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*Supplement of*

# Process-based and observation-constrained SOA simulations in China: the role of semivolatile and intermediate-volatility organic compounds and OH levels

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**Table S1.** Summary of the location, sampling information of the 86 surface online OA measurements in China as well as the campaign-average mass concentrations of the observed OA and the PMF-derived POA and SOA. The yearly measurements from 2011 to 2019 are 6, 8, 16, 19, 17, 8, 4, 7, and 1, respectively.

| Site # | Site Name / Region | References | Site Type | Sampling Period | Lon. | Lat. | OA (μg m$^{-3}$) | POA (μg m$^{-3}$) | SOA (μg m$^{-3}$) |
|---|---|---|---|---|---|---|---|---|---|
| 1 | IAP / NCP | J. K. Zhang et al. (2014) | urban | 1/1/2013-2/1/2013 | 116.37 | 39.97 | 44.65 | 20.54 | 24.11 |
| 2 | PKU / NCP | Hu et al. (2017) | urban | 1/23/2013-3/2/2013 | 116.31 | 39.99 | 29.7 | 14.9 | 14.8 |
| 3 | IAP / NCP | Y. Sun et al. (2016a) | urban | 12/17/2013-1/17/2014 | 116.37 | 39.97 | 38.1 | 21.5 | 16.5 |
| 4 | IAP / NCP | J. K. Zhang et al. (2015) | urban | 1/1/2014-2/3/2014 | 116.37 | 39.97 | 27.27 | 8.45 | 18.82 |
| 5 | IRSDE / NCP | Elser et al. (2016) | urban | 1/9/2014-1/26/2014 | 116.38 | 40.00 | 43 | 35.67 | 7.4 |
| 6 | IAP / NCP | Xu et al. (2019a) | urban | 11/17/2014-12/13/2014 | 116.37 | 39.97 | 30.4 | 16.1 | 14.7 |
| 7 | IAP / NCP | Zhou et al. (2018) | urban | 12/1/2014-1/18/2015 | 116.37 | 39.97 | 31.8 | 18.4 | 14.3 |
| 8 | THU / NCP | H. Li et al. (2019) | urban | 12/6/2014-2/27/2015 | 116.30 | 40.00 | 30.4 | 17.9 | 12.6 |
| 9 | IAP / NCP | J. K. Zhang et al. (2016a) | urban | 12/10/2014-12/31/2014 | 116.37 | 39.97 | 20.25 | 12.14 | 8.1 |
| 10 | NCNT / NCP | Huang et al. (2020) | urban | 12/29/2014-2/28/2015 | 116.38 | 40.00 | 38.38 | 26.86 | 11.9 |
| 11 | IAP / NCP | Y. Zhang et al. (2016) | urban | 2/2/2015-4/1/2015 | 116.37 | 39.97 | 39 | 24.75 | 14.25 |
| 12 | NCN / NCP | Duan et al. (2020) | urban | 12/4/2015-2/6/2016 | 116.32 | 39.99 | 30.97 | 18.8 | 12.2 |
| 13 | IAP / NCP | Xu et al. (2019a) | urban | 11/17/2016-12/13/2016 | 116.37 | 39.97 | 36.4 | 18.3 | 19 |
| 14 | IAP / NCP | Zhao et al. (2019) | urban | 12/17/2016-12/30/2016 | 116.37 | 39.97 | 53.7 | 22.6 | 30.9 |
| 15 | THU / NCP | H. Li et al. (2019) | urban | 12/11/2017-2/2/2018 | 116.30 | 40.00 | 11.9 | 6.1 | 5.8 |
| 16 | IAP / NCP | J. Li et al. (2020) | urban | 11/10/2018-1/31/2019 | 116.37 | 39.97 | 11.5 | 6.7 | 4.8 |
| 17 | IGDB / NCP | Huang et al. (2019) | urban | 1/11/2014-2/18/2014 | 114.54 | 38.03 | 89 | 64.97 | 24.03 |
