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An Integrated Source Apportionment Methodology and Its Application over the Yangtze River Delta Region, China

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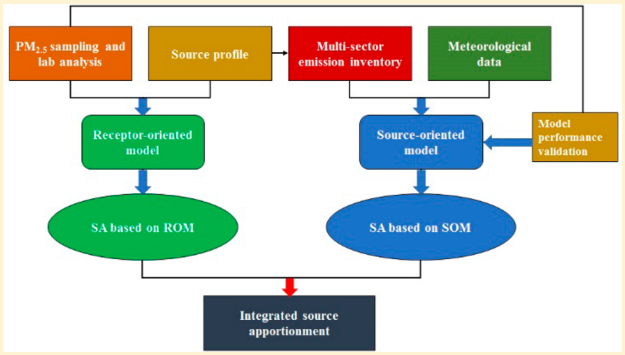
Supporting Information

ABSTRACT: An integrated source apportionment methodology is developed by amalgamating the receptor-oriented model (ROM) and source-oriented numerical simulations (SOM) together to eliminate the weaknesses of individual SA methods. This approach attempts to apportion and dissect the $PM_{2.5}$ sources in the Yangtze River Delta region during winter. First, three ROM models (CMB, PMF, ME2) are applied and compared for the preliminary SA results, with information from $PM_{2.5}$ sampling and lab analysis during the winter seasons. The detailed source category contribution of SOM to $PM_{2.5}$ is further simulated using the WRF-CAMx model. The two pieces of information from both ROM and SOM are then stitched together to give a comprehensive information on the $PM_{2.5}$ sources over the region. With the integrated approach, the detailed contributing sources of the ambient $PM_{2.5}$ at different receptors including rural and urban, coastal and in-land, northern and southern receptors are analyzed. The results are compared with previous data and shows good agreement. This integrative approach is more comprehensive and is able to produce a more profound and detailed understanding between the sources and receptors, compared with single models.



INTRODUCTION

The ever-growing air pollution in recent years has become a serious environmental issue in China.[1,2] Severe haze episodes with high concentration of $PM_{2.5}$ frequently occur in winter due to the high emissions intensity and unfavorable meteorological conditions. The high $PM_{2.5}$ pollution is generally contributed by both primary particles and secondary aerosol formed through the interaction of numerous gaseous precursors from various sources. $PM_{2.5}$, an aggregation of aerosols with fine particle sizes is comprised of directly emitted aerosols such as local combustion, vehicular, industrial, biogenic source, and secondary aerosols including nitrate, sulfate, ammonium salt, and secondary organic aerosol. In recent years, the air pollution with $PM_{2.5}$ as the major air pollutant has garnered great attention over the world, especially in China. As of 2015, there is more than 91% of the Chinese population exposed to level above of annual $PM_{2.5}$ mean concentration standard according to National Ambient Air Quality Standard II (NAAQS-II) of China (35 $\mu g/m^3$) and almost none of the Chinese cities has met the World Health Organization (WHO) standard (10 $\mu g/m^3$).[2] These aerosols of aerodynamic diameter less than 2.5 μm are inhalable and poses great health threat to the respiratory and cardiovascular system of human despite short-term exposure.[3,4] The effective reduction of the $PM_{2.5}$ level requires implementation of multipollutant control strategy.[5,6] Several approaches have been developed over the years to identify the respective sources of $PM_{2.5}$ including receptor-oriented models (ROM) and source-oriented models (SOM). These approaches are collectively known as the source apportionment (SA) techniques.

ROM have been one of the most widely applied SA method in China due to its simplicity.[7−9] As the name implies, this model analyses the physical and chemical properties from the standpoint of receptor and hence deduce the contribution from the pollution source based on prescribed emission profile. The information on the receptor is usually obtained from the in situ measurement of $PM_{2.5}$ related chemical species. The SA

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result based on ROM is reliable; however, this methodology has the problem of collinearity and, often, unidentified sources of secondary emission components. Besides, it cannot quantify the effect of local and regional transportation between or beyond the measurement networks. These have limited further effort to refine the source analysis.

On the other hand, the SOM approach analyze the contribution of respective $PM_{2.5}$ sources from the output of chemical weather prediction (CWP) model through sensitivity analysis or species tracing approach. Due to the usage of CWP result, the SOM excels its counterparts by accounting the weather condition, transportation, physical, and chemical reaction of the particulate matter in the atmosphere.[10−12] Nevertheless, the uncertainties of CWP including emission inventory, weather condition, chemical mechanism, and other aspects retain and might cause the discrimination of the $PM_{2.5}$ sources and its components.

As far as $PM_{2.5}$ is concerned, a number of stringent government policies has successfully lowered the concentration in the Yangtze River Delta (YRD) region. Hence the management of $PM_{2.5}$ has shifted from mitigation to precise management and quality control. As indicated above the current traditional sampling-apportionment techniques do not satisfy the need for the provision of precise information on the emission source and characteristics; while the widely used modeling approach has a large uncertainty. There is an urgent need to establish a new technique to achieve this goal for $PM_{2.5}$ apportionment. Moreover, in China most of the $PM_{2.5}$ apportionment is conducted using a single apportionment methodology which forbids validation. At present, almost every province uses a different or multiple apportionment techniques which is both expensive and clumsy. Due to the limitations and uncertainties of single SA methodology, more and more researches have focused on the combined approach targeting source apportionment in recent years. Some of these studies are related to the intercomparison of receptor models;[13] comparisons between receptor model and dispersion model;[14,15] some of the researches developed different integrated source apportionment methodologies based on different techniques, including synergic SA using PMF and PSAT;[14] combination of ROM and national emissions inventory (NEI);[16] ensemble methods of ROM and chemical transport models (CTM);[17,18] and nonlinear optimization.[19−21] For example, Bove et al.[14] performed a preliminary synergic source apportionment exercise using PMF and PSAT in Genoa, which mainly attempted to overcome the difficulties affecting comparisons between receptor and chemical transport models, particularly for the grouping of PM sources and the apportionment of secondary components. Contini et al.[15] applied PMF and Si/Al diagnostic ratio for the source apportionment study of PM_{10} to figure out the impact of a large coal-fired power plant to PM_{10} in Italy. Sturtz et al.[18] incorporated the source contributions to total fine particle carbon predicted by a CTM into the PMF receptor model to form a receptor-oriented hybrid model. Hu et al.[19] developed a hybrid approach integrating receptor model and chemical transport model, which utilizes kriging to spatially interpolate source-specific impact adjustment factors to generate revised CTM source impact fields from the CTM-ROM method results.[20] A subset of these hybrid modeling approaches includes a deterministic CTM that includes secondary formation of particulate matters and receptor information on particle compositions.[19]

In this research, we further developed an integrated source apportionment methodology to incorporate the advantages of both ROM and SOM. It combined the observation and receptor modeling (ROM) techniques, together with the chemical weather prediction (SOM) modeling to perform the $PM_{2.5}$ source apportionment for the major cities over the Yangtze Delta (YRD) region. First, an intercomparison has been done regarding the performance of three receptor models including PMF, CMB and ME2 based on a field campaign, and ME2 is selected as the SOM method; Next, two scenarios based on WRF-SMOKE-CAMx-PSAT has been run to figure out the detailed source contribution (including 15 source sectors and local vs regional transport) to both primary and secondary particles. Last, with ROM results as constrains and SOM results as relative contribution factors, the mapping between sources in ROM and SOM is established. The integrated source impacts are figured out by scaling ME2 (ROM) source impacts by CAMx-PSAT (SOM) source impacts. The $PM_{2.5}$ emission sources could now be optimized and adjusted to refine the source apportionment result. The breakthrough technique is applied onto the atmospheric composition of $PM_{2.5}$ in YRD region to efficiently and accurately identify the corresponding emission sources. It is expected to provide important scientific proof and support for the said sources. The development of such methodology increases the confidence level to identify the major contributing emission sources, from which corresponding prevention and control policy can be implemented to improve the air quality. Such effect can provide strong scientific support at midst of severe pollution scenarios for realistic abatement strategy.

