weather and stable PBL dynamics, plays a very important role in the formation of air pollution [16,32,33]. Intensive air pollution, especially high concentrations of radiatively active aerosols, on the other hand, could substantially influence meteorological conditions. As mentioned above, the geographical location of the SORPES station makes it an ideal location to study interactions of different sources of air pollutants and meteorological conditions. During the past few years, we have conducted many studies to investigate this kind of interactions from a two-way perspective based on field measurements at the SORPES station and modeling studies.

With continuous measurements at the SORPES station, Ding et al. [16] reported four pollution episodes under different synoptic weather conditions and found that stagnant anti-cyclones favor the transport of city cluster plumes from the south-east direction to Nanjing, resulting in multi-day O₃ or PM episodes. This work also found that the PBL meteorology substantially influenced the diurnal evolution of air pollutants in different ways. For example, different from other primary pollutants like CO and NO_x, the SO₂ concentration observed at the SORPES station generally shows a late-morning peak due to the fumigation of residual-layer plumes from elevated sources, such as power plants and industrial chimneys, associated with the vertical mixing in the developing PBL in the morning.

High concentrations of radiatively-active air pollutants, such as BC or brown carbon (BrC), have been found to play an important role in modifying weather/climate conditions [69,70]. Biomass burning is one of the important sources for BC and BrC [70]. In east China, agricultural fires, specifically field burning of wheat and rice straw, occur mainly in the harvest seasons and have been identified as one of the culprits of regional air pollution [16,71,72].

During the “brown sky” haze episode on 10 June 2012, representative of an exceptional intense fire event, a thick brown haze blanketed Nanjing and adjacent cities in the west YRD region. Extremely high PM concentrations were observed, together with regional anomalies in the near-surface air temperature and precipitation (Fig. 6): the air temperature dropped by almost 10 K, and the spatial distribution of rainfall changed during both daytime and nighttime [29,64]. Based on the comprehensive field measurement data at the SORPES station, solar radiation, sensible heat flux, air temperature and precipitation were found to be modified to a large extent [29]. Huang et al. [64] quantified this mechanism by conducting WRF-Chem

simulations. By considering the emission from biomass burning estimated from satellite fire counts, they found that the agricultural straw burning was responsible for the majority of the observed regional haze pollution in East China. In addition, they found that biomass burning plumes in the lower troposphere caused a significant cooling at the ground surface and warming in the atmosphere (Fig. 7), resulting in a change in boundary layer dynamics and precipitation patterns in both daytime and nighttime [64].

Based on comprehensive data analysis and modeling, Ding et al. [29] proposed a conceptual model to explain the main processes through which air pollution modified regional weather (Fig. 8). At the very beginning, agricultural burning plumes were transported aloft and mixed with anthropogenic fossil fuel combustion pollutants. Due to the considerable amount of light-absorbing components (e.g. BC and BrC) in the fire plumes, these aerosols would heat the atmosphere and cool the ground surface, thereby suppressing the vertical mixing and dispersion of pollutants. Enhanced boundary layer stability in turn led to more intense pollution in the lower boundary layer. Meanwhile, the cooling of the PBL and the resulting increase in the relative humidity amplified the feedback further by increasing the aerosol scattering coefficient through hygroscopic effects. The weakened convection caused by the more stagnant boundary layer, combined with changed cloud properties due to concentrated aerosol particles, further modified cloud properties and precipitation patterns.

Rather than being limited to an extraordinary biomass burning event, this kind of interaction between pollution and meteorology is a common phenomenon across East China [35]. Continuous (8-month) radiation measurements and pollution monitoring in Nanjing showed that both short-wave and long-wave radiation correlated negatively with the measured particle mass concentration, as did also the turbulent flux. The suppressed vertical mixing would confine the existing particles into a shallower boundary layer height, increasing their concentrations further. A theoretical calculation indicated that this amplifying effect might act as a plausible explanation for severe haze episodes in East China [35].