| 18 | Handan / NCP | H. Li et al. (2017) | urban | 12/4/2015-2/5/2016 | 114.50 | 36.57 | 82.50 | 50.61 | 31.87 |
| 19 | PKU / NCP | Hu et al. (2017) | urban | 3/30/2012-5/7/2012 | 116.31 | 39.99 | 14 | 7.2 | 6.8 |
| 20 | PKU / NCP | Hu et al. (2016b) | urban | 8/3/2011-9/15/2011 | 116.31 | 39.99 | 26.4 | 8.9 | 17.1 |
| 21 | PKU / NCP | Hu et al. (2017) | urban | 7/29/2012-8/29/2012 | 116.31 | 39.99 | 12.5 | 3.9 | 8.6 |
| 22 | IAP / NCP | J. Zhang et al. (2015) | urban | 8/1/2012-8/31/2012 | 116.37 | 39.97 | 13 | 5.9 | 7.2 |
| 23 | IAP / NCP | Xu et al. (2017) | urban | 6/3/2014-7/11/2014 | 116.37 | 39.97 | 18.1 | 7.78 | 10.32 |
| 24 | THU / NCP | H. Li et al. (2018) | urban | 6/30/2015-7/27/2015 | 116.30 | 40.00 | 12.2 | 4.15 | 8.05 |
| 25 | NCN / NCP | Duan et al. (2020) | urban | 7/1/2015-8/19/2015 | 116.32 | 39.99 | 19.27 | 4.9 | 14 |
| 26 | NCN / NCP | Duan et al. (2019) | urban | 8/15/2015-9/10/2015 | 116.32 | 39.99 | 13.8 | 3.9 | 9.8 |
| 27 | IAP / NCP | Xu et al. (2019b) | urban | 6/4/2017-7/13/2017 | 116.37 | 39.97 | 9.8 | 3.4 | 6.4 |

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| 28 | IAP / NCP | Xu et al. (2019b) | urban | 5/20/2018-6/23/2018 | 116.37 | 39.97 | 12.7 | 3.7 | 9.2 |
| 29 | Xinxiang / NCP | H. Li et al. (2018) | urban | 6/8/2017-6/25/2017 | 113.90 | 35.30 | 18.00 | 4.32 | 13.5 |
| 30 | IAP / NCP | J. Zhang et al. (2015) | urban | 10/1/2012-10/31/2012 | 116.37 | 39.97 | 27 | 10.8 | 16.5 |
| 31 | PKU / NCP | Hu et al. (2017) | urban | 10/13/2012-11/13/2012 | 116.31 | 39.99 | 18.2 | 9.7 | 8.6 |
| 32 | IAP / NCP | J. K. Zhang et al. (2016a) | urban | 10/1/2014-10/27/2014 | 116.37 | 39.97 | 34.57 | 15.56 | 19.01 |
| 33 | IAP / NCP | W. Xu et al. (2015) | urban | 10/10/2014-11/2/2014 | 116.37 | 39.97 | 29.4 | 15 | 14.9 |
| 34 | IAP / NCP | J. K. Zhang et al. (2016b) | urban | 10/17/2014-11/2/2014 | 116.37 | 39.97 | 44.44 | 17.33 | 27.11 |
| 35 | IAP / NCP | Xu et al. (2017) | urban | 10/14/2014-11/12/2014 | 116.37 | 39.97 | 29.58 | 14.84 | 14.74 |
| 36 | IAP / NCP | Zhao et al. (2017) | urban | 9/4/2015-9/30/2015 | 116.37 | 39.97 | 18.31 | 6.33 | 11.97 |
| 37 | NCN / NCP | Duan et al. (2019) | urban | 9/11/2015-10/10/2015 | 116.32 | 39.99 | 21.2 | 8.4 | 13 |
| 38 | NCN / NCP | Duan et al. (2019) | urban | 10/11/2015-12/04/2015 | 116.32 | 39.99 | 29.6 | 15.7 | 13.9 |
| 39 | IAP / NCP | Zhou et al. (2018) | urban | 11/13/2014-11/30/2014 | 116.37 | 39.97 | 38.1 | 23.8 | 13.1 |
| 40 | Qingdao / NCP | Zhu et al. (2018) | urban | 11/1/2013-11/30/2013 | 120.47 | 36.10 | 10.43 | 5.57 | 4.86 |
| 41 | Xinglong / NCP | J. Li et al. (2020) | polluted regional | 11/10/2018-1/31/2019 | 117.67 | 40.40 | 5.4 | 2.2 | 3.2 |