EXPERIMENTAL APPARATUS AND MATERIALS

The experiment was conducted during the winter season that recorded with high concentration of $PM_{2.5}$ due to the emissions and unfavorable meteorological conditions.[2,22] First, the chemical components of measurement $PM_{2.5}$ at designated receptor sites were analyzed and ingested into ROM to recognize the contributing sources. Second, the SOM traced the sources of $PM_{2.5}$ and its components (primary and secondary) from source sectors in the emission inventory. Lastly, with the ROM as the basis and constrains, the contributing factors of the sources identified from numerical model is incorporated for secondary analysis of the emission source. The integration of these two approaches have refined the evaluation of the dominant regional emission sources at the receptor sites.

Receptor-Oriented Models (ROM). Positive matrix factorization (PMF), chemical mass balance (CMB), and multilinear engine2 (ME2) are among some commonly used ROM methods. In this study, we applied PMF5.0, EPA CMB8.2, and ME2 to do source apportionment based on field measurements. We have compared the three types of ROM in our work to decide on which one is more suitable for the case study, we have eventually settled on ME2. Since such comparative study has been performed in several literatures before, we decided not to elaborate much in the main content of the paper. It is now included in the Supporting Information (SI) for readers who are interested.

The ROM study collected and analyzed offline $PM_{2.5}$ samples for the composition of ions, carbonaceous aerosols, and trace elements. The samples were collected from 10th November 2014 to 14th January 2015 to represent winter at the eight typical representations of air quality condition on land use in

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Table 1. Location and Description of Offline Sampling Sites for ROM

no	stations	location	location description
1	Shanghai urban (SHH_URB)	31.1717°N 121.425°E	surrounded by residential housing and commercial building, without apparent industrial emissions. The land surface is similar to the Shanghai urban
2	Nanjing urban (NAJ_URB)	32.0578°N 118.775°E	surrounded by commercial and residential buildings without apparent industrial emissions. The land surface is similar to the Nanjing urban
3	Hangzhou urban (HAZ_URB)	30.2733°N 120.138°E	surrounded by commercial and residential buildings without apparent industrial emissions. The land surface is similar to the Hangzhou urban
4	Suqian suburban (SUQ_SURB)	33.2347°N 118.326°E	there are no apparent industrial emissions near the station
5	Dafeng suburban (DAF_SURB)	33.5044°N 120.559°E	it is surrounded by wetlands and has no apparent industrial emissions near the station
6	Cixi suburban (CIX_SURB)	30.3125°N 121.64°E	there is no apparent industrial emission near the station
7	Lin'an suburban (LNA_SURB)	30.3025°N 119.75°E	the surroundings are made up of hills, forests and farmlands. The vegetated lands are mainly composed of shrublands and bamboos. There is no large village within proximity of 3 km
8	Shanghai suburban (SHH_SURB)	31.0506°N 121.796°E	there are no apparent industrial emissions near the station

different cities as shown in Table 1 and Figure 1. The sampling period was 1000 LST to 0900 LST of the second day, with an hour allocated for the manual replacement of filter. The samples were then analyzed in the laboratories to obtain the chemical components for ROM analysis.

All the measurement points used four-channel particulate collector (TH-16A, Wuhan TianHong Environmental Protection Industry Co., Ltd., China) with $PM_{2.5}$ impactor installed in each channel. The channels were installed with two Teflon filters (47 mm, Whatman Inc., UK) and two preheated Quartz filters (47 mm, PALL Life Sciences) at inlet flow rate of 16.7 L/min. Nevertheless, the SHH_SURB has used the four-channel particulate collector (Partisol 2300, Thermo Scientific). The settings were same as above except the inlet flow rate of the two Teflon channels was set to 10 L/min. The samples collected from Teflon filter were analyzed for the mass concentration of $PM_{2.5}$ and the water-soluble ionic components and inorganic elements, while sample collected from Quartz filter were analyzed for the carbonaceous components, elemental carbon (EC) and organic carbon (OC). The latter also served as supporting data for the water-soluble ionic components when measurement of Teflon filter was deemed unsuitable for analysis.

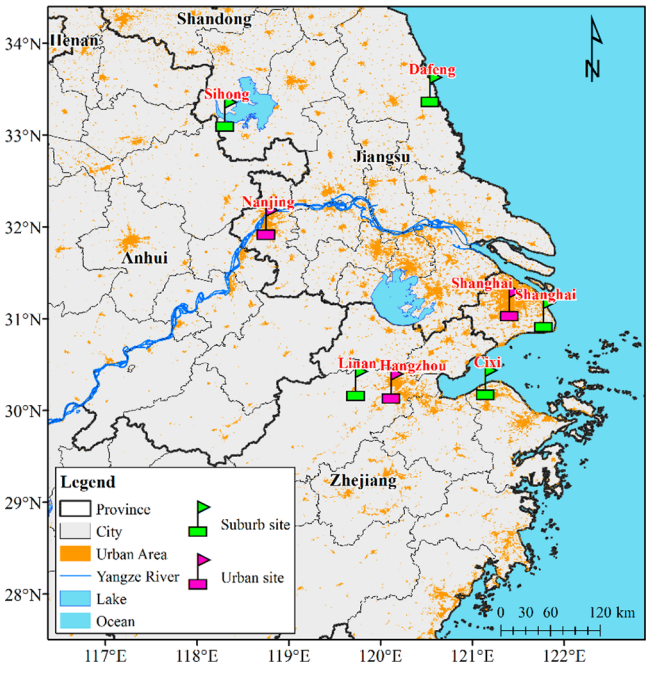

Figure 1. Location of the offline sampling sites.

The mass concentration was quantified from Teflon filter following the weighing procedure specified in "The Technical Specifications of Airborne Particulates ($PM_{2.5}$) Manual Measurement Method (Gravimetric)" (HJ 656−2013). The filter samples were extracted in ultrapure water with the low temperature (below 20 °C) ultrasonic technique and subsequently analyzed with the Ion chromatography (940 Professional IC Vario, Metrohm AG, Swiss) for the water-soluble ionic components, including F^-, Cl^-, NO_2^-, NO_3^-, PO_4^{3-}, SO_4^{2-}, $C_2O_4^{2-}$, Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}. The inorganic components from filter sample were analyzed with the Epsilon 5 ED-XRF (PANalytical B.V., Netherlands), including Na, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Br, Rb, Sr, Mo, Cd, Sb, Ba, Pb. The Quartz filter samples were analyzed with thermal optical carbon analyzer (DRI model 2001A) to determine the carbonaceous components (EC and OC) in $PM_{2.5}$, according to the U.S. Interagency Monitoring of Protected Visual Environment (IMPROVE) protocol. The optical correction was analyzed according to the thermal optical reflective theory. The limit of detection (LOD) and relative standard deviations for each species were listed in SI Table S1.