Following the previous two works, Ding et al. [17] further investigated the role of this feedback in haze pollution extending from the North China Plain to the YRD region during December 2013 by using the meteorology-chemistry coupled regional model WRF-Chem. Parallel experiments made it possible to investigate the key processes and components that connect the air pollution and boundary layer meteorology. Black carbon, which is intensively emitted by residential combustion, industrial activities and transportation, was identified as the main culprit. The relatively high heating efficiency of BC in the upper air changes the boundary layer dynamics and depresses the boundary layer height. Ding et al. [17]

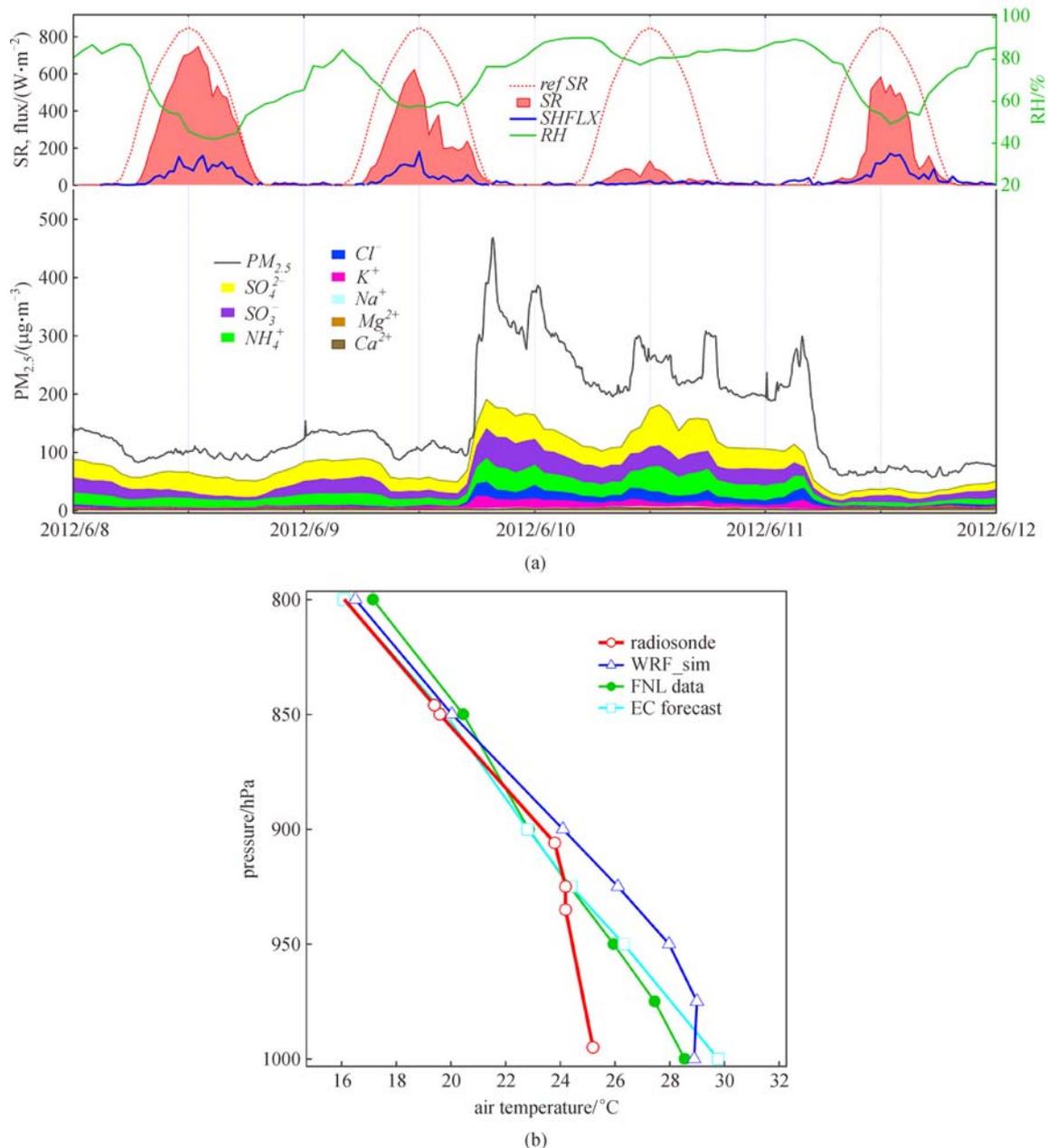


Fig. 6 (a) Time series of solar radiation, sensible heat flux and $\text{PM}_{2.5}$ mass and water soluble ions concentration, and (b) comparison of air temperature vertical profiles from the WRF simulation, FNL data and ECMWF forecast products and radiosonde measurement at Nanjing at 20:00 LT of 10 June, 2012. Note: Ref SR gives a reference of clear-sky solar radiation based on solar radiation measurement in the afternoon of 13 June with cloud-free sky. (Modified from Ding et al. [29])

named this effect as BC's "dome effect". Together with dimming at the surface, it favored a capping inversion in the upper boundary layer, followed by concentrated near-surface PM concentration during this haze case. The BC's role in the initiation of a positive feedback between air pollution and boundary layer meteorology highlights the importance of continuous and effective control on BC emission in China's air quality improvement, which could simultaneously contribute to the mitigation of global warming at the same time.

4 Future outlook

The above discussions summarize the current key findings obtained from the SORPES measurements and relevant modeling studies in the past few years. Besides these, there are a series of activities that are still ongoing. These results demonstrate the unique role of this "flagship" station in improving our current understanding on air quality, chemical mechanisms of secondary pollution, and the two-way interactions between air pollution and boundary

