Author Response to Anonymous Reviewer #2

General Comment:

The manuscript worth a publication in ACP but some of its conclusive statements especially those related to emissions reductions needs more careful and convincing analysis.

We thank Reviewer #2 for the support to publish this paper and for the comments to improve it. Each comment by the reviewer is reproduced below, **in bold type**. Our replies to these comments are given below, and changes to the manuscript are marked in blue. Please refer to the responses to Specific Comments.

Specific comments

1. **Regulations reduce emissions, weather conditions do too. According to Figure 3, temperature drops significantly during the three periods. Emissions change due to change of temperature. For example, VOC evaporation decreases due to lower temperature, meanwhile emissions increase due to increasing heating needs (as authors found out).Also, when wind pattern changes, emissions change too. It clearly shows in Figure 3 that more northerly winds happened in “during” period than in “before” period, while with northerly winds air mass bring much less emissions to Beijing from upwind. To make some conclusive statements sounder, it would be better to compare changes of VOC concentrations and contributions under similar weather conditions.**

   Response: Accepted. Thank you very much for your valuable suggestion for improving our manuscript. We are very sorry for the unclear and misleading description about the conclusive statements related to emissions reduction. We respond to the comments in Specific Comments 1 of Reviewer #2 one by one, as follows:

(1) **Regulations reduce emissions, weather conditions do too. According to Figure 3, temperature drops significantly during the three periods. Emissions change due to change of temperature. For example, VOC evaporation decreases due to lower temperature, meanwhile emissions increase due to increasing heating needs (as authors found out).**

Reviewer#2 raises a good comment that temperature difference may lead to the change of emissions.

First, as reviewer points out VOC evaporation may decrease due to lower temperature. We have ever considered the influence from the change of temperature on VOC evaporation and condensation. The temperature difference during this campaign was less than 8 ℃. Based on Antoine equation and previous studies, we think the influence from temperature difference during this campaign on VOC evaporation and condensation were relatively small (Wolkoffa et al., 1998; Nevers, 2000; Goss et al., 2006; Okamoto et al., 2009). So we didn’t discuss the influence from temperature on VOC evaporation in this manuscript.

Second, VOC emissions may increase due to increasing heating needs. We found the proportions of some VOC species increased significantly after the early November; these species were tracers of fuel combustion (details can be found in section 3.2). Considered November is a transition month for central heating in northern China, we speculate that the combustion may have been an important nighttime source of VOCs during the second and the third periods.

To make conclusive statements sounder, P12467 L29 "The temperature difference during this campaign is relatively low, the influence from meteorological variability on VOC evaporation were not considered in this study" is added.

Reference

Goss, K. U.: Prediction of the temperature dependency of Henry's law constant using poly-parameter linear free energy relationships, Chemosphere, 64, 1369-1374, 10.1016/j.chemosphere.2005.12.049, 2006.

Paatero, P., and Tapper, U.: Postive matrix factorization a nonnegative factor model with optimal utilization of error estimates of data values, Environmetrics, 5, 111-126, 10.1002/env.3170050203, 1994.

Noel de Nevers, Air pollution control engineering, second edition, P330-335, McGraw-Hill, 2000.

Okamoto, K., Watanabe, N., Hagimoto, Y., Miwa, K., and Ohtani, H.: Changes in evaporation rate and vapor pressure of gasoline with progress of evaporation, Fire Safety Journal, 44, 756-763, 10.1016/j.firesaf.2009.03.004, 2009.

Wolkoff, P.: Impact of air velocity, temperature, humidity, and air on long-term VOC emissions from building products, Atmospheric Environment, 32, 2659-2668, 10.1016/s1352-2310(97)00402-0, 1998.

(2) When wind pattern changes, emissions change too. It clearly shows in Figure 3 that more northerly winds happened in "during" period than in "before" period, while with northerly winds air mass bring much less emissions to Beijing from upwind. To make some conclusive statements sounder, it would be better to compare changes of VOC concentrations and contributions under similar weather conditions.

This suggestion certainly has merit. As Reviewer#2 pointed out, wind pattern can influence the emissions. We have once considered the effect from wind pattern. But this paper aims to compare the relative variation of each VOC sources before, during, and after the air quality control period, which would not be influenced significantly by wind pattern. So we didn't discuss the influence from wind pattern in this manuscript.

