Response Letter

Atmospheric Chemistry and Physics

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Long–term Trends in $PM_{2.5}$ Chemical Composition and Its Impact on Aerosol Properties: Field Observations from 2007 to 2020 in Pearl River Delta, South China

We sincerely thank reviewers for your time and constructive comments. We have carefully revised the manuscript to improve its clarity and enhance the readers' understanding. Our point-by-point responses are marked in blue and the corresponding changes to the original text are shown below each response. We hope that these revisions adequately address the comments and concerns.

Anonymous Referee #1

General comments

In this work, the authors examine the 2007 – 2020 trends in PM2.5 and its composition in the Pearl River Delta area of China. This time period saw dramatic decreases in PM2.5 concentrations and changes in the PM2.5 composition driven by successful regulatory actions. These air quality and associated emission trends have been discussed by others; however, in this work the authors explore the causes of the trends including changes in the oxidation rates of SO_2 and NO_2 to sulfate and nitrate respectively. I think this is an important contribution to our understanding of the aerosol in this region and the underlying causes of their trends. I also found no major technical issues in the work and recommend publication after the authors address a number of minor comments.

Comments

1) Properly define $PM_{2.5}$.

Response: Thanks for reminding. We change it as "particulate matter with aerodynamic diameter less than 2.5 μm" in the introduction.

Line 40 – 41

"Particulate matter with aerodynamic diameter less than 2.5 μm ($PM_{2.5}$) is a major air pollutant with significant implications for global climate, air quality, and human health (Burnett et al., 2018; Chen et al., 2021; Ding et al., 2021; Pye et al., 2021; Vohra et al., 2022)."

2) Define ALWC.

Response: We have added definition of ALWC as "aerosols liquid water content" in the abstract.

3) The IMPROVE equation was developed by the US National Park Service with some support from the EPA. There are two IMPROVE equations. The first, IMPROVE equation 1 (EPA, 2003), was based on the work in Malm et al., (1994). This was replaced in 2007 with the IMPROVE equation 2 (Pitchford et al., 2007) based on the work of Malm et al., (2007) and Hand et al., (2007). The

IMPROVE equation 1 uses constant scattering efficiencies base on fixed size distributions for the different aerosol components. In IMPROVE equation 2 the scattering efficiencies are dependent on the aerosol concentrations. Specifically, the scattering efficiencies are a weighted average of the scattering efficiency derived from a small and large size distribution and the weights are proportional to the aerosol concentration (Pitchford et al., 2007). Which IMPROVE equation is used in this work is not discussed and needs to be clarified.

Response: Thanks for providing information and valuable references for IMPROVE equations. It is important to clarify which equation was used in this study. Here, we used the revised IMPROVE equation proposed in 2007. We have added introduction about them clarified which one was used in discussion.

Line 77 – 81

"To estimate the light extinction coefficient (b_{ext}), the first Interagency Monitoring of Protected Visual Environments (IMPROVE) equation was developed by the U.S. National Park Service with support from the U.S. Environmental Protection Agency (EPA) (Malm et al., 1994; EPA, 2003), but this equation tended to underestimate (overestimate) the highest (lowest) b_{ext} values. Consequently, the revised IMPROVE equation was then proposed (Malm and Hand, 2007; Pitchford et al., 2007)."

Line 444 – 445

"We also calculated b_{ext} by the revised IMPROVE equation proposed in 2007 (Malm and Hand, 2007; Pitchford et al., 2007), and compared to the local parameter scheme (Fig. S21)."

4) "The hygroscopic growth factor (f(RH)), which has been suggested to depend on secondary inorganic fractions (e.g., sulfate, nitrate, and ammonium), sea salt components, and water-soluble organic carbon, is solely a function of relative humidity (RH) in the algorithm" If IMPROVE equation 2 is being used then this is an incorrect statement.

Response: Thanks for comments. We used IMPROVE equation 2, which is the revised IMPROVE equation proposed in 2007 in this study. The hygroscopic growth factor in the equation is not only a function of relative humidity but also particle size distribution (or mass concentration). We have corrected our statement.

Line 83 – 85

"In addition, the calculation of hygroscopic growth factor (f(RH)) in the revised equation depends on relative humidity (RH) and particle size distribution (or aerosols mass), but does not account for the chemical composition in aerosols, which has been shown to significantly affect f(RH) (Li et al., 2021)."

5) "IMPROVE program in the United States, initiated in 1985, tracks visibility trends and their driving factors (Epa, 2011)". The EPA reference is not in the reference list. There are many journals article reporting on the purpose of the IMPROVE program and use of the data and should be used

as the reference instead of an EPA report. For example, see Hand et al., (2024) and references there in.

Response: We apologize for the error in citation. We have changed it to journal article.

6) "anions (.e. Cl-, NO3- and SO42-) were analyzed with an ion-chromatography system…" The authors should note that some ammonium nitrate volatilizes from the quartz fiber filters during sampling and handling causing underestimations in NH4+ and NO3- concentrations (Yu et al., 2006). In addition, if possible, provide an estimate of the underestimation, which should be carried over into the discussion of particulate nitrate concentrations.

Response: We acknowledge that the volatilization of ammonium nitrate can lead to negative bias in measurements of NO_3^- and NH_4^+ and have included relevant discussion into Section 2.2 and 3.2.2. According to previous studies, this volatilization may result in underestimations of 8%–16% for NO_3^- and 10%–28% for NH_4^+. Because our measurements were conducted in winter, the relatively lower temperature and relative humidity did not favor the volatilization, thereby reducing the extent of underestimation. In addition, the measurements were conducted in the same season and such losses are expected to be systematic over time. Consequently, it would not significantly influence the long-term trends in NO_3^- and NH_4^+.

Line 158 – 159

"Due to the negative mass artifacts associated with the volatilization of ammonium nitrate, the measured concentrations of NO_3^- and NH_4^+ may be underestimated (Chow et al., 2005; Yu et al., 2006)."

Line 305 – 309

"Previous studies reported that the volatilization of ammonium nitrate during sampling can cause negative mass artifacts, leading to the underestimation of both NO_3^- (8%–16%) (Chow et al., 2005) and NH_4^+ (10%–28%) (Yu et al., 2006). The volatilization is highly dependent on relative humidity and temperature. However, such losses are expected to be systematic over time and therefore are unlikely to significantly affect the general trends in this study, because our measurements were conducted in the same season."

7) Measuring long-term trends is very challenging, and seemingly, small changes in sampling and analysis protocols can introduce discontinuities in the PM trends. This is particularly true for thermal optical carbon analyses, since the measured OC and EC are operationally defined and sensitive to changes in the method. The authors should note any changes in the monitoring protocols over the 14-year time span and discuss any evidence for or against these changes introducing discontinuities in the trends.