The total number of samples involved in this ROM calculation is 496 sets. SI Table S2 listed the average

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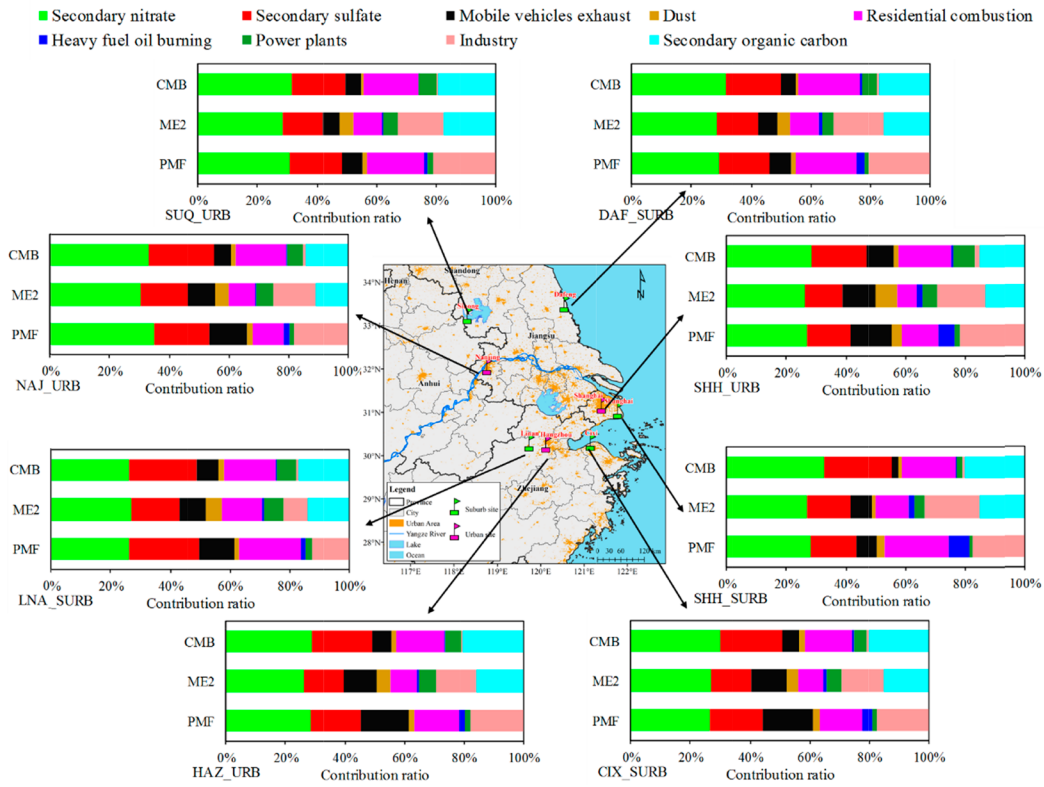

Figure 2. Comparisons among the source apportionment results based on three receptor models.

concentrations of each species in $PM_{2.5}$ obtained in different sampling sites. The offline sampling results of $PM_{2.5}$ in YRD region with conventional filter samples are analyzed with three types of ROMs, namely ME2, PMF, and CMB. A more detailed description and discussion of these ROMs can be found in Watson et al.,[23] Viana et al.,[24] Hopke,[25] and Belis et al.[26] The evaluation of ROM approach has been widely conducted and is not the focus of the paper, hence the comparison of the three ROM is only briefly discussed. Figure 2 shows the comparisons among the three receptor models. For comparison, only the identified emission sources are shown, whereas the unidentified part can be compared in SI Table S3. In Figure 2, the result obtained for PMF and ME2 is rather similar, while the result for CMB is greatly influenced by the source profile precision. For example, the result of dust of CMB deviates greatly since it generalized the source profile into the entire region without difference among the cities. It is also realized that the comparison is better in SHH_URB and SHH_SUBR. This suggested that the source profile used in the region is more locally calibrated compared to the remaining area. The ME2 approach is chosen since it has integrated the advantages of PMF and CMB to extract the source factors. The relatively stable and accurate source profile from the CMB is processed and ingested into ME2. The additional source profile can limit the randomness of the model to tackle the common accuracy problem of PMF.[27,28] More information on the comparison of ROM were given in the "Source Apportionment Methodology based on receptor models" section in the SI and Table S3. Table S3 indicates that there exists the unidentified mass between CMB and ME2 simulated against the observed data. For CMB, the reconstructed $PM_{2.5}$ percentage ranges between 84 and 89%; for ME2, it ranges between 82 and 85%. In this study, the sum of the measured chemical species does not include double counting species and unmeasured ions, like metal oxides, or hydrogen and oxygen associated with organic carbon, which can explain approximately 78% ∼ 83% of the $PM_{2.5}$ mass. For PMF and CMB models, both the concentrations of $PM_{2.5}$ mass and its identified chemical species were set as fitting species, and the $PM_{2.5}$ mass was set as total variable. Compared with PMF model, the sum of the CMB-calculated source contribution could not completely fit to $PM_{2.5}$ mass due to the complexity and limitation of source profiles like industrial source and other sources. For ME2 model, only the concentrations of measured species were set as fitting species. Therefore, the sum of the ME2-calculated source contribution fitted to the sum of measured species better than the observed $PM_{2.5}$ mass concentration. However, the reconstructed $PM_{2.5}$ concentrations in the range from 80% to 120% were acceptable.[29,30]

The ME2 model (hereinafter ME2-ROM) is used to apportion the sources after comparison. The source profiles (Refer to Supporting Information) of secondary nitrate, secondary sulfate and secondary carbon are set as limiting conditions to fit into the ME2 model, hence eight emission sources are identified. The contributing sources are listed according to the weight with sequence of large to small: secondary nitrate (NH_4^+, NO_3^-), secondary sulfate (NH_4^+, SO_4^{2-}), secondary organic carbon (SOC), motor vehicle exhaust (EC, OC, Cu), industrial and coal combustion (As, Se, Pb), dust (Al, Si, Ca, Ti, and other crustal elements), heavy fuel oil burning (Ni, V), biomass burning (K, OC) and industrial source (Cr, Ni, Pb). The result of ROM-ME2 is further discussed in "Results and Discussion".

Source-Oriented Model (SOM). The SOM method uses the result of chemical weather prediction, in this study, we have used WRF-CAMx result. The SOM has simulated the chemical field with Comprehensive Air Quality Model and coupled with

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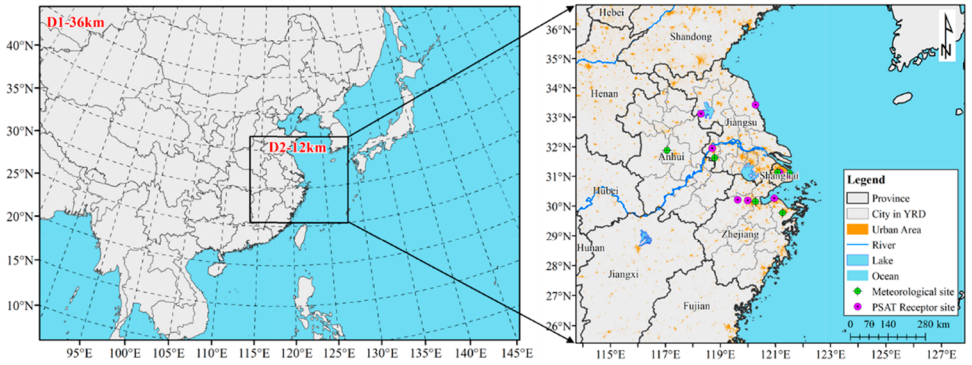

Figure 3. Domain settings of CAMx-PSAT with parent domain (D01) covering entire China and nest covering YRD (D02).