Reviewer#2 raises a good point in that compare changes of VOC concentrations and contributions under similar weather conditions. We compared the peak VOC mixing ratios of pollution episodes occurred before, during, and after the control in the first paragraph of section 3.2. Under similar weather conditions the peak VOC mixing ratios during the control were significantly lower than those found before and after the control period.

It is misleading to use mixing ratios to describe reductions of VOC source contributions and make conclusions. Instead, we should make conclusions based on the relative source contributions to the reduction. To avoid the misleading, we revised the conclusion statement related to the reductions:

The last column of table 4 and table 5 is deleted.

P12454 L12-22, "Contributions of vehicular exhaust were most reduced (19.65 ppbv, the contributions before the control period minus the values after the control period), followed by industrial manufacturing (10.29 ppbv) and solvent utilization (6.20 ppbv). Contributions of evaporated or liquid gasoline and industrial chemical feedstock were slightly reduced, with values of 2.85 and 0.35 ppbv, respectively." is revised to "Compared with the values before control, contributions of vehicular exhaust were most reduced, followed by industrial manufacturing and solvent utilization. Reductions of these three sources were responsible for 50%, 26%, and 16% of the reductions in ambient VOCs. Contributions of evaporated or liquid gasoline and industrial chemical feedstock were slightly reduced."

P12467 L21-22, "…indicating that the control measures on traffic and industry were effective." is deleted.

P12467 L23 "Reductions of vehicular exhaust, industrial manufacturing, solvent utilization, evaporated or liquid gasoline, and industrial chemical feedstock were responsible for 50%, 26%, 16%, 7%, and 1% of the reductions in ambient VOCs, indicating that the control measures on traffic were most effective." is added.

P12467 L24-26, "In contrast, due to central heating and weak control on fuel combustion and LPG, contributions from these sources were elevated with the contributions of 5.65 and 1.34 ppbv, respectively.", is revised to "In contrast, due to central heating and weak control on fuel combustion and LPG, contributions from these sources were elevated by 80% and 22%, respectively."

2. **Was reduction of SOA formation a fact between the periods? Please provide measurement data to support it before stating and discussing the "reduction". SOA formation is complex; precursor emissions can change SOA formation, while temperature can change its formation too. Lower temperature somehow leads to more SOA. How come the unit of SOAP-weighted mass contribution is μg cm^{-3}?**

Response: Accepted. Great thanks to your carefulness and suggestion. We respond to the comments in Specific Comments 2 of Reviewer #2 one by one, as follows.

(1) Was reduction of SOA formation a fact between the periods? Please provide measurement data to support it before stating and discussing the "reduction". SOA formation is complex; precursor emissions can change SOA formation, while temperature can change its formation too.

Your suggestion is very valuable. We are sorry that due to the lack of available data, we can't provide measurement data to support the reduction of SOA formation. But it is reported that during control the concentration of $PM_{2.5}$ reduced 75% compared with the period before control, and the concentration of OC (organic carbon) reduced 54% (Beijing Municipal Environmental Protection Bureau, http://www.bjepb.gov.cn/). SOA constitute a significant fraction of $PM_{2.5}$ in China, and contribute 20-70% and 30%–90% of $PM_{2.5}$ and of organic aerosol concentrations, respectively (Huang et al., 2014). So we suspected that the concentration of SOA would likely be lower during control.

As Reviewer#2 point out, SOA formation is complex; precursor emissions can change SOA formation, while temperature can change its formation too. In this study, we focus on the precursor emissions. VOC is an important precursor of SOA, and $PM_{2.5}$ reduced a lot during control .We suspect that the large reduction of VOCs may contribute the $PM_{2.5}$ reduction. So we use SOAP approach to estimate the influence of variations of precursor emissions on SOA and discuss the effectiveness of the air quality controls. Temperature may influence the gas/particle conversion of gaseous hydrocarbon precursors (Lin et al., 2002). It will be essential to study the role of temperature in the formation of SOA in our future work.