Response: In this long-term observation, we consistently employed the same measurement instruments and analytical protocols. Field and blank samples were analyzed in the same way as field samples. All the OC/EC and cation/anion data were corrected using the field blanks. We added

Fig. S1 in supplement to illustrate that all measured components did not exhibit large variability in the blank filter samples. This indicated that the influence of analytical or sampling biases related to blank subtraction and experimental procedures was limited, further supporting the reliability of the long-term trends observed in this study. We add this statement in QA/QC section.

Line 170 – 173

"Blank samples were analyzed in the same way as field samples. All the OC/EC and cation/anion data were corrected by subtracting the field blank samples. As shown in Fig. S1, all measured components exhibited minimal variability in the blank filter samples. This indicated that the influence of analytical and sampling biases related to blank subtraction and experimental procedures was limited, further supporting the reliability of the long-term trends observed in this study."

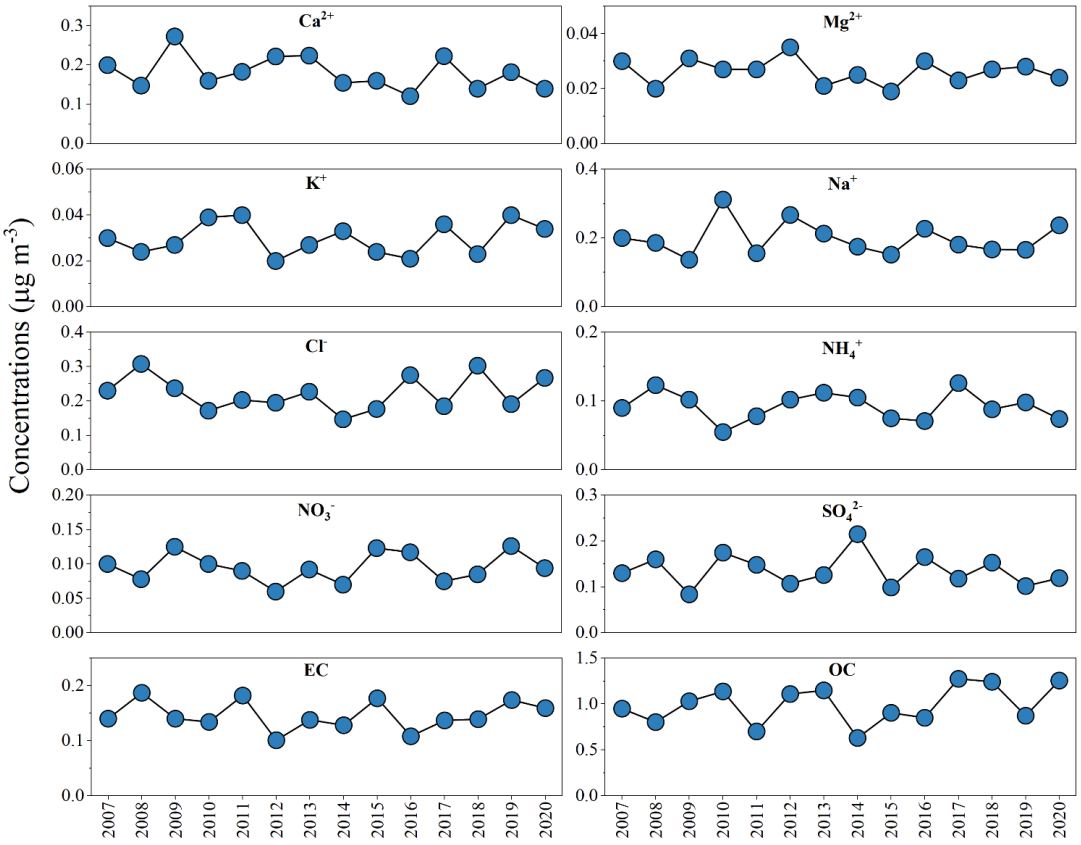

Figure S1. Annual variations of measured components in blank filter samples.

8) "Bayesian Inference Approach and suggested it had significant advantages in accurately estimating POC and SOC". All of methods to estimate POC and SOC from OC and EC data are highly uncertain. This should be conveyed in the paper. For example, instead of saying "suggested it had significant advantages in accurately estimated..." could use "suggested it more accurately estimated…"

Response: Thanks for correcting that. We should deliver that all methods to estimate POC and SOC will introduce uncertainty.

Line 194 – 195

"Liao et al. (2023) proposed Bayesian Inference (BI) approach and suggested it more accurately estimated POC and SOC compared to the conventional method."

9) The annual bar chart in Figure 2 provides the change in the absolute concentrations overtime. It is difficult to see the trends in the changing PM2.5 composition. I suggest the authors include a graph similar to Figure 2a of the annual relative contributions of aerosol components to PM2.5 in the main document or supplemental information.

Response: Thanks for suggestion. This graph will make changes in mass fractions of chemical composition more visible. We have added the similar graph into supplement.

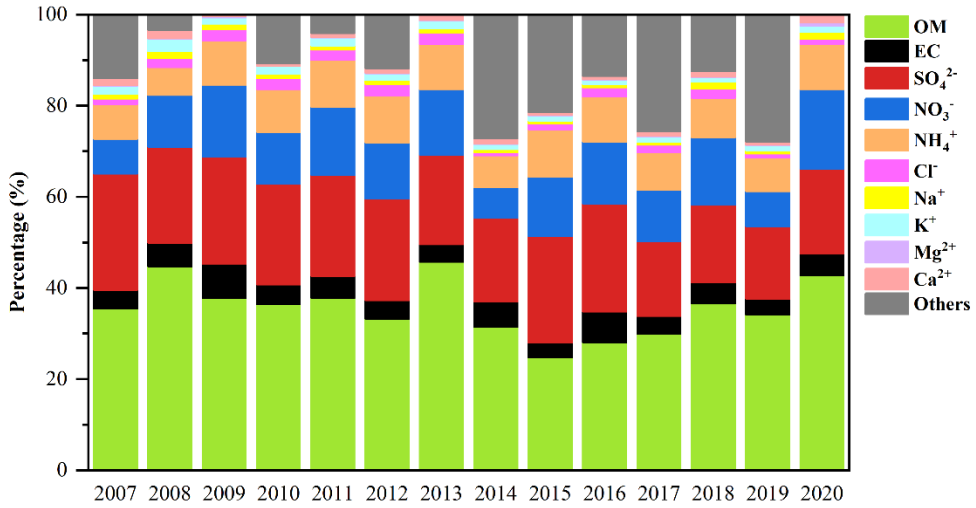

Figure S6. The changes in percentage of $PM_{2.5}$ chemical composition.

10) In Figure 2, could some indication of the SOC and POC concentrations be included?