| 42 | Gucheng / NCP | Kuang et al. (2020) | polluted regional | 11/11/2018-12/24/2018 | 115.73 | 39.15 | 66.08 | 42.29 | 21.81 |
| 43 | Changdao / NCP | Hu et al. (2013) | polluted regional | 3/21/2011-4/24/2011 | 120.70 | 37.99 | 13.4 | 4.4 | 9.4 |
| 44 | Xingtai / NCP | Y. Zhang et al. (2018) | suburban | 4/30/2016-6/20/2016 | 114.37 | 37.18 | 11.59 | 2.54 | 9.04 |
| 45 | Rizhao / NCP | Lei et al. (2020) | polluted regional | 3/2/2019-3/29/2019 | 119.40 | 35.18 | 15.66 | 2.19 | 13.31 |
| 46 | Xianghe / NCP | Y. Sun et al. (2016b) | polluted regional | 6/1/2013-6/30/2013 | 116.96 | 39.80 | 28.3 | 8.2 | 19 |
| 47 | BIPT / NCP | Chen et al. (2020) | suburban | 8/16/2018-9/16/2018 | 116.33 | 39.73 | 12.24 | 3.79 | 8.44 |
| 48 | Rizhao / NCP | Lei et al. (2020) | polluted regional | 9/2/2018-9/29/2018 | 119.40 | 35.18 | 8.32 | 0.67 | 7.65 |
| 49 | Nanjing / YRD | Tang et al. (2014) | urban | 1/4/2013-1/31/2013 | 118.73 | 32.21 | 26.26 | 14.11 | 11.89 |
| 50 | JEMC / YRD | Y. Zhang et al. (2015) | urban | 12/1/2013-12/31/2013 | 118.77 | 32.05 | 38.4 | 15.74 | 22.66 |
| 51 | NOC / YRD | J. Wang et al. (2016) | urban | 4/13/2015-4/29/2015 | 118.73 | 32.01 | 12.69 | 5.64 | 7.04 |
| 52 | Nanjing / YRD | Y. J. Zhang et al. (2015) | urban | 6/1/2013-6/15/2013 | 118.76 | 32.08 | 15.4 | 3.3 | 11.2 |
| 53 | JEMC / YRD | Y. J. Zhang et al. (2014) | urban | 8/1/2013-8/31/2013 | 118.77 | 32.05 | 10.3 | 2.73 | 6.86 |
| 54 | BJ / YRD | K. Li et al. (2018) | urban | 8/5/2016-8/21/2016 | 120.21 | 30.21 | 17 | 6.7 | 10.7 |
| 55 | Nanjing / YRD | Y. J. Zhang et al. (2015) | urban | 10/15/2013-10/30/2013 | 118.76 | 32.08 | 20.3 | 7.2 | 13.1 |
| 56 | JEMC / YRD | Y. Zhang et al. (2017) | urban | 10/20/2015-11/19/2015 | 118.75 | 32.04 | 25.2 | 7.06 | 18.14 |
| 57 | BJ / YRD | K. Li et al. (2018) | urban | 9/7/2016-9/23/2016 | 120.21 | 30.21 | 18.5 | 6.1 | 12.1 |

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| 58 | Lin'an / YRD | Y. W. Zhang et al. (2015) | polluted regional | 11/16/2013-12/18/2013 | 119.73 | 30.30 | 29 | 13.4 | 15.7 |
| 59 | Dongguan / PRD | Zhu et al. (2018) | urban | 12/12/2013-1/1/2014 | 113.75 | 23.03 | 23.3 | 7.36 | 15.75 |
| 60 | PKUshenzhen / PRD | Cao et al. (2018) | urban | 12/31/2015-1/23/2015 | 113.90 | 22.60 | 18.45 | 7.93 | 10.51 |
| 61 | GIG / PRD | Guo et al. (2020) | urban | 11/20/2017-1/5/2018 | 113.37 | 23.15 | 17.3 | 5.36 | 12.11 |
| 62 | Mong Kok / PRD | Lee et al. (2015) | urban | 3/7/2013-5/15/2013 | 114.17 | 22.32 | 12.8 | 7.9 | 4.9 |
| 63 | Mong Kok / PRD | Lee et al. (2015) | urban | 5/16/2013-7/19/2013 | 114.17 | 22.32 | 7.9 | 5.6 | 2.2 |
| 64 | HKEPD / PRD | C. Sun et al. (2016) | urban | 9/3/2013-12/31/2013 | 114.17 | 22.32 | 15.1 | 6.3 | 8.8 |