To make it clear, in the first paragraph of section 3.4 "SOAP-weighted mass contributions of each VOC source were used to determine the most important precursor source likely to be responsible for the reduction of SOA formation." is deleted. "It is reported that during control the concentration of $PM_{2.5}$ reduced a lot (Beijing Municipal Environmental Protection Bureau, http://www.bjepb.gov.cn/). SOA constitute a significant fraction of $PM_{2.5}$ in China (Ding et al., 2012; Guo et al., 2012; Huang et al., 2014) and VOCs play an important role in the formation of SOA. The large reduction of VOCs may lead to the reduction of SOA and contribute to the $PM_{2.5}$ reduction. SOAP-weighted mass contributions of each VOC source were used to estimate the influence of variations of precursor emissions on SOA" is added.

P12468 L22-26, "We have to point out that the SOA potentials could not be quantitatively estimated by this approach. We focus on only the variations of the contribution of precursor sources to SOA formation and evaluating the major sources of the reduction of SOA during the air quality control period. Detailed VOC data in this work will provide useful information for further study on the SOA formation." is changed to "Current knowledge about formation mechanisms of SOA is still very limited (Guo et al., 2012). We have to point out that the SOAP is computed to understand the potential to form SOA for VOC species, which cannot estimate the SOA formation from VOCs actually in certain atmospheric conditions. In this study, we used SOAP approach to discuss the effectiveness of the air quality controls. Detailed VOC data in this work will provide useful information for further study on the formation mechanisms of SOA."

Reference:

Ding X, Wang X M, Gao B, et al. Tracer-based estimation of secondary organic carbon in the Pearl River Delta, south China (J). J. Geophys. Res.-Atmos., 2012,117, D05313,doi:10.1029/2011JD016596.

Guo, S., Hu, M., Guo, Q., Zhang, X., Zheng, M., Zheng, J., Chang, C. C., Schauer, J. J., and Zhang, R.: Primary Sources and Secondary Formation of Organic Aerosols in Beijing, China, Environ. Sci. Technol., 46, 9846-9853, 10.1021/es20425641, 2012.

Huang, R. J., Zhang, Y. L., Bozzetti, C., Ho, K. F., Cao, J. J., Han, Y. M., Daellenbach, K. R., Slowik, J. G., Platt, S. M., Canonaco, F., Zotter, P., Wolf, R., Pieber, S. M., Bruns, E. A., Crippa, M., Ciarelli, G., Piazzalunga, A., Schwikowski, M., Abbaszade, G., Schnelle-Kreis, J., Zimmermann, R., An, Z. S., Szidat, S., Baltensperger, U., El Haddad, I., and Prevot, A. S. H.: High secondary aerosol contribution to particulate pollution during haze events in China, Nature, 514, 218-222, 10.1038/nature13774, 2014.

Lin, J. J.: Characterization of the major chemical species in PM2.5 in the Kaohsiung City, Taiwan, Atmospheric Environment, 36, 1911-1920, 10.1016/s1352-2310(02)00193-0, 2002.

(2) How come the unit of SOAP-weighted mass contribution is μg cm^{-3}?

We are sorry for the unclear expression. In this study, the unit of measured VOC species is mixing ratio (ppbv). “The SOAP represents the propensity for an organic compound to form SOA when an additional mass emission of that compound is added to the ambient atmosphere expressed relative to that SOA formed when the same mass of toluene is added (Derwent et al., 2010).” We should convert mixing ratios for each VOC species to mass concentrations. Linking with the molar mass of VOC species and basing on ideal gas law, the paper converts the unit of VOC species from ppbv to ug m^{-3}. $SOAP_{(i)}$ is a number with no unit. Based on Eq. (3), the unit of SOAP-weighted mass contribution is μg cm^{-3}.

To make it more clearly, P12460 L14-18 is changed to: “The SOAP-weighted mass contribution of each VOC source (ug cm^{-3}) can be calculated using Eq. (3):

$$SOAP_{weighted\ mass\ contribution} = \sum(VOCs)_{(i)} \times SOAP_{(i)} \tag{3}$$

where $(VOCs)_{(i)}$ is the mass contribution of a VOC source to species *i* (ug cm^{-3}), estimated by PMF analysis (linking with the molar mass of VOC species and basing on ideal gas law, we converts the unit of VOC species from ppbv to ug m^{-3}); $SOAP_{(i)}$ is the SOA formation potential for species *i* (unitless, Table S3).”

3. P12456, L5 M. Wang -> Wang; P12464, L14, resident -> residential; P12468, L23, concentrated -> focus.

Response: Accepted. We appreciate your careful reading very much and sorry for the writing mistakes in the manuscript. These have been corrected accordingly.