Response: We considered that OM = conversion factor 1 × POC + conversion factor 2 × SOC, or OM = POA + SOA. It is unreasonable to replace OM with POC and SOC. If we used POA and SOA to replace OM, it will introduce some uncertainties because of the selection of conversion factor 1 and 2. Here, we want to highlight the changes in OM. In addition, we have presented the variations in POA and SOA in supplement (Fig. S8-9), which could indicate the changes in POC and SOC. In addition, we add the detailed information about POC and SOC into Table S1 in supplement.

11) "Despite the slight decline in their concentrations, our result showed that the relative reductions of EC (-9% yr-1)…" This is confusing. What is meant by "slight decline"? A 9% decrease per year is not slight.

Response: We apologize for this misunderstanding. What we want to deliver is that the reduction in absolute concentration (–0.97 μg m^{-3} yr^{-1}) was slight. It will lead to misunderstand and is irrelevant with the main point so we changed our statement.

Line 294 – 296

"Our result showed that the relative reductions in EC (–9% yr^{-1}), K^+ (–12% yr^{-1}) and Ca^{2+} (–11% yr^{-1}) were greater than that of $PM_{2.5}$ (–7% yr^{-1})"

12) "When NO2 levels are low, the accumulation of nitrate is hindered due to volatilization losses." This is a confusing sentence. What are the volatilization losses? Those from the filter? Also, ammonium nitrate volatilization from the filter is not really dependent on NO2 levels, but is dependent on temperature and relative humidity.

Response: Thanks for pointing this confusing statement. The "volatilization losses" should be put as "partitioning of nitrate from particle phase to gas phase". Here, we want to discuss that the lower intercepts observed in the NO_3^-/NO_2 regression compared to those in the SO_4^{2-}/SO_2 regression. When NO_2 level is low, the formation of HNO_3, gaseous precursor of NO_3^- is suppressed. Therefore, the reaction in R2 tends to proceed to the left. This indicates that more NO_3^- will partition into gas phase, leading to less NO_3^- accumulates in particle phase.

$$OH\ (g) + NO_2\ (g) + M \rightarrow HNO_3\ (g) + M \quad (R1)$$

$$HNO_3(g) + NH_3\ (g) \leftrightarrow NO_3^-(aq) + NH_4^+(aq) \quad (R2)$$

Line 312 – 316

"The generally lower intercepts observed in the NO_3^-/NO_2 regression compared to those in the SO_4^{2-}/SO_2 regression can be explained by the semi-volatile nature of nitrate (Yu et al., 2006). The formation of HNO_3, gaseous precursor of NO_3^-, is suppressed under very low NO_2 level. Therefore, the reaction in R2 tends to proceed to the left. This facilitates partitioning of NO_3^- into gas phase, leading to less accumulation of NO_3^- in particle phase."

13) "As shown in Fig 5a, a dramatic increase in SOR was observed during 2007–2020 ($p < 0.05$). The SOR value in 2020 (0.24 ± 0.09) was twice as high as that in 2008 (0.12 ± 0.07)". Comparing 2008 to 2020 is a bit of cherry picking. There is a lot of variability in the data and 2007 SOR are only 30-40% smaller than 2020. I suggest a robust trend line, e.g. use Theil regression, is calculated from the data, and then use the slope of the trend line to estimate the change over time.

Response: Thanks for pointing out our mistake. We double checked the result and found that an extreme point in 2020 was included when we conducted average calculation, which was not included in the plot. We have corrected it. Comparing 2008 to 2020 is not very reasonable. Instead, we compared the beginning point (2007) with the end point (2020). We used the slopes derived from Theil-Sen regression in to represent the change rates of different species in this study, and we added this clarification in QA/QC section. A robust line was also added into the SOR plot.

Line 175 – 177

"The change rates were calculated using the slopes derived from Theil–Sen regression and evaluated for statistical significance via the non-parametric Mann–Kendall test, providing a robust and reliable assessment of temporal variations."

Line 331 – 332

"The SOR value in 2020 (0.26 ± 0.09) was 44% higher than that in 2007 (0.18 ± 0.07). In general, SOR exhibited a significant upward trend during 2007–2020, increasing at a rate of 0.005 yr^{-1} (3% yr^{-1}, $p < 0.05$)."

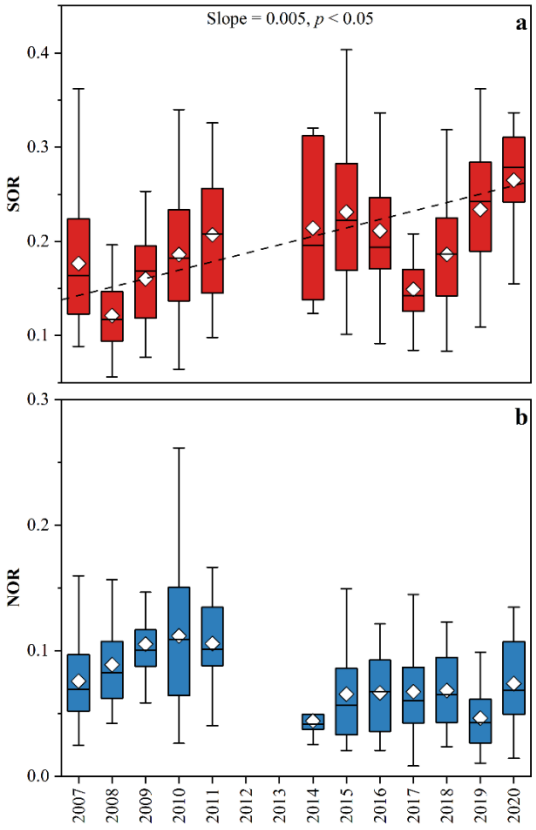

Figure 5. The variations in SOR (a) and NOR (b). A dramatic increase in SOR was observed (0.005 yr^{-1}, $p < 0.05$) and the SOR value in 2020 (0.26 ± 0.09) was 44% higher than that in 2007 (0.18 ± 0.07). Although there was no significant trend in NOR, the values before 2012 were higher than those after 2013.

14) This meant that the conversion of NO2 to NO3- became weaker, resulting in a greater reduction in NO3- compared to NO2." This is not obvious to me. How do you know that the difference in the trends is not driven by changes in the partitioning of NO3 between the gas and particle phase?

Response: Our statement here was not precise. To make it more structure and logical, we combined the results of correlation analysis to illustrate that the lower NOR was caused by both weaker heterogeneous formation pathway of nitrate and enhanced partitioning of nitrate from particle phase into gas phase.

Line 363 – 366

"The largest regression coefficient and the strongest correlation between ALWC and NOR suggested that the change in NOR was primarily driven by ALWC. The lower ALWC levels after 2013 (Fig. 6b) suppressed heterogeneous formation pathway of nitrate and enhanced the partitioning of nitrate from particle phase into gas phase, leading to the overall lower NOR after 2013."