| 65 | GPACS / PRD | Qin et al. (2017) | suburban | 11/07/2014-1/3/2015 | 113.35 | 23.00 | 20.36 | 7.81 | 12.55 |
| 66 | HKUST / PRD | Y. J. Li et al. (2015) | clean regional | 1/19/2012-3/1/2012 | 114.26 | 22.34 | 5.1 | 0.98 | 4.12 |
| 67 | HKUST / PRD | Y. J. Li et al. (2015) | clean regional | 4/25/2011-6/1/2011 | 114.26 | 22.34 | 4 | 0.8 | 3.2 |
| 68 | HKUST / PRD | Y. J. Li et al. (2015) | clean regional | 9/1/2011-9/29/2011 | 114.26 | 22.34 | 4.1 | 0.75 | 3.35 |
| 69 | HKUST / PRD | Y. J. Li et al. (2015) | clean regional | 10/28/2011-12/15/2011 | 114.26 | 22.34 | 6 | 0.82 | 5.18 |
| 70 | Xi’an / NW | Elser et al. (2016) | urban | 12/13/2013-1/6/2014 | 108.88 | 34.23 | 128.5 | 100.32 | 21 |
| 71 | LZU / NW | Xu et al. (2016) | urban | 1/10/2014-2/4/2014 | 103.85 | 36.05 | 29.33 | 18.41 | 10.91 |
| 72 | Baoji / NW | Y. C. Wang et al. (2017) | urban | 2/26/2013-3/27/2014 | 107.14 | 34.35 | 29.7 | 16.63 | 13.07 |
| 73 | CAREERI / NW | Xu et al. (2014) | urban | 7/11/2012-8/7/2012 | 103.86 | 36.05 | 11.51 | 4.6 | 6.79 |
| 74 | GMA / NW | X. Zhang et al. (2017) | urban | 10/27/2014-12/3/2014 | 103.88 | 36.04 | 18.2 | 10.87 | 7.33 |
| 75 | Sichuan Basin / OTR | Hu et al. (2016a) | suburban | 12/3/2012-1/5/2013 | 104.64 | 30.15 | 21.5 | 6.2 | 15.4 |
| 76 | Nan'ao island / OTR | Cao et al. (2019) | polluted regional | 12/22/2015-1/16/2016 | 117.02 | 23.42 | 7.06 | 2.61 | 4.45 |
| 77 | Xiamen / OTR | Cao et al. (2017) | suburban | 5/1/2015-5/18/2015 | 118.05 | 24.60 | 13.07 | 3.61 | 9.46 |
| 78 | Xinzhou / OTR | Q. Wang et al. (2016) | suburban | 7/17/2014-9/5/2014 | 112.12 | 38.07 | 11.72 | 2.2 | 9.5 |
| 79 | Mt. Yulong / OTR | Zheng et al. (2017) | remote | 3/22/2015-4/14/2015 | 100.20 | 27.20 | 3.88 | 0.5 | 3.37 |
| 80 | Lake Hongze / OTR | Zhu et al. (2016) | remote | 3/19/2011-4/24/2011 | 118.33 | 33.23 | 9.8 | 3 | 6.8 |
| 81 | Mt. Wuzhi / OTR | Zhu et al. (2016) | remote | 3/18/2015-4/15/2015 | 109.49 | 18.84 | 4.9 | 0.01 | 4.89 |
| 82 | QOMS / OTR | X. Zhang et al. (2018) | remote | 4/12/2016-5/12/2016 | 86.95 | 28.36 | 2.39 | 1.04 | 1.34 |
| 83 | WLG / OTR | Zhang et al. (2019) | remote | 7/1/2017-7/31/2017 | 100.90 | 36.28 | 3.14 | 0.79 | 2.35 |
| 84 | Nam Co / OTR | J. Wang et al. (2017a) | remote | 5/30/2015-6/30/2015 | 90.98 | 30.77 | 0.71 | 0.13 | 0.59 |
| 85 | Nam Co / OTR | Xu et al. (2018) | remote | 5/31/2015-7/1/2015 | 90.95 | 30.77 | 1.36 | 0 | 1.36 |
| 86 | NBS / OTR | Du et al. (2015) | remote | 9/5/2013-10/15/2013 | 101.26 | 37.61 | 4.9 | 0.8 | 4.1 |

**Table S2.** Summary of the location, sampling information of the 49 surface online measurements of VOCs as well as the campaign-average mixing ratios of benzene, toluene, and xylene in China.