Table 2. Settings for WRF and CAMx-SAT Numerical Model

WRF settings	options	CAMx-PSAT settings	options
vertical layers	27 layers (100 hPa as top)	vertical layers	collapsed from WRF settings to 14 layers
land use map	30-m high resolution Global Land Cover	initial and lateral chemical boundary condition	MOZART
initial and lateral boundary condition	NCEP/NCAR 1°×1° at 6 h interval	anthropogenic emission inventory	YRD: SMOKE-YRD; Outside YRD: MEIC-2012, MIX-2010
microphysics	Purdue Lin	biogenic emission inventory	MEGANv2.10
cumulus	Grell-3	gas-phase chemistry	CB6
land surface model (LSM)	Noah LSM	water-phase chemical mechanism	RADM
planetary boundary layer	Yonsei University	inorganic chemistry mechanism	ISORROPIA
longwave radiation	RRTM	aerosol chemical mechanism	CF
shortwave radiation	Goddard	secondary aerosol chemical mechanism	SOAP2

Extensions-Particle Source Apportionment Technology (CAMx-PSAT version 6.40; http://www.camx.com)[31] to track the emission sources of $PM_{2.5}$ and their species. The Weather Research and Forecast (WRF) version 3 supplied the meteorological field for CAMx-PSAT while the Sparse Matrix Operator Kernel Emissions (SMOKE)[32] emission inventory building system supplied the area- and source- speciated emission data. The SOM approach is also hereinafter known as WRF/CAMx/PSAT. The model domain is made up of two nests: D01 (199 column ×139 row, 36 km) is centered at the 118°E, 32°N to cover the entire China, some Asian countries such as Korea and Japan, while D02 (120 column ×105 row, 12 km) cover the entire YRD area and its surrounding area including sea. The coverage of CAMx-PSAT is similar to WRF but 3 grids smaller on each side to remove the influence of lateral boundary condition, as illustrated in Figure 3. In this study, the locally customized emission system for YRD, SMOKE-YRD was used to produce the anthropogenic emission in YRD while emission beyond YRD was supplied by Multiresolution Emission Inventory for China (MEIC-2012) (http://www.meicmodel.org) and MIX-2010.[33] The emissions data, characteristics and factors information were simultaneously fed into SMOKE model and are processed according to three-dimensional space, time, and species. The anthropogenic data was then combined with the biogenic data from Model for Emissions of Gases and Aerosol from Nature (MEGAN version 2.10)[34] as the final input of emission inventory for the CWP. The details of WRF/SMOKE/CAMx-PSAT settings are given in Table 2.

An additional simulation was run to disable the chemical mechanism to isolate the contribution of the regional transportation on primary and secondary sources from outside (initial and lateral boundary transportation) on the $PM_{2.5}$ component. There are 15 types of emission sources in the inventory, including power plants, industrial boilers, cement, iron and steel, petrochemical engineering, other industries, diesel vehicle, gasoline vehicle, nonroad mobile, ships, dust, agricultural, cooking, other residential emissions and biogenic emissions. Locations of the receptors and meteorological sites used for model verification are shown in Figure 3.

Integrated Source Apportionment Technique. three steps are included in this methodology.

Step 1 (ROM-ME2). Base on the chemical compositions of $PM_{2.5}$ observed through filter analysis, we run ROM-ME2 (selected in this study) to get the basic SA results. With this methodology, the primary sources, secondary components and undefined sources are identified, respectively. The ROM approach apportioned the sources of $PM_{2.5}$ according to eq 1. RMC_t indicates the total mass concentration of $PM_{2.5}$ at the receptors, RMC_{ps}, RMC_{sc}, RMC_{os} indicate the concentration of $PM_{2.5}$ from primary, secondary components, unidentified emission sources at the receptor. *i* indicates the contributions to primary $PM_{2.5}$ from m source categories; *m* indicates the number of primary $PM_{2.5}$ source categories identified via ROM. *j* indicates the secondary contributions to $PM_{2.5}$, *n* indicates the number of secondary $PM_{2.5}$ categories, including secondary sulfate, secondary nitrate, secondary organic aerosol.

$$\mathrm{RMC}t = \sum_{i=1}^{m} \mathrm{RMC}ps_i + \sum_{j=1}^{n} \mathrm{RMC}sc_j + \mathrm{RMcos} \quad (1)$$

Step 2 (SOM-WRF/CAMx/PSAT). We applied the SOM methodology (here refers to WRF/SMOKE/CAMx-PSAT numerical methodology) to quantify source contributions to primary $PM_{2.5}$ and secondary $PM_{2.5}$ with two scenarios. Scenario 1: each traced source emission category is divided into two parts, gas emissions (SO_2, NO_2, VOCs species, NH_3, etc.) and primary particle emissions (sulfate, element carbon, primary organic mass, other unclassified species etc.). Contribution from the gas

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emissions is identified as secondary contribution, whereas the other is treated as primary contribution. Scenario 2: chemistry mechanism was turned off to quantify the primary contribution from super regional transport, then its secondary contribution is calculated as the difference between Sce.1 and Sce.2. Finally, source contributions to each chemical component of $PM_{2.5}$ for every refined source is quantified. Eq 2 defines the process based on SOM.

$$SMCt = \sum_{s=1}^{p} \{SMCpsc_s + \sum_{j=1}^{n} SMCssc_{(j,s)} + SMCosc_s\} \quad (2)$$

SMCt, $SMCpsc_s$, $SMCssc_{(j,s)}$, $SMCosc_s$ refer to the total mass concentration of $PM_{2.5}$ at the receptors predicted via SOM models, the contributions from s source to primary $PM_{2.5}$, secondary aerosol, and others at the receptor, respectively. *s* and *p* refer to the source category and total number of sources, respectively. *j* and *n* refer to the secondary species of $PM_{2.5}$ and the total number, respectively.

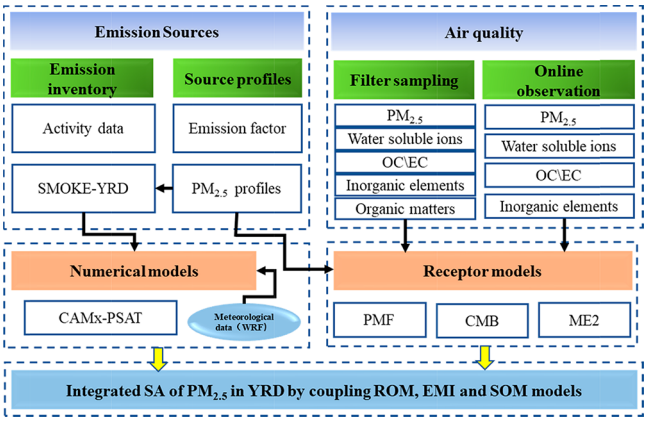

Figure 4. Integrated source apportionment technique applied in this paper.