15) "Aerosol pH increased from 1.51 ± 1.07 to 3.29 ± 1.43, at a rate of 0.06 yr–1 ($p < 0.05$)." In figure 6a, the largest pH is about 2.7 not 3.29. Also, I do not see a trend in these data. Similar to the suggestion for figure 5, it would be best to fit a robust trend line through the data and use its slope to define the change over time.

Response: Thanks for pointing out our mistake. Same as mentioned in response 13, there was an extreme point which was not excluded in calculation and we have corrected that. The pH value in 2020 should be 2.86 ± 0.49. In addition, we also derived a robust line into the graph, with the slope derived from Theil-Sen regression.

Line 397 – 398

"Aerosol pH increased from 1.51 ± 1.07 to 2.86 ± 0.49, at a rate of 0.04 yr^{-1} ($p < 0.05$)."

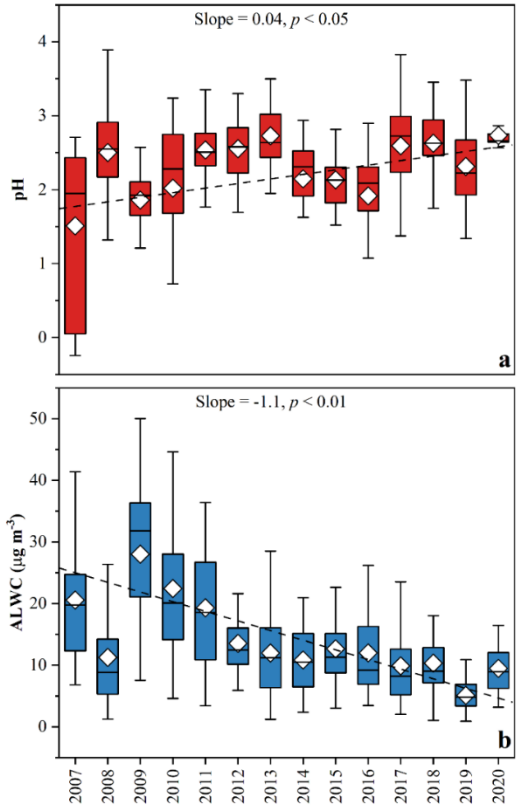

Figure 6. (a) Annual average pH increased at a rate of 0.04 yr^{-1}. (b) Annual average ALWC decreased at a rate of –1.1 μg m^{-3} yr^{-1}.

16) Which IMPROVE equation is being used to estimate the light extinction?

Response: We add clarification before comparing the result.

Line 444 – 445

"We also calculated b_{ext} by the revised IMPROVE equation proposed in 2007 (Malm and Hand, 2007; Pitchford et al., 2007)"

17) "Our results indicated that the IMPROVE equation tend to overestimate bext in elevated

pollution periods." The authors did not provide any evidence that one bext equation was better than the other was, so they should only say that the IMPROVE equation had higher bext than that estimated from equations 5-8 for high PM2.5 concentrations. This is important, because many studies have shown that scattering efficiencies of secondary aerosols are correlated with concentrations. It was this observation that drove the development of the IMPROVE equation 2. Therefore, it is quite possible that the fixed scattering efficiencies in equation 5 would cause an underestimation of the bext.

Response: Due to lack of real measurements of b_{ext}, it unreasonable to say the revised IMPROVE equation tend to overestimate b_{ext} in elevated pollution periods. Instead, we can only conclude that b_{ext} estimated by the revised IMPROVE equation was significantly higher than that estimated by local parameter scheme. We have changed the statement.

Line 445 – 446

"b_{ext} estimated by the revised IMPROVE equation (335.72 ± 219.64 Mm^{-1}) was significantly higher than that estimated by local parameter scheme (262.67 ± 143.82 Mm^{-1})."

Anonymous Referee #2

General comments

This work presents long-term data on PM2.5 chemical composition for a regionally representative site in the Pearl River Delta region of China. The authors report decreases in PM2.5 and its chemical constituents. They find an increase in secondary species. They add an analysis investigating the sulfur and nitrogen oxidation rate and find that sulfur oxidation rates increase while nitrogen oxidation rates decrease. Other physicochemical properties are assessed.

There are many studies already published that are similar to the one presented here, and there is not much new information. The novel part of this study is the investigation into sulfur and nitrogen oxidation rates, and for that reason, I think this paper is of value to add to the literature. I have some concerns that should be addressed before I can recommend it for publication.

Comments

1) There is a lack of placing into context the findings in this study to other prior studies that have found similar phenomena. Specifically, I noted that the discussion of the impact of aerosol water on the organic aerosol species lacks many references to other studies that initially identified aerosol water as a major influencing factor on organic aerosol concentrations, some of which are from over 10 years ago. This is especially true in the paragraph starting in Line 302 and Section 3.3.

Response: Thanks for reminding this and providing valuable literature. It is important to discuss the previous studies with similar findings, which can also support the conclusion of our study. We have added the related discussion and cited these references in the Introduction, Section 3.2.2 and 3.3.

Line 59 – 60

"Furthermore, Attwood et al. (2014) reported that the changes in ALWC significantly influenced the aerosol light extinction and radiative forcing."

Line 377 – 383

"Previous studies have demonstrated that ALWC is a key factor driving the partitioning of organic compounds from the gas phase into the particle phase, thereby promoting SOA formation (Ervens et al., 2011; Carlton and Turpin, 2013; Attwood et al., 2014). Nguyen et al. (2015) observed concurrent decreasing trends in ALWC and OC in the Southeast U.S., and further suggested that anthropogenic inorganic species modulated SOA formation through ALWC effects. In addition, higher aerosol acidity has been shown to enhance SOA formation via acid-catalyzed reactions (Surratt et al., 2007). These findings indicated that the reduction in SOA during our study period aerosol could be attributed to the changes in acidity and ALWC, which will be discussed in Sect. 3.3."

Line 401

"The rapid reduction in hygroscopic components, especially SO_4^{2-}, led the decline of ALWC (Attwood et al., 2014)."

Line 410 – 412

"Many studies have demonstrated that high aerosol acidity, ALWC, and O_3 will facilitate SOA formation (Ervens et al., 2011; Carlton and Turpin, 2013; Nguyen et al., 2015; Zhang et al., 2022; Ma et al., 2024; Zhang et al., 2024)."

2) I noted that the timeframe of the study includes 2020, yet there is no acknowledgement of the potential impacts of this anomalous year on the long-term trend. The authors need to at least make note of the fact that large emissions changes during 2020 may impact the trend, and where possible, quantify the uncertainty that brings to their analysis.