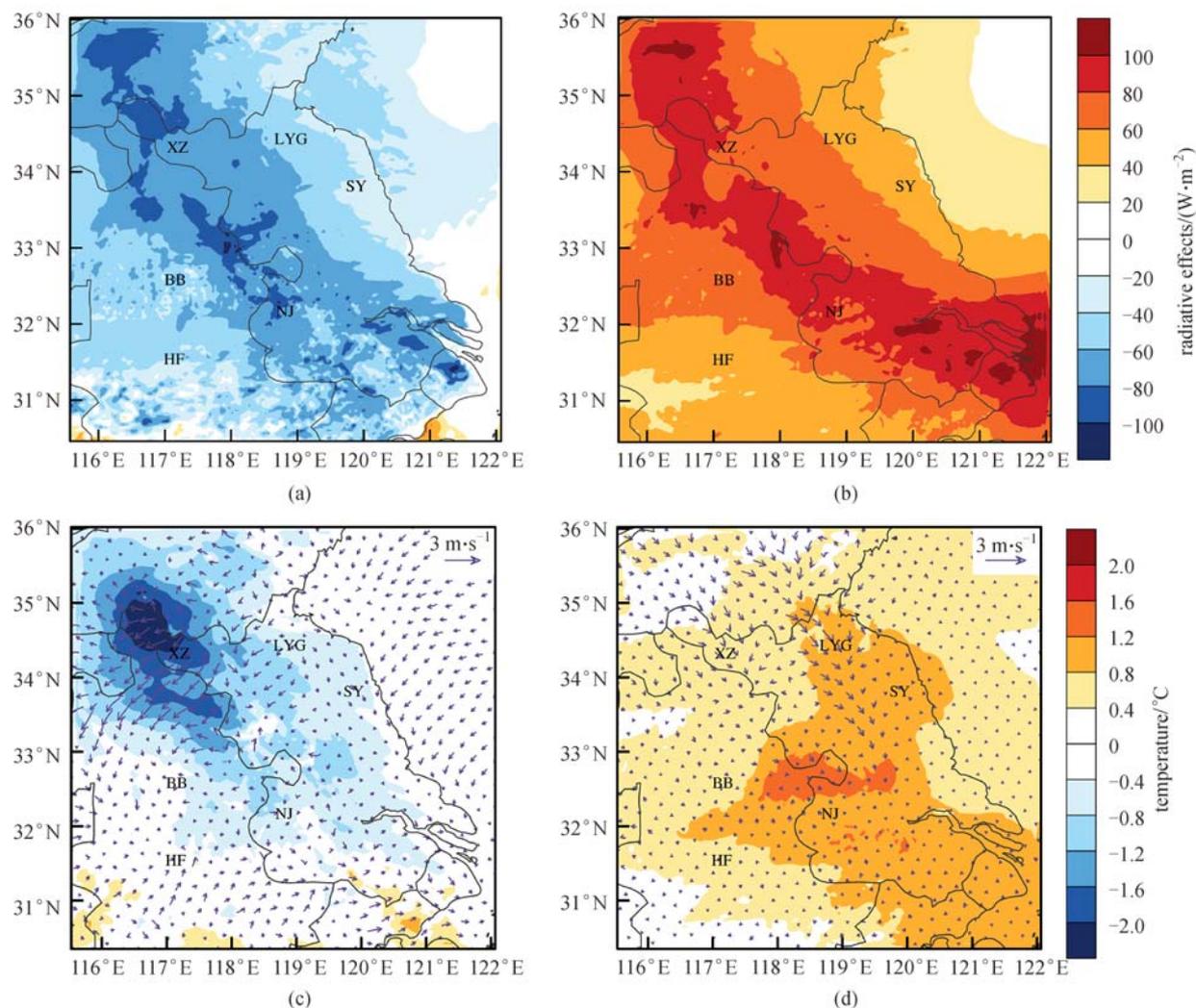


Fig. 7 Radiative forcing (a) at the surface and (b) in the atmosphere due to anthropogenic and biomass burnings aerosols on 10 June 2012; and aerosol-induced changes in air temperature and wind fields (c) near the surface and (d) at the altitude of 2 km (Figure modified from Huang et al. [64])

layer meteorology. Some of the new findings in chemical and physical processes or mechanisms will be further introduced into state-of-the-art numerical models in the future. Of course, the measurement data will be used to the evaluation these models, and to further understand the impact of the processes on a larger scale.

According to the overall concept design of the SORPES and the obtained scientific results, future development of the station will be focused on the following aspects:

1) From the atmospheric chemistry research point of view, the measurement components and instruments of SORPES will be further updated with more advanced instruments. For example, to meet the need for an in-depth understanding of secondary organic aerosols and ozone photochemistry at the SORPES station, online measurements of VOCs based on GC-MS and/or PTR-MS and organic aerosols based on ACSM, for example, will be further applied for long-term measurements.

2) To understand some specific chemical processes, especially detailed chemical mechanisms from molecular scales, some instruments for radical measurements (such as $\text{HO}_2\text{-RO}_2$ radicals by TOF-CIMS) [73] and size-resolved chemical species detection (such as TOF-AMS) will be further applied at this station. These instruments will be used for some specifically-designed intensive field campaigns and for flow tube-based laboratory studies combined with the online measurements at the station.

3) Because many previous studies have demonstrated the significance of air pollution-meteorology interactions in this regions with complex pollution sources, the vertical observation capacities based on remote sensing techniques (including DOAS-based retrieval, wind profilers and microwave radiometer etc.) and other sensor-based vertical sounding techniques onboard tagged and free-released balloons as well as unmanned aerial vehicles will be further developed and applied at this station.