| Site # | Site Name | References | Sampling Period | Lon. | Lat. | Benzene (ppbv) | Toluene (ppbv) | Xylene (ppbv) |
|---|---|---|---|---|---|---|---|---|
| 1 | PKU | Wang et al. (2014) | 12/29/2011-1/18/2012 | 116.31 | 40.00 | 2.34 | 2.67 | |
| 2 | PKU | J. Li et al. (2019a) | 1/1/2015-1/31/2015 | 116.33 | 39.99 | 1.30 | 1.20 | |
| 3 | NCNST | K. Li et al. (2019) | 1/31-2/15/2015 | 116.32 | 39.99 | 1.93 | 1.51 | |
| 4 | RCEES | Liu et al. (2017) | 12/15/2015-1/14/2016 | 116.30 | 40.00 | 1.81 | 1.67 | 1.12 |
| 5 | PKU | Y. Shi et al. (2020) | 12/01/2016-1/31/2017 | 116.33 | 39.99 | 3.27 | 3.63 | 1.50 |
| 6 | PKU | Y. Shi et al. (2020) | 12/2017 | 116.33 | 39.99 | 1.06 | 1.47 | 0.61 |
| 7 | PKU | Y. Shi et al. (2020) | 1/2018 | 116.33 | 39.99 | 1.03 | 1.26 | 0.51 |
| 8 | TEPB | Liu et al. (2016) | 12/2014-2/2015 | 117.15 | 39.10 | 0.54 | 0.27 | 0.38 |
| 9 | UCAS | K. Li et al. (2019) | 11/24-12/24/2014 | 116.67 | 40.41 | 0.91 | 0.73 | |
| 10 | Wangdu | Zhang et al. (2020) | 11/1/2017-1/21/2018 | 115.25 | 38.67 | 3.16 | 2.48 | 1.07 |
| 11 | NUIST | An et al. (2017) | 12/1/2013-2/28/2014 | 118.72 | 32.21 | 3.21 | 3.20 | 0.74 |
| 12 | Ziyang | Li et al. (2014) | 12/6/2012-1/4/2013 | 104.64 | 30.15 | 1.80 | 0.80 | |
| 13 | BJ | L. Li et al. (2015) | 5/2014 | 116.33 | 39.99 | 0.82 | 1.33 | |
| 14 | PKU | J. Li et al. (2019a) | 4/1/2015-4/30/2015 | 116.33 | 39.99 | 0.76 | 1.06 | |
| 15 | TEPB | Liu et al. (2016) | 3/2015-5/2015 | 117.15 | 39.10 | 0.24 | 0.30 | 0.21 |
| 16 | SAES | Y. Liu et al. (2019c) | 5/20-5/30/2017 | 121.72 | 31.28 | 0.42 | 1.31 | 1.52 |
| 17 | WHU | Hui et al. (2020) | 4/26-6/6/2017 | 114.36 | 30.54 | 0.49 | 0.71 | 0.43 |
| 18 | PKUSZ | Yu et al. (2019) | 4/1/2016-4/30/2016 | 113.90 | 22.60 | 0.60 | 3.12 | 1.07 |
| 19 | PKU | M. Wang et al. (2015) | 8/3/2011-9/13/2011 | 116.31 | 40.00 | 1.29 | 2.08 | 1.74 |
| 20 | PKU | M. Wang et al. (2015) | 8/1/2012-8/31/2012 | 116.31 | 40.00 | 0.98 | 2.25 | 1.75 |
| 21 | PKU | M. Wang et al. (2015) | 8/7/2013-8/25/2013 | 116.31 | 40.00 | 0.98 | 2.00 | 1.50 |
| 22 | PKU | J. Li et al. (2019a) | 7/1/2015-7/31/2015 | 116.33 | 39.99 | 0.80 | 1.55 | |
| 23 | PKU | Li et al. (2016) | 8/11/2015-9/3/2015 | 116.33 | 39.99 | 0.58 | 1.20 | 0.68 |
| 24 | TEPB | Liu et al. (2016) | 6/2015-8/2015 | 117.15 | 39.10 | 8.26 | 1.54 | 0.89 |
| 25 | Gucheng | L. Li et al. (2015) | 6/5/2013-7/16/2013 | 115.67 | 39.13 | 1.08 | 1.31 | |