Response: Thanks for this comment. Previous studies have reported a dramatic decline in anthropogenic pollutants during the COVID-19 lockdowns. Therefore, it is necessary to evaluate the impact of this anomalous period on the trends of $PM_{2.5}$ and its chemical composition. We used the slopes derived from Theil–Sen regression to represent the change rates for different components, and performed a sensitivity analysis of long-term trends with and without the year 2020. The estimated uncertainties ranged from 1% to 20% (see below, Table S2), indicating that including this anomalous year in the trend analysis would not introduce large bias on the overall long-term trends. We showed the sensitive analysis results in supplement and clarified it in the revised manuscript (Section 3.1).

Line 242 – 246

"It is worth noting that the year 2020 was characterized by unprecedented emission reductions associated with COVID-19 lockdowns (Wang et al., 2021), which may have temporarily affected the trends in $PM_{2.5}$ and its chemical composition. As shown in Table S2, a sensitivity analysis was conducted to evaluate the uncertainty introduced by including 2020 in the long-term trends analysis. The results suggested that this anomalous year would not introduce large bias on the overall long-term trends."

Table S2. Sensitivity analysis of long-term trends with and without the year 2020.

	Slope (with 2020)	Slope (without 2020)	Uncertainty
$PM_{2.5}$	-4.0 **	-4.2 **	4%
OM	-1.70 **	-1.91 **	12%
EC	-0.23 **	-0.25 **	9%
SO_4^{2-}	-1.13 **	-1.21 **	7%
NO_3^-	-0.40 **	-0.47 **	18%
NH_4^+	-0.31 **	-0.33 **	6%
Cl^-	-0.10 **	-0.10 **	1%
Na^+	-0.05 **	-0.06 **	20%
K^+	-0.10 **	-0.12 **	16%
Mg^{2+}		-0.01 *	
Ca^{2+}	-0.06 *	-0.06 *	3%

One asterisk, two asterisks denote p value < 0.05, 0.01, respectively. Blank cells denote p value > 0.05. The uncertainty was calculated as: $\boldsymbol{Uncertainty} = \frac{|Slope_{with} - Slope_{without}|}{|Slope_{with}|}$.

3) "EC is a product of carbon fuel-based combustion processes and is exclusively associated with primary emission…" I am a bit confused on this paragraph. Are the authors referring to the EC measured by the OC/EC analyzer? If so, then this statement needs more clarification and discussion of uncertainties. Instrument-measured OC and EC are determined optically, and are thus more operational definitions than true determinants of carbon sources. The cutoff temperatures between OC and EC varies by instrument type and network protocol followed, and errors for the cutoff tend to be large. The authors should specify what temperatures were used for their analysis, and quantify where possible the uncertainty associated with their chosen protocol.

Response: We thank the reviewer for raising this important point regarding the definition and measurement of OC/EC and providing valuable references. To minimize potential measurement biases and enhance data comparability, we used the same thermal-optical carbon analyzer and followed the same analytical protocol throughout the study. We have added detailed information of measurement procedure and discussion of uncertainties in the Methodology (Section 2.2 and 2.3).

Line 144 – 154

"Because the determination of OC and EC are highly sensitive to analytical conditions, different thermal-optical methods may lead to discrepancies in the OC/EC measurements (Khan et al., 2012; Giannoni et al., 2016). To minimize potential measurement biases and enhance data comparability, we used the same thermal-optical carbon analyzer and followed the same analytical protocol throughout the study. Specifically, the OC and EC were

determined by the thermal–optical transmittance (TOT) method (NIOSH, 1999) using an OC/EC analyzer (Sunset Laboratory Inc., USA), with a punch (1.5 × 1.0 cm) of the sampled filters. The samples were analyzed by stepwise heating. First, the sample was heated sequentially to 870 °C (310 °C for 60 s, 475 °C for 60 s, 620 °C for 60 s, and 870 °C for 90 s) under a pure helium (He) atmosphere. OC was volatilized and a portion of it underwent pyrolysis, forming pyrolyzed carbon during this period. After cooling, the sample was reheated under a 2% O_2/He atmosphere up to 920 °C (625 °C for 30 s, 700 °C for 30 s, 775 °C for 30 s, 850 °C for 30 s, and 920 °C for 30 s) to oxidize EC and pyrolyzed carbon."

Line 164 – 165

"Prior to OC/EC analysis, we calibrated the instrument using glucose standards at multiple concentrations. The instrument responses were highly linear ($R^2 > 0.99$) and the relative errors between measured and prepared concentrations were within ± 5%."

4) Please provide more details on the EC-tracer method and how it differs from your Bayesian Inference Approach. In addition, please describe the Bayesian approach in more detail. What is the significance of the K values?

Response: The EC-tracer method assumes that elemental carbon (EC) originates from primary combustion sources and uses a fixed $(OC/EC)_{pri}$ ratio to estimate primary organic carbon (POC), from which secondary organic carbon (SOC) is obtained by subtracting POC from total OC. Although a few methods have been proposed to estimate the $(OC/EC)_{pri}$, such as minimum (OC/EC) and minimum R squared method, the choice of this $(OC/EC)_{pri}$ value is often arbitrary and could introduce significant uncertainties. Bayesian Inference Approach adopts a probabilistic framework that combines prior knowledge (in the form of prior distributions of the K values) with observational data (OC, EC, and secondary inorganic aerosol species like sulfate) to estimate the source contributions to OC. The Bayesian Inference Approach assumes that observed OC is a linear combination of contributions from POC and SOC tracers (e.g., EC and SO_4^{2-}), and it uses the Markov Chain Monte Carlo (MCMC) technique to derive posterior distributions for the K values. This probabilistic treatment allows the model to update parameter estimates based on actual measurements. The K values represent the proportionality constants that link OC to its respective tracers (e.g., K_{EC} for EC-to-POC and K_{SO42-} for sulfate-to-SOC). We have added related introduction into section 2.4.

Line 183 – 192

"Given that EC is emitted exclusively from primary combustion sources (e.g., fossil fuel and biomass burning), it is commonly used as a tracer for POC. Under this assumption, POC is estimated by multiplying EC by a representative primary OC/EC ratio, and SOC is determined as the residual between total OC and estimated POC (eq. 1–2). One of the most commonly used approaches to determine $(OC/EC)_{pri}$ is the minimum OC/EC ratio approach (Castro et al., 1999), which assumes that the lowest observed OC/EC value corresponds to conditions

dominated by primary emissions with negligible SOC formation. In addition, Pio et al. (2011) recommended using the 5% percentile of observed OC/EC values instead, and Wu and Yu (2016) proposed minimum R squared (MRS) method to obtain $(OC/EC)_{pri}$."

$$POC = (\frac{OC}{EC})_{pri} \times EC \quad (1)$$

$$SOC = OC - POC \quad (2)$$

Line 195 – 200

"The BI approach adopts a probabilistic framework that combines prior knowledge (in the form of prior distributions of the K values) with observational data (OC, EC, and SIA) to estimate the source contributions to OC. The BI model assumes that observed OC is a linear combination of contributions from POC and SOC tracers (e.g., EC and SO_4^{2-}), and it uses the Markov Chain Monte Carlo (MCMC) technique to derive posterior distributions for the K values. This treatment allows the approach to update parameter estimates based on actual measurements and offers more flexibility."