Step 3 (ROM-SOM). The $PM_{2.5}$ sources obtained from ROM are briefly categorized into three types: primary sources, secondary components and other unidentified sources. The SOM approach further accounted the chemical transportation and reaction processes, including (1) refine primary emission sources from ROM with improved source categorization, (2) differentiate the secondary $PM_{2.5}$ components in the primary emission and secondary formation by marking these species and their primary particulate matters in the emission inventory for sources tracing, (3) the gaseous chemical mechanism of CWP to include more intermediate-VOC species into the emission inventory to improve the modeling result of secondary organic aerosol. With ROM results as constrains and SOM results as relative contribution factors, the mapping between sources in ROM and SOM is established. As shown in Figure 5, the sources identified via ROM are classified into three types including primary, secondary and undefined. The relative concentration of the primary source from ROM is refined with the corresponding concentration obtained from SOM. The approach used is shown in eq 3. Similar methodology is applied on the secondary and unidentified sources as shown in eqs 4 and 5.

$$IMCp_s = RMCps_i \times \frac{SMCpsc_s}{\sum_{s=1}^{p_i} SMCpsc_s} \quad (3)$$

$$IMCs_s = \sum_{j=1}^{n} \left\{ RMCsc_j \times \frac{SMCssc_{(j,s)}}{\sum_{s=1}^{p} SMCssc_{(j,s)}} \right\} \quad (4)$$

$$IMCo_s = RMcos \times \frac{SMcosc_s}{\sum_{s=1}^{p} SMcosc_s} \quad (5)$$

Based on results above, the integrated source apportionment result is given as IMC(s), as shown in eq 7, which is the sum of primary, secondary and unidentified sources as shown in eq 6.

$$IMC_s = IMCp_s + IMCs_s + IMCo_s \quad (6)$$

$$IMCt = \sum_{s=1}^{p} IMC_s \quad (7)$$

The methodology integrates offline sampling and numerical models to obtain a thorough and complete offline approach to study the $PM_{2.5}$ apportionment of YRD. The overall concept of integrated SA is shown in Figure 4.

RESULTS AND DISCUSSION

Source Apportionment Result with ROM (ME2). Through the ROM approach, the $PM_{2.5}$ source apportionment result from the eight stations during the winter season are as shown in Figure 6:

Secondary Sulfate, Secondary Nitrate, And Secondary Organic Carbon. These three sources are the main pollution sources in YRD region with total percentage around 50%. Owing to the severe pollution during winter, the secondary nitrate has a greater contribution compared to the secondary sulfate. Secondary nitrate contributed around 26.1−30.4% to the total $PM_{2.5}$ amount, with largest contribution at NAJ_URB (30.4%) and least at SHH_SURB (26.1%). The secondary sulfate ranks second contributor, with the contributions ranging between 12.8−16.4%. The recorded largest contribution from secondary sulfate occurs at LNA_SURB (16.4%) and least from the SHH_URB (12.8%). The secondary organic carbon constitutes of around 10.7−17.4% which is the largest contribution from DAF_SURB.

Motor Vehicles Exhaust. The motor vehicle exhaust in YRD is substantial with around 5.8−12.2% of total $PM_{2.5}$ concentrations. Especially high contributions in SHH_URB, HAZ_URB and NAJ_URB, much higher than rural sites. The significant emission is attributed to the large amount of motor vehicles in the mega cities (Shanghai: 3.3 million; Hangzhou 2.7 million; and Nanjing: 2.2 million in the year 2015; from statistical yearbook).

Industry and Power Plants. Industry refers to emissions from industrial boilers and metal smelting, which contributes a lot to the ambient $PM_{2.5}$ concentrations, with 6.8−16.7%. The contribution of power plants is relatively smaller than industrial source, which is because the more than 60% of the coal burning is in powerplants and are installed with ultralow emission control facilities.

Dust. The dust makes up around 1.4−7.2% of the $PM_{2.5}$ source over all measured sites.

Heavy Fuel Oil Burning. It has contributed around 1.0−2.0% and mostly from the SHH_URB and least from SUQ_SURB and DAF_SURB (1%). The pollutants are mainly produced at the harbor and river ships. However, due to the northerlies monsoon during the winter seasons, the contribution of the source is relatively small than summer seasons.

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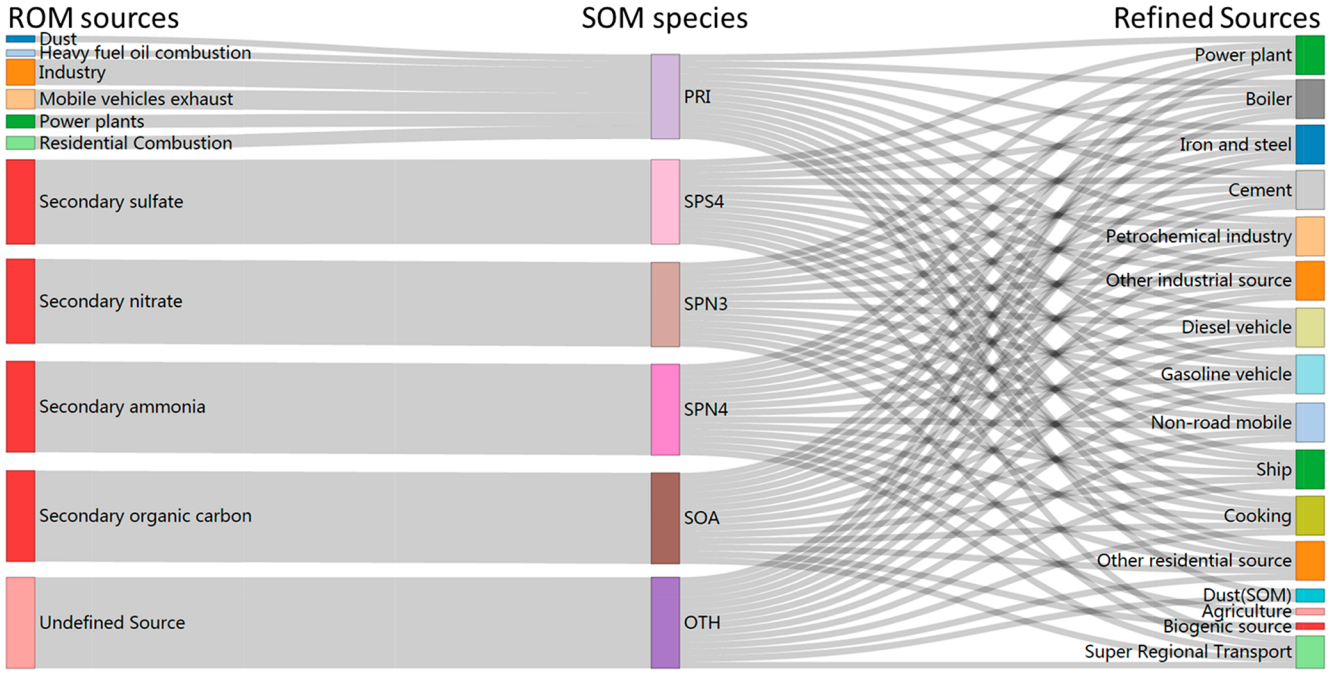

Figure 5. Source mapping between ROM and SOM used in the integrated source apportionment technique applied in this paper. (PRI: primary contribution besides unclassified species in $PM_{2.5}$, SPS4: secondary sulfate, SPN3: secondary nitrate, SPN4: secondary ammonia, SOA: secondary organic mass, OTH: unclassified species in $PM_{2.5}$).