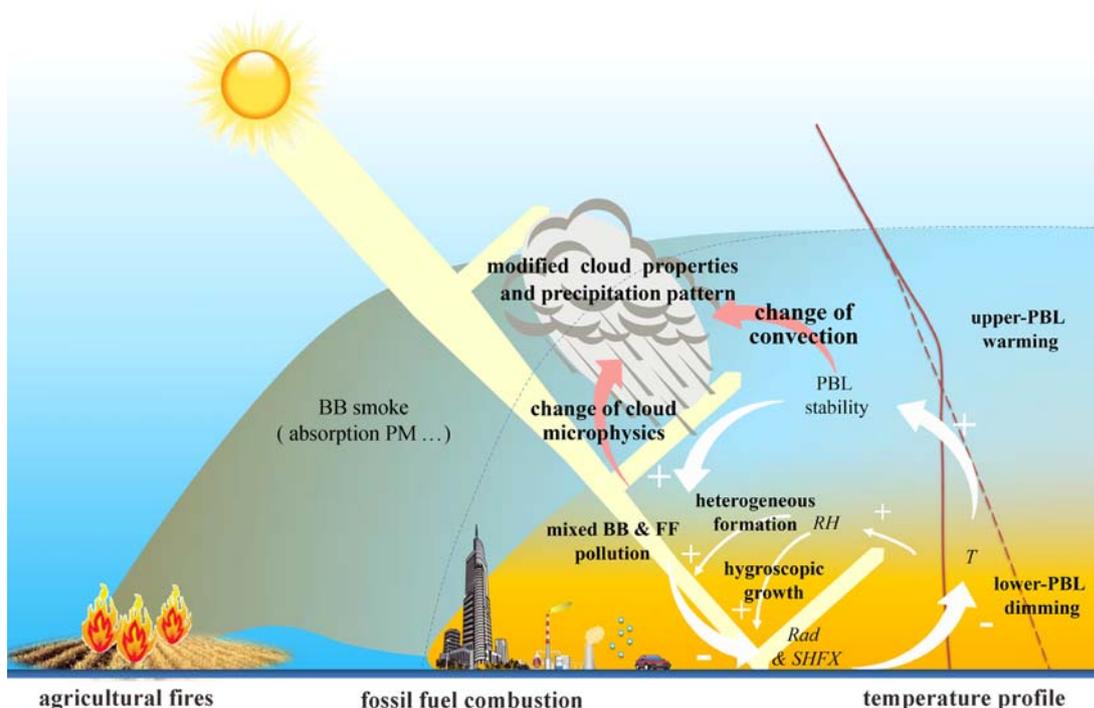


Fig. 8 A schematic figure for interactions of air pollution–PBL dynamics and aerosol–radiation–cloud for the mixed agriculture burning plumes and fossil fuel combustion pollutants (Modified from Ding et al. [29])

4) The current SORPES station is only at its first stage of development. Other components of the earth system process will be developed in the coming future. For example, according to the overall concept of earth system processes study, the two specific processes, ecosystem–atmosphere interactions and hydrological cycle need to be further developed in the future. More components has been added to understand the emission/flux of biogenic reactive VOCs [74] and greenhouses, deposition and concentration of toxic species like mercury [75] and their impacts on air quality and health of the land–ecosystem. A synthesis networking of the flagship station with other “satellite” sites in the surrounding region will also be improved.

5) The achievement in the development of SORPES has benefited from the close collaboration between Nanjing University and University of Helsinki, which has been further developed into a solid joint research platform, the Joint international research Laboratory of ATmospheric and Earth SysTEM sciences (JirLATEST). In the framework of JirLATEST, the SORPES and SMEAR II stations will be concurrently run for long-term. The data from both stations will be compared to understand the different mechanisms in the remote boreal forest and the subtropical East Asia region with intensive human activities.

6) The SORPES station has been continuous run for almost 5 years since 2011 and it has already archived a large number of high quality data. These data will be provided to the community, especially to the modelers, in

order to improve the current understanding of air quality, atmospheric chemistry mechanisms, and their impacts on climate change and human health in China and the East Asian regions.

For the last, but not the least, the development and progress of SORPES clearly demonstrate how an integrated “flagship” station with different components could improve the current understanding of physical and chemical processes in the earth system. Given that China and many other countries are facing a great challenge in environment and climate change, there is an urgent need to build more such kind of integrated stations in different geological regions in the world, especially the developing countries. Only with more measurements and more data, we can achieve a holistic understanding about the earth system and reach a better capacity for predicting the “future earth” using the earth system models.

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