| 26 | Quzhou | L. Li et al. (2015) | 6/11/2014-7/9/2014 | 115.02 | 36.86 | 0.81 | 0.48 | |
| 27 | NUIST | An et al. (2017) | 6/1-8/31/2013 | 118.72 | 32.21 | 1.86 | 1.47 | 0.74 |
| 28 | Zhaohui | K. Li et al. (2017) | 7/1/2013-8/15/2013 | 120.16 | 30.29 | 0.39 | 1.28 | 1.51 |
| 29 | Nanling | Gong et al. (2018) | 7/15/2016-8/17/2016 | 112.90 | 24.69 | 0.05 | 0.15 | 0.04 |
| 30 | PKU | Wu et al. (2016) | 10/1-10/16/2014 | 116.33 | 39.99 | 1.21 | 1.78 | 1.65 |
| 31 | PKU | J. Li et al. (2015) | 10/18-11/22/2014 | 116.33 | 39.99 | 1.62 | 2.43 | 1.24 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 32 | PKU | J. Li et al. (2019a) | 10/1/2015-10/31/2015 | 116.33 | 39.99 | 1.01 | 1.66 | |
| 33 | TEPB | Liu et al. (2016) | 11/2014; 9/2015-10/2015 | 117.15 | 39.10 | 2.12 | 1.18 | 1.06 |
| 34 | Shenyang | Z. Ma et al. (2019) | 8/20-9/16/2017 | 123.42 | 41.78 | 1.41 | 1.63 | 0.88 |
| 35 | Pixian | Deng et al. (2019) | 8/28/2016-10/7/2016 | 103.87 | 30.80 | 0.79 | 1.78 | 1.17 |
| 36 | CJZ | J. Li et al. (2018) | 8/24/2015-9/22/2015 | 106.50 | 29.62 | 1.10 | 1.20 | 0.70 |
| 37 | JYS | J. Li et al. (2018) | 8/24/2015-9/22/2015 | 106.38 | 29.83 | 0.60 | 0.40 | |
| 38 | NQ | J. Li et al. (2018) | 8/24/2015-9/22/2015 | 106.59 | 29.43 | 1.00 | 0.80 | 0.90 |
| 39 | Heshan | M. Song et al. (2019) | 10/22-11/22/2014 | 112.93 | 22.73 | 1.04 | 2.96 | 2.39 |
| 40 | RCEES | Q. Li et al. (2020) | 3/1/2016-1/31/2017 | 116.50 | 39.80 | 1.00 | 1.20 | 1.00 |
| 41 | BNU | Liu et al. (2020) | 1,4,7,10/2016 | 116.37 | 39.97 | 0.92 | 0.98 | 0.80 |
| 42 | Langfang | C. Song et al. (2019) | 4/2016-3/2017 | 116.76 | 39.57 | 0.86 | 0.84 | 0.62 |
| 43 | NUIST | An et al. (2014) | 3/2011-2/2012 | 118.72 | 32.21 | 3.14 | 2.10 | 1.37 |
| 44 | WH | Lyu et al. (2016) | 2/2013-10/2014 | 114.37 | 30.54 | 1.70 | 2.00 | 0.60 |
| 45 | WHU | Yang et al. (2019) | 9/2016-8/2017 | 114.62 | 30.88 | 0.73 | 0.95 | 0.37 |
| 46 | Qingyang | Song et al. (2018) | 10/27/2016-9/30/2017 | 104.08 | 30.93 | 0.95 | 1.80 | 1.71 |
| 47 | LEMS | Jia et al. (2016) | 1/2013-12/2013 | 103.92 | 36.05 | 1.94 | 1.01 | 1.23 |
| 48 | GPACS | Zou et al. (2015) | 6/2011-5/2012 | 113.35 | 23.00 | 0.62 | 4.59 | 2.07 |
| 49 | Mong Kok | Z. Li et al. (2020) | 1/2013-12/2014 | 114.17 | 22.32 | 0.44 | 1.74 | 0.63 |

**Table S3.** Summary of the locations, sampling information, and campaign-average mixing ratios of the 28 surface HONO measurements as well as the corresponding simulation results in the Sp_base and Sp_R1+2 simulations.