Line 205 – 206

"The *K* values represent the proportionality constants that link OC to its respective tracers (e.g., K_{EC} for EC-to-POC and K_{SO42-} for sulfate-to-SOC)."

5) It is unclear why the conversion from SOC to SOA is needed.

Response: We appreciate the reviewer's comment. The conversion from secondary organic carbon (SOC) to secondary organic aerosol (SOA) is necessary because SOC represents only the carbonaceous portion of the organic aerosol, while SOA includes the entire mass of organic compounds formed through secondary processes, including associated non-carbon atoms (e.g., hydrogen, oxygen, nitrogen). To better evaluate the atmospheric mass loading of organic aerosols and their mass proportions in $PM_{2.5}$, it is important to quantify SOA in terms of mass concentration ($\mu g\ m^{-3}$), not just carbon content. A conversion factor (commonly referred to as the organic mass to organic carbon ratio) is thus applied. In this study, we adopted a factor of 2.4 based on previous literature (Yan et al., 2020), which reflects the higher oxidation state of SOA formed under intense photochemical activity in the PRD region. This factor accounts for the added oxygenated functional groups in aged aerosols and is consistent with values used in other field studies in similar environments.

Line 207 – 210

"Because SOC represents only the carbonaceous portion of the organic aerosol, while SOA includes the entire mass of organic compounds formed through secondary processes, including associated non-carbon atoms (e.g., hydrogen, oxygen, nitrogen). To better evaluate the atmospheric mass loading of organic aerosols and their mass proportions in $PM_{2.5}$, it is important to convert SOC to SOA."

6) The authors refer to annual average PM2.5 concentrations, but earlier state that most

measurements were done from October to December. Are the concentrations shown here true annual averages, or are they the averages from October to December (wintertime)? How many of the samples fall outside this October to December range?

Response: We thank the reviewer for the comment. Indeed, all of the samples in this study were collected during the October to December, which corresponds to the winter season. Therefore, the $PM_{2.5}$ concentrations and chemical compositions presented in this paper should be interpreted as representative of the wintertime conditions rather than true annual averages. We acknowledge this seasonal limitation and have revised the relevant descriptions in the revised manuscript to avoid confusion.

7) There are several undefined acronyms (POA, SOA, SIA) – this is true in other areas of the manuscript as well (ALWC, SOR, NOR). These need to be defined at their first usage.

Response: We thank the reviewer for pointing this out. We have carefully reviewed the entire manuscript and ensured that all acronyms are defined at their first occurrence.

8) Where the authors state that the Bayesian Inference approach is more reliable. On what basis are you making this claim? To what are you comparing the approaches to determine reliability?

Response: We thank the reviewer for pointing this out. We conducted correlation analysis between oxalic acid (a typical and abundant organic secondary molecular marker) and SOC estimated by Bayesian Inference approach, as well as SOC estimated by EC-tracer method. The result showed that the correlation between oxalic acid and SOC (BI approach) ($r = 0.62$, $p < 0.05$) was stronger than correlation between oxalic acid and SOC (EC-tracer method) ($r = 0.54$, $p < 0.05$), which indicated the estimation from BI approach was more reliable. We showed this in Fig. S5 in supplement but not mentioned in manuscript, which could lead to confusion. We have added related description in the revised manuscript.

Line 274 – 276

"In addition, the correlation between oxalic acid (a typical secondary organic molecular marker) and SOC estimated by the BI approach ($r = 0.62$, $p < 0.05$) was stronger than that with SOC estimated by the EC-tracer method ($r = 0.54$, $p < 0.05$)"

9) What control measures have been put in place for biomass burning and dust?

Response: We thank the reviewer for the valuable question. In the past decades, several control measures have been implemented in the Pearl River Delta (PRD) region to reduce emissions from biomass burning and dust sources. The main measures and relevant policy frameworks are summarized below:

Control category	Main Measures	Sources

Biomass burning	Based on the types and distribution of crop straw, an integrated plan for comprehensive straw utilization has been formulated and steadily implemented. The focus is placed on strictly banning open burning and promoting the comprehensive utilization of surplus straw. These measures aim to enhance the efficient use of straw resources and address the issues of resource waste and environmental pollution caused by improper disposal and illegal burning.	2
	Accelerate the establishment and improvement of policies and mechanisms for the comprehensive utilization of biomass waste, and effectively control the uncontrolled burning of biomass waste in rural areas and around urban fringes.	6
	Open burning is strictly prohibited, and the responsibility for straw burning control must be rigorously implemented at the municipal, county, and township government levels. Effective measures should be taken to strengthen the supervision and management of straw burning bans.	8
Dust	Strengthen comprehensive control of urban fugitive dust throughout the entire process. In each city, designated dust control zones should cover more than 80% of the built-up area, and dust pollution must be effectively managed. Dust control at material storage and handling sites must be enhanced. Facilities storing or piling dust-prone materials such as coal, gangue, cinder, fly ash, sand, and soil should be equipped with enclosed structures, spraying systems, and surface solidification measures. At construction sites, strict implementation of enclosure measures, proper disposal of construction waste, and water spraying for dust suppression is required.	1, 3
	Strengthen the control of dust pollution from construction activities and roadways. Promote the application of dust suppression technologies at construction sites, and establish dynamic databases of dust sources along with online monitoring systems for particulate matter. Actively promote green construction practices by requiring construction units to implement measures such as site enclosure, installation of vehicle washing facilities, and road pavement hardening, while strictly prohibiting open-air operations. In the main urban areas of each city, the transportation of construction waste and powdery materials should gradually be carried out in enclosed vehicles equipped with satellite positioning systems.	4
	Strengthen the regulation of spillage from vehicles transporting construction waste, earth and rock, and industrial raw and auxiliary materials in urban areas. Enclosed transport vehicles or tightly covered truck beds should be used, and all transport activities must follow designated routes and time schedules. Road cleaning practices should be improved by promoting standardized operations and increasing the rate of mechanized street sweeping and water spraying. The mechanized cleaning rate of roads in built-up urban areas should exceed 85%.	5, 7
	All prefecture-level and above cities are required to establish citywide online monitoring and management platforms for construction site dust emissions. The use of fully enclosed vehicles for transporting construction waste and powdery materials should be promoted, and the phasing out and replacement of outdated transport vehicles is encouraged.	8