Residential Combustion. In winter, the residential combustion is still one of the source contributors to ambient $PM_{2.5}$, with the contributions ranging between 6.8 and 13.6%. The contributions from residential combustion in rural area are significantly higher than urban region, with the contributions DAF_SURB (9.7%), SHH_SURB (11.1%), LNA_SURB (13.6%). This indicates that residential combustion both outside and within households is still a major source in rural area to ambient $PM_{2.5}$ pollution.

It is expected that the ROM result is solely based on the nine identified sources given in the source profile. Nevertheless, the EMI data set for CWP has considered a larger varieties of emission sources. Some of which are branched out from the ROM sources, the other account for the unidentified sources left out by the ROM, notably regional transportation.

Source Apportionment with SOM (WRF/SMOKE/CAMx-PSAT). The result of respective weather and chemical model are verified to ensure the applicability and reliability of the numerical models. The meteorological parameters of WRF model is verified with the measurement data from the weather stations in YRD (marked in Figure 1) for 2014−2015 year. Table 3 shows that the temperature is well predicted in the YRD region. The MB, NMB, NME and MFB of temperature maintained below 1, 6%, 12%, 0.1 °C, respectively, and IOA and R are close to 1. Similarly, the MB, NMB, NME, and MFB of predicted wind speed is maintained below 1 m/s, 29%, 41%, and 0.41 m/s, while IOA above 0.7. The model performance of station on higher elevation is limited by the WRF model performance.

During the study period, the modeled $PM_{2.5}$ and its chemical components from CAMx-PSAT is verified against the real-time measured pollutant concentration and manual filter sampling respectively. As shown in Table 3, the MB of modeled $PM_{2.5}$ concentration has differed among the stations. The performance of the models is generally acceptable. Around 8 stations have obtained NMB, NME, MFB, and MFE between 13−68%, 36−69%, −7−54%, and 25−56% respectively; the R and IOA are also well within 0.43−0.53 and 0.5−0.7. Although the model has under-predicted the $PM_{2.5}$ mass concentration during several high pollution episodes, the overall trend and concentration agreed with the observation result. The concentration of $PM_{2.5}$, sulfate, nitrate, and ammonium salt produced close agreement of value and trend with observed values. The incomplete secondary aerosol chemical mechanism might have caused the underestimation of these secondary aerosols.[35] The elemental carbon concentration is also lower than observed. The discrimination is believed to be caused by the underestimate emission of biomass burning in rural area. The result from the numerical model performs reasonably accurate and reliable during the study period.

The CAMx-PSAT have produced an informative spatial map of the emission source contributions of each designated area in the YRD region as shown in Figure 7. Overall the super-regional transportation from outside of the boundary cannot be ignored over the region. The prevailing northerlies during winter tends to carry the air mass from northern China which is great emitter of coal burning emissions.[36,37] The major contributing sources to $PM_{2.5}$ at SHH_URB during winter season are boiler, industrial process, mobile, and dust with 14.1%, 11.7%, 12.9%, and 20.5% respectively. The main emission sources for northern Jiangsu (DAF_SURB, SUQ_SURB) are mainly boiler, mobile, dust and agricultural emission, with 12.6−14.9%, 6.2−6.6%, 10.5−11.0%, and 7.1−7.5% respectively. Similarly, the boiler and dust contributed a substantial amount of $PM_{2.5}$ with 21.8−25.4% respectively in NAJ_URB in southern Jiangsu. The emissions from power plant, processing industry, mobile source, and agricultural activities contributed to 3.2%, 12.0%, 7.2%, and 3.9%, respectively. The shipping source contributes higher $PM_{2.5}$ concentration near the river and coast (CIX_SURB, SHH_URB, SHH_SURB).

The overall $PM_{2.5}$ concentration in southern Zhejiang is lower than the northern Zhejiang. In the northern Zhejiang

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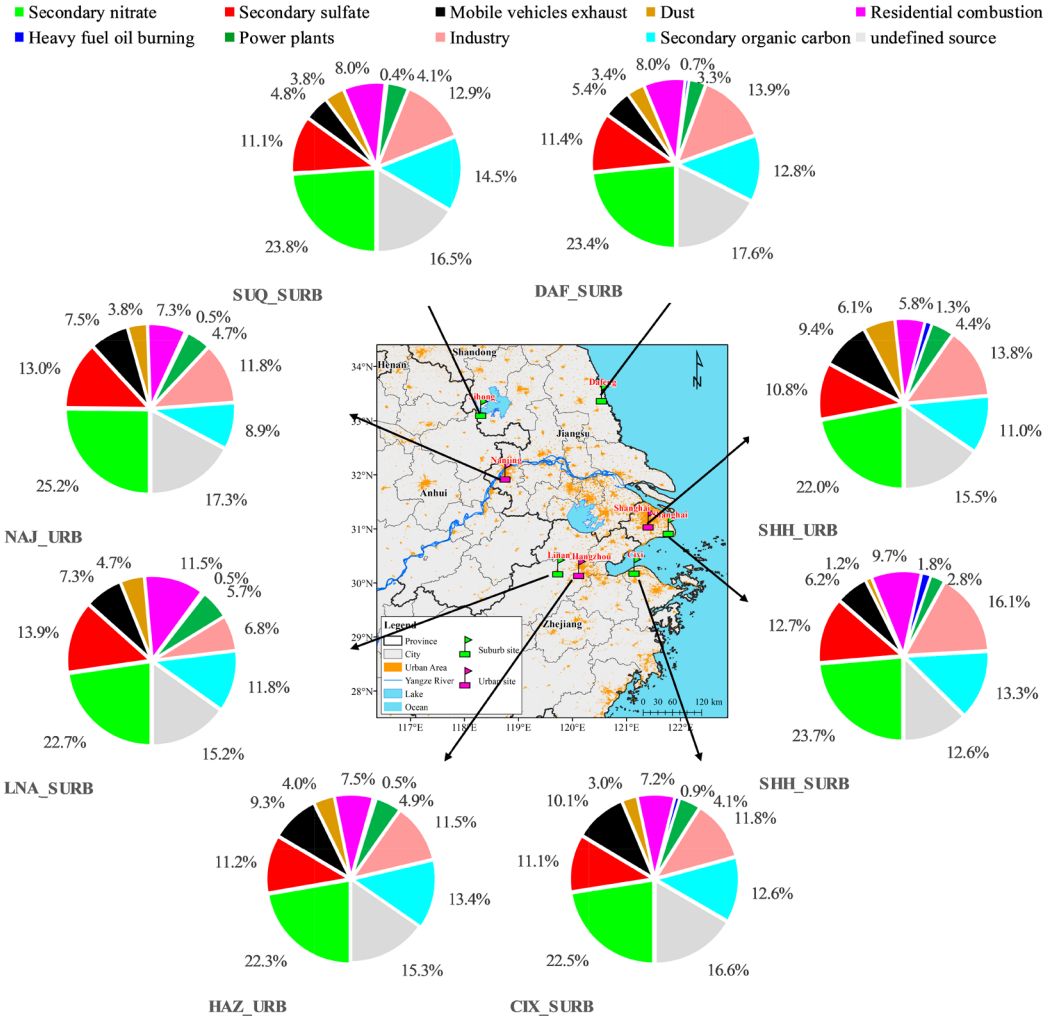

Figure 6. ROM-ME2 derived spatial distribution of the $PM_{2.5}$ contributing sources in YRD during winter.