No.	Release year	Main documents
1	2005	Notice on the Issuance of the “Pearl River Delta Environmental Protection Plan Outline (2004–2020)”
2	2008	Circular on Forwarding the Opinions of the General Office of the State Council on Accelerating the Comprehensive Utilization of Crop Straw
3	2010	Notice on the Issuance of the “Integrated Environmental Protection Plan

		for the Pearl River Delta (2009–2020)"
4	2014	Notice of the Guangdong Provincial People's Government on Issuing the Guangdong Province Air Pollution Prevention and Control Action Plan (2014–2017)
5	2016	Notice of the Guangdong Provincial Department of Environmental Protection on Issuing the "13th Five-Year Plan for Environmental Protection in Guangdong Province"
6	2016	Notice of the Guangdong Provincial Department of Environmental Protection on Issuing the "13th Five-Year Plan for Environmental Protection in Guangdong Province"
7	2017	Notice of the General Office of the Guangdong Provincial People's Government on Issuing the Enhanced Measures and Task Allocation Plan for Air Pollution Prevention and Control in Guangdong Province
8	2018	Notice of the Guangdong Provincial People's Government on Issuing the "Guangdong Province Blue Sky Protection Campaign Implementation Plan (2018–2020)"

10) Nitrate accumulation would be more impacted by temperature than NO2 concentrations.

Response: Thanks for pointing this. We acknowledge that nitrate accumulation is strongly influenced by temperature and relative humidity, and we have clarified this point in the revised manuscript. In addition, we have included an estimate of the potential underestimation caused by the volatilization of ammonium nitrate. Here, we want to discuss the differences between intercepts observed in the NO_3^-/NO_2 regression and those in SO_4^{2-}/SO_2 regression. When NO_2 level is low, the formation of HNO_3, gaseous precursor of NO_3^- is suppressed. Therefore, the reaction in R2 tends to proceed to the left. This indicates that more NO_3^- will partition into gas phase, leading to less NO_3^- accumulates in particle phase. Thus, the intercepts observed in the NO_3^-/NO_2 regression were lower.

$$OH\ (g) + NO_2\ (g) + M \rightarrow HNO_3\ (g) + M \quad (R1)$$

$$HNO_3(g) + NH_3\ (g) \leftrightarrow NO_3^-(aq) + NH_4^+(aq) \quad (R2)$$

Line 305 – 309

"Previous studies reported that the volatilization of ammonium nitrate during sampling can cause negative mass artifacts, leading to the underestimation of both NO_3^- (8%–16%) (Chow et al., 2005) and NH_4^+ (10%–28%) (Yu et al., 2006). The volatilization is highly dependent on relative humidity and temperature. However, such losses are expected to be systematic over time and therefore are unlikely to significantly affect the general trends in this study, because our measurements were conducted in the same season."

Line 312 – 316

"The generally lower intercepts observed in the NO_3^-/NO_2 regression compared to those in the

SO_4^{2-}/SO_2 regression can be explained by the semi-volatile nature of nitrate (Yu et al., 2006). The formation of HNO_3, gaseous precursor of NO_3^-, is suppressed under very low NO_2 level. Therefore, the reaction in R2 tends to proceed to the left. This facilitates partitioning of NO_3^- into gas phase, leading to less accumulation of NO_3^- in particle phase."

11) The authors should specify which IMPROVE extinction equation they are using. There is an updated one from 2023 (see https://vista.cira.colostate.edu/Improve/wp-content/uploads/2023/10/IMPROVE_Data_User_Guide_24October2023.pdf, Section 8.1). Is that the equation used here? In addition, what is the local parameter scheme?

Response: Thanks for pointing this. It is necessary to illustrate which IMPROVE equation was used in this study. To estimate the light extinction coefficient (bext), the first Interagency Monitoring of Protected Visual Environments (IMPROVE) equation was developed by the U.S. National Park Service with support from the U.S. Environmental Protection Agency (EPA) (Malm et al., 1994; EPA, 2003), but this equation tended to underestimate/overestimate the highest/lowest bext values. Consequently, the revised IMPROVE equation was then proposed (Malm and Hand, 2007; Pitchford et al., 2007). Here, we used the revised one proposed in 2007:

$$b\text{ext} \approx 2.2 \times f_S(RH) \times [\textit{Small Ammonium Sulfate}] + 4.8 \times f_L(RH) \times [\textit{Large Ammonium Sulfate}] + 2.4 \times f_S(RH) \times [\textit{Small Ammonium Nitrate}] + 5.1 \times f_L(RH) \times [\textit{Large Ammonium Nitrate}] + 2.8 \times [\textit{Small Organic Mass}] + 6.1 \times [\textit{Large Organic Mass}] + 10 \times [\textit{Elemental Carbon}] + 1 \times [\textit{Fine Soil}] + 1.7 \times f_{SS}(RH) \times [\textit{Sea Salt}] + 0.6 \times [\textit{Coarse Mass}] + \textit{Rayleigh Scattering (Site Specific)} + 0.33 \times [NO_2\ (ppb)]$$

We have added related introduction in the revised manuscript.

Because the scattering/absorbing efficiency (MSE/MAE) in the revised IMPROVE equation is an approximation based on measurements from clean areas. In addition, the calculation of hygroscopic growth factor (f(RH)) in the revised equation depends on relative humidity (RH) and particle size distribution (or aerosols mass), but does not account for the chemical composition in aerosols, which has been shown to significantly affect f(RH) (Li et al., 2021). These simplifications could lead to large discrepancies in polluted regions. In this study, we adopted the MSE/MAE obtained in the PRD region and calculated f(RH) based on $PM_{2.5}$ chemical composition in the PRD. Then we used them to replace the corresponding parameters in the revised IMPROVE equation, which was called local parameter scheme.

Line 77 – 81

"To estimate the light extinction coefficient (b_{ext}), the first Interagency Monitoring of Protected Visual Environments (IMPROVE) equation was developed by the U.S. National Park Service with support from the U.S. Environmental Protection Agency (EPA) (Malm et al., 1994; EPA, 2003), but this equation tended to underestimate (overestimate) the highest (lowest) b_{ext} values. Consequently, the revised IMPROVE equation was then proposed (Malm and Hand, 2007; Pitchford et al., 2007)."

Line 428 – 429

"We adopted MSE/MAE suggested by Fu et al. (2016), as well as relationship between chemical composition and f(RH) suggested by Li et al. (2021), to reconstruct b_{ext} (herein called local parameter scheme) using equations (9-12)."

12) Does the IMPROVE equation overestimate, or does the local parameterization underestimate? It is not clear that this can be said with any certainty. It is probably better to just state the differences between the methods.

Response: We thank the reviewer for the valuable suggestion. Due to lack of real measurements of b_{ext} in the PRD during our study period, it does not make sense to say the revised IMPROVE equation tend to overestimate b_{ext} or the local parameter scheme underestimate it. We have changed the statement in the revised manuscript.