Table 3. Statistical Verification of Meteorological Field in WRF and $PM_{2.5}$ and Its Chemical Components in CAMx Model Performance against the Observations

parameters[a]		data pairs	observed average	simulated average	MB	NMB	NME	MFB	MFE	R	IOA
meteorological factors	T	11904	8.1	9.0	0.9	11%	20%	0%	7%	0.94	0.96
	RH	11904	64.1%	60.4%	−3.6%	−5%	17%	0%	0%	0.79	0.88
	P	11904	1026.0	1025.4	−0.6	0%	0%	0%	0%	0.98	0.98
	WS	11904	3.0	3.8	0.8	32%	51%	5%	12%	0.68	0.77
	WD	11904				9%	39%	0%	0%	0.49	0.73
$PM_{2.5}$ concentrations and its chemical components	$PM_{2.5}$	496	73.5	56.0	−17.5	−24%	39%	−15%	28%	0.53	0.69
	sulfate	496	10.6	9.2	−1.4	−13%	36%	−7%	25%	0.50	0.70
	nitrate	496	14.6	11.2	−3.4	−23%	51%	−12%	38%	0.43	0.63
	ammonia	496	8.7	6.2	−2.5	−29%	42%	−19%	31%	0.54	0.68
	EC	496	4.9	1.6	−3.4	−68%	69%	−54%	56%	0.49	0.50
	OM	496	11.9	5.4	−6.5	−55%	58%	−40%	45%	0.51	0.55

[a]T—temperature; RH—relative humidity; P—pressure of sea surface; WS—wind speed; WD—wind direction; EC—element carbon; OM—organic mass; MB—the mean bias; NMB—the normalized mean bias; NME—the normalized mean error; MFB—the mean fractional bias; MFE—the mean fractional error; R—the correlation coefficient; IOA—the index of agreement.

(LNA_SURB, HAZ_URB, CIX_SURB), the stationary point sources including boiler and power plant have contributed to 21.0−22.7% of the $PM_{2.5}$ in the region. The cement, metallurgy and other processing industries contributed to 11.3−16.3% of emissions in total. Dust has a large influence on these stations also with around 10.3−31.3%.

In general, the $PM_{2.5}$ caused by local emissions from YRD in winter mainly originated from power plant, industrial boiler, industrial process, dust, mobile, and agricultural sources, making

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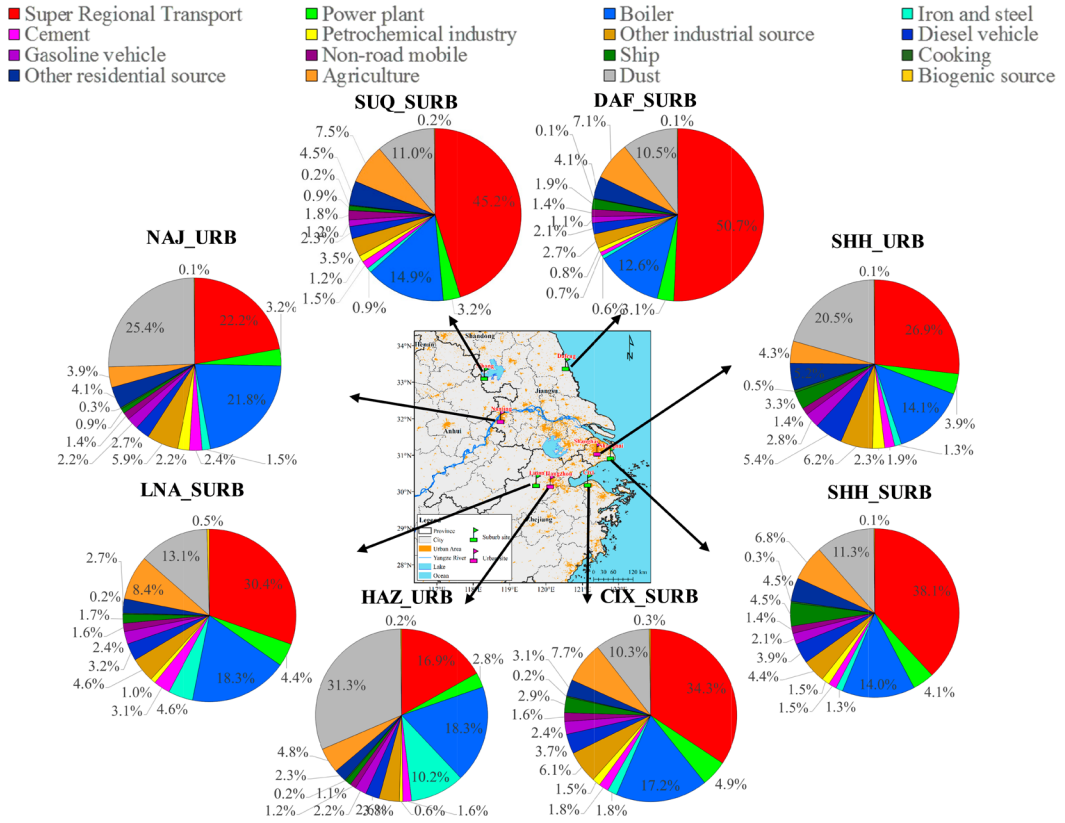

Figure 7. Source contributions to ambient $PM_{2.5}$ at the eight receptors in YRD during winter based on SOM methodology.

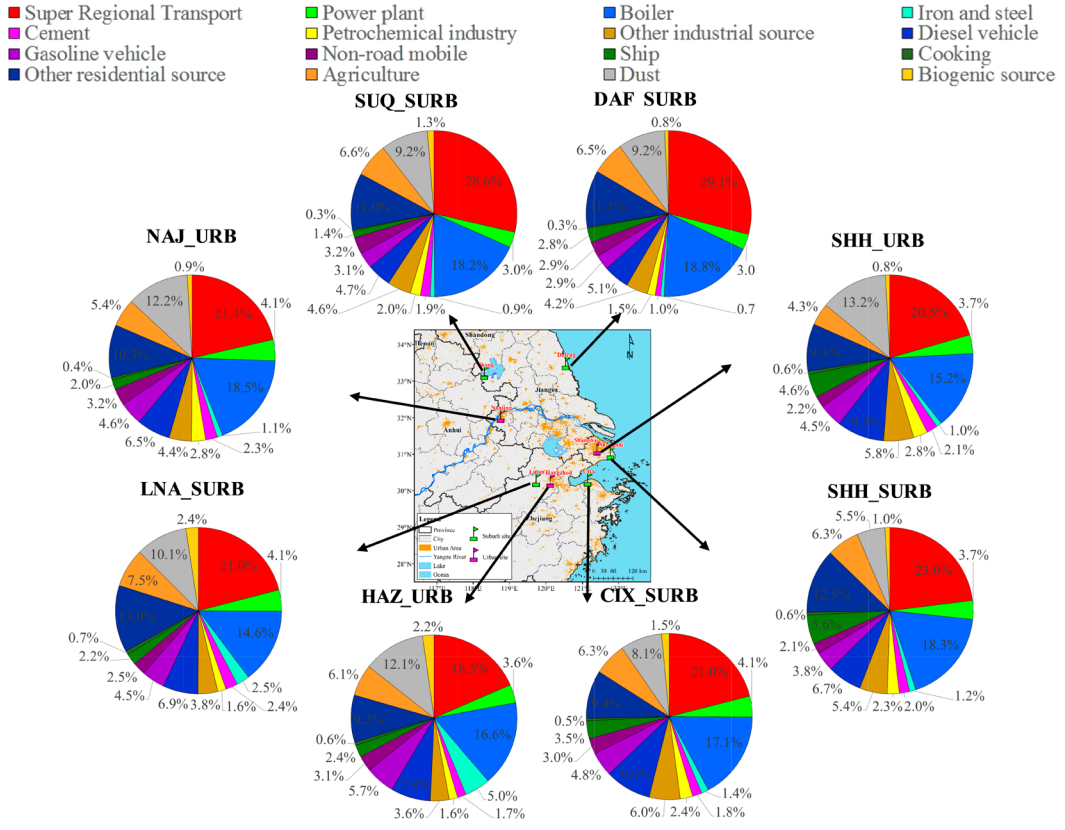

Figure 8. Contributing sources of $PM_{2.5}$ in YRD during winter at each offline sampling sites with the ROM-SOM integrated SA technique.

2.8−4.9%, 12.6−21.8%, 4.9−16.3%, 10.3−31.3%, 6.2−12.9%, and 3.9−8.4% respectively. SOM approach have produced an extensive spatial distribution profile for the emission sources with more detailed emission categories. The motor vehicle

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emission is broken down into diesel and gasoline vehicle while industries emission is broken down into power plants, cement, metallurgy, petrochemical, and other industries. More emission sources from regional influx, agricultural activities, biogenic emissions, are accounted to polish the ability of model to trace the sources of secondary aerosols. The integrative approach is able to enhance the source apportionment ability of the ROM and SOM models.

Integrated $PM_{2.5}$ Source Apportionment of YRD. With the integrated approach described in the previous section, the contributing sources to $PM_{2.5}$ in the YRD region during the winter season is shown in Figure 8. Result shows that the super-regional transport has contributed to 21.4−29.1% and 18.5−21.0% of the $PM_{2.5}$ emission at receptor sites in Jiangsu and Zhejiang respectively. It is then understood that the cross-boundary transportation has a greater effect on the northern side during the winter due to the dominant northwesterlies. It carries the pollutants from the upwind cities in northern China to the downwind cities on the southern side. Moreover, due to the generically higher amount of local pollution in cities, the cross-boundary transportation has greater contribution in the suburban regions.

The stationary point emissions have contributed an average of 18.8−22.6% of the $PM_{2.5}$ at the receptor point, with more significant contribution from industrial boiler (14.6% - 18.8%). Among the processing industry, the metallurgy, cement and petrochemical industries contributed 0.7−5.0%, 1.0−2.4%, and 1.5−2.8%, respectively at most receptor sites, while the remaining industries contribute around 3.6−6.0%. The gasoline, diesel nonroad mobile sources, ships, and other mobile sources have contributed around 12.4−20.6% in most receptor sites. The overall contributions are similar to the stationary point emissions. The diesel car contributes the most significant amount of 4.7−9.3% while the gasoline, nonroad mobile and ship sources has contributed to 2.9−5.7%, 2.1−3.2%, and 1.4−5.6%, respectively. The influence of diesel car is equivalent to in the suburban to the urban, with 4.7−9.0% to 6.5−9.3%. Gasoline car has a larger contribution in the cities compared to suburban, with 4.5−5.7% to 2.9−4.8%. Except the before mentioned emission sources, the agriculture is the major source of the NH_3 and NH_3 is the most direct source of the ammonium salt in $PM_{2.5}$. It contributes to around 4.3−7.5% over each receptor sites. Dust is one of the primary emission source of $PM_{2.5}$ and it contributes to around 5.5−13.2%.

The most commonly used methods to apportion sources of $PM_{2.5}$ have a number of limits and merits as well. Results from the application of different ROM methods are convincible since they are based on receptor observations, but with obvious limits: (1) the number of source types are limited, with generally 5−7 source categories; (2) the secondary aerosol cannot be apportioned to the source sectors; (3) local pollution and regional transport cannot be separated; (4) the results may vary a lot among different receptor models. The source oriented models (SOM) to apportion sources of $PM_{2.5}$ also have both limits and merits. SOM methodology can give detailed source contribution to both primary and secondary particles based on chemical weather prediction and emissions inventory. However, there exist certain uncertainty due to the uncertainty of emissions inventory, meteorology simulation, and chemical mechanism. In this study, we developed an integrated SA methodology, which combines two types of SA methods including ROM and SOM. The integrated methodology has improved detailed understanding of source apportionment compared to individual methods, and provides a larger number of source impact estimates. The methodology is further applied to do SA of $PM_{2.5}$ over the Yangtze River Delta region. Results indicate that super-regional transport has contributed 21.4%−29.1% to the cities in Jiangsu and 18.5%−21.0% to stations in Zhejiang, which indicates that the northern YRD region is more influenced by super-regional transport than southern area. Comparisons between the stations have found that the suburban region is more affected by the super-regional transportation compared to the cities, with 21.0−29.1% to 18.5−21.4%. It is mainly caused by the larger proportion of local pollution in the crowded and congested cities. Stationary combustion source including power plants and industrial boilers is the major source sector contributing to the ambient $PM_{2.5}$, with contributions ranging between 18.8 and 22.6% at different stations, among which industrial boilers contribute 14.6−18.8%, and industrial processing ranks second. Among the industrial processing, steal industry, cement, and petrochemical and chemical industry contribute 0.7−5.0%, 1.0−2.4%, and 1.5−2.8%, respectively. Contributions from mobile source including gasoline and diesel vehicle exhaust, off-road and ships account for 12.4−20.6%, which is generally close to stationary combustion source. Among the mobile source, diesel vehicle exhaust contributes 4.7−9.3%, followed by gasoline vehicle (2.9−5.7%), nonroad mobile (2.1−3.2%), and ships (1.4−5.6%). Urban stations are more affected by gasoline vehicles than rural sites. Agriculture is the major source of NH_3 and NH_3 is the most direct source of the ammonium salt in $PM_{2.5}$. It contributes to around 4.3−7.5% over each receptor sites. Road dust is one of the primary emission source of $PM_{2.5}$ and it contributes to around 5.5−13.2%. These results are compared with previous data and shows good agreement. It is found that this integrative approach is more comprehensive and is able to produce a more profound spatial and temporal understanding between the sources and the receptors.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.8b01211.

The measured $PM_{2.5}$ mass concentrations and its chemical compositions, the comparison of measured and simulated $PM_{2.5}$ by receptor models, source profiles, and equations for PMF, CMB and ME2 (Tables S1−S3, Figure S1, and eq S1) (PDF)

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Notes

The authors declare no competing financial interest.

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