Line 445 – 446

"b_{ext} estimated by the revised IMPROVE equation (335.72 ± 219.64 Mm^{-1}) was significantly higher than that estimated by local parameter scheme (262.67 ± 143.82 Mm^{-1})."

13) Figure 3: What do the error bars represent?

Response: Thank you for comment. The error bars in Figure 3 represent the standard deviation of the total concentration, calculated as the sum of primary and secondary species. We have added this in revised manuscript.

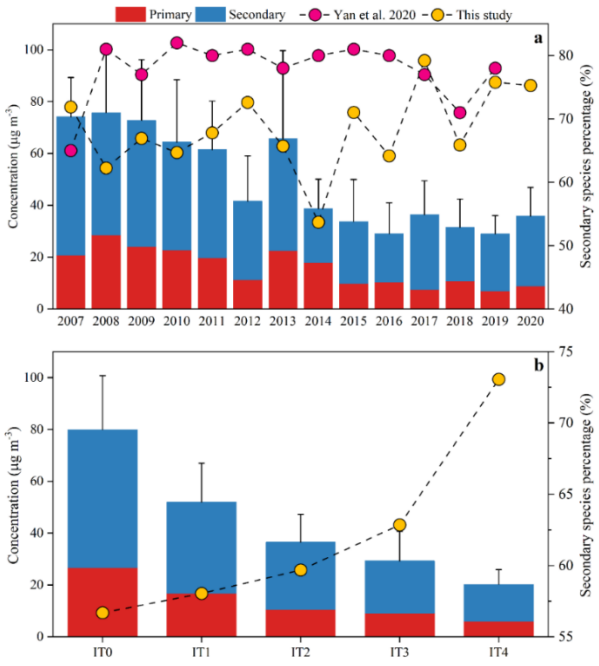

Figure 3. The variations in primary and secondary species during 2007–2020 (a) and their variations under different pollutants levels (b). Bars represent concentrations of them and circles represent the mass proportion of secondary species in $PM_{2.5}$. Secondary species (account for 54%–79%) dominated over primary species. The proportion of secondary species increased from 57% to 73% with improvement of air quality (From IT0 to IT4). The error bars represent the standard deviation of the total concentration, calculated as the sum of primary and secondary species.

14) Text S2 should be moved to the main document.

Response: Thanks for your suggestion. The calculation of aerosol pH and ALWC is a important part of our results. It indeed should be illustrated in the main document.

15) Figure S6 may benefit from an additional line showing the uncorrected changes in Cl-.

Response: Thanks for suggestion. We have added the line on the chart.

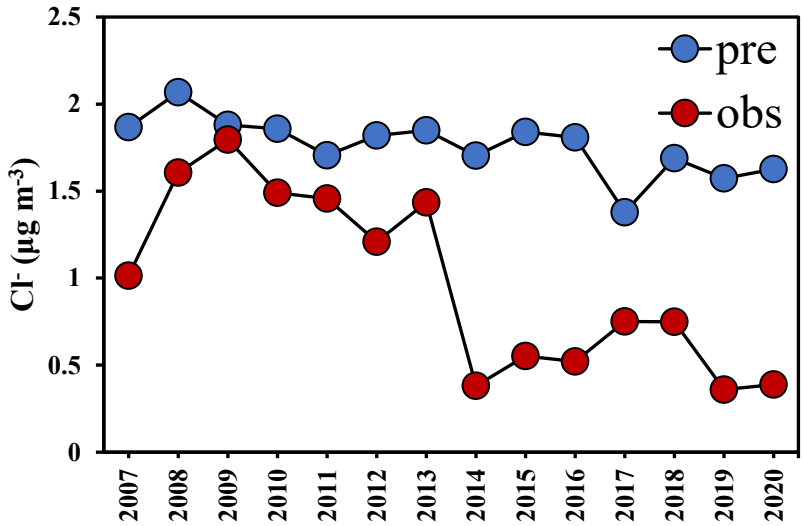

Figure S8. The observation (red) and prediction (blue) concentration of Cl^-. After eliminating variations in anthropogenic sources on Cl^-. It decreased slightly at a rate of –2% yr^{-1} during 2007-2020 (blue).

16) Figure S7: The colors of the lines do not match the colors in the legend.

Response: Thanks for reminding. We have corrected that.

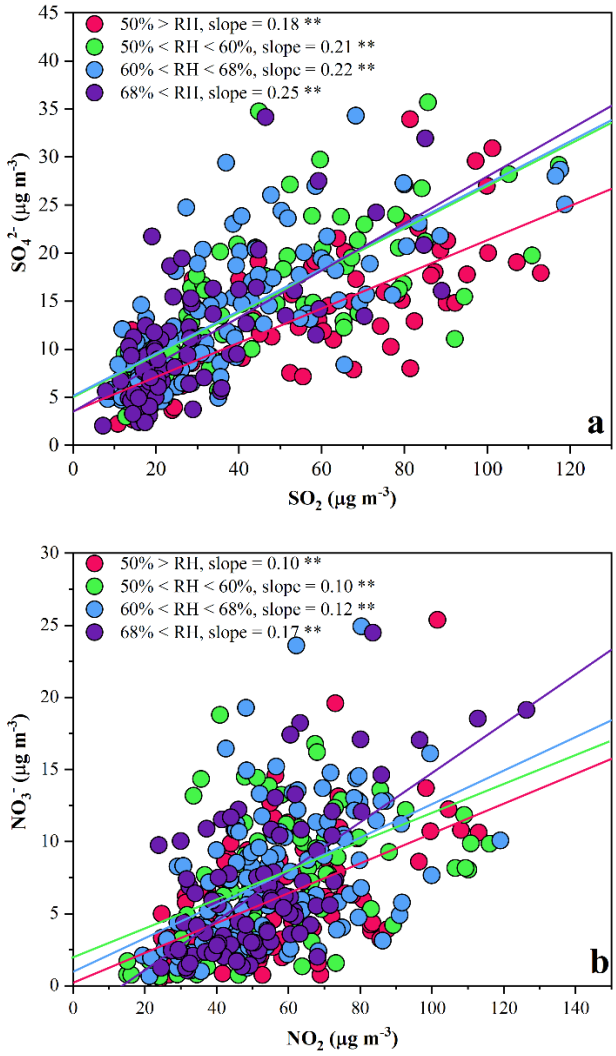

Figure S11. Correlations between SO_4^{2-}/SO_2 (a), as well as NO_3^-/NO_2 (b). Two asterisks denote *p* value less than 0.01. All samples were categorized into four groups according to the quartile ranges of RH. The slope became greater with rising RH, indicating conversion of primary pollutants to secondary species was more efficient.

17) Figure S8: Define SOR.

Response: Thanks for reminding. We have added definition of SOR and NOR in the revised supplement.

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