| Site # | Site Name | References | Sampling Period | Lon. | Lat. | Observed HONO (ppbv) | Sp_base simulation | | Sp_R1+2 simulation | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | HONO (ppbv) | Sim/ Obs | HONO (ppbv) | Sim/ Obs |
| Winter | | | | | | | | | | |
| 1 | EESCAS | J. Wang et al. (2017b) | 1/3-1/27/2016 | 116.34 | 40.01 | 1.03 | 0.71 | 0.68 | 1.04 | 1.01 |
| 2 | Jinan | D. Li et al. (2018) | 12/1/2015-2/29/2016 | 117.05 | 36.67 | 1.14 | 0.95 | 0.84 | 1.64 | 1.44 |
| 3 | Jinan | L. Wang et al. (2015) | 11/26/2013-1/5/2014 | 117.05 | 36.67 | 0.36 | 1.12 | 3.15 | 1.93 | 5.42 |
| 4 | YelRD | Gu et al. (2020) | 2/8-3/24/2017 | 118.98 | 37.76 | 0.26 | 0.41 | 1.59 | 0.60 | 2.34 |
| 5 | NUIST | Zheng et al. (2020) | 12/1-12/31/2015 | 118.71 | 32.21 | 1.28 | 0.85 | 0.66 | 1.62 | 1.27 |
| 6 | SORPES | Y. Liu et al. (2019b) | 12/2017-2/2018 | 118.95 | 32.12 | 0.67 | 0.80 | 1.19 | 1.43 | 2.12 |
| 7 | Tung Chung | Z. Xu et al. (2015) | 2/3-3/9/2012 | 113.93 | 22.30 | 0.90 | 0.06 | 0.07 | 0.21 | 0.23 |
| 8 | Zhengzhou | Hao et al. (2020) | 1/9-1/31/2019 | 113.52 | 34.80 | 2.51 | 1.56 | 0.62 | 2.20 | 0.88 |
| Spring | | | | | | | | | | |
| 9 | EESCAS | J. Wang et al. (2017b) | 4/1-5/14/2016 | 116.34 | 40.01 | 1.05 | 0.15 | 0.14 | 0.40 | 0.38 |
| 10 | Jinan | D. Li et al. (2018) | 3/1-5/31/2016 | 117.05 | 36.67 | 1.11 | 0.45 | 0.41 | 1.05 | 0.95 |
| 11 | Changzhou | X. Shi et al. (2020) | 4/3-4/24/2017 | 119.90 | 31.80 | 1.52 | 0.32 | 0.21 | 0.78 | 0.51 |
| 12 | SORPES | Y. Liu et al. (2019b) | 3-5/2018 | 118.95 | 32.12 | 0.45 | 0.43 | 0.94 | 1.10 | 2.42 |
| 13 | SORPES | Nie et al. (2015) | 4/17-6/24/2012 | 118.95 | 32.12 | 0.75 | 0.30 | 0.40 | 0.90 | 1.19 |
| 14 | Tung Chung | Z. Xu et al. (2015) | 5/1-5/31/2012 | 113.93 | 22.30 | 0.40 | 0.15 | 0.38 | 0.56 | 1.40 |
| Summer | | | | | | | | | | |
| 15 | Wangdu | Y. Liu et al. (2019a) | 6/8-7/5/2014 | 115.18 | 38.68 | 0.92 | 0.22 | 0.24 | 0.66 | 0.72 |
| 16 | EESCAS | J. Wang et al. (2017b) | 6/20-7/25/2016 | 116.34 | 40.01 | 1.39 | 0.15 | 0.11 | 0.50 | 0.36 |
| 17 | Jinan | D. Li et al. (2018) | 6/1-8/31/2016 | 117.05 | 36.67 | 0.76 | 0.42 | 0.55 | 1.09 | 1.42 |
| 18 | YelRD | Gu et al. (2020) | 6/1-7/10/2017 | 118.98 | 37.76 | 0.17 | 0.06 | 0.36 | 0.23 | 1.36 |
| 19 | SORPES | Y. Liu et al. (2019b) | 11/2017; 9-11/2018 | 118.95 | 32.12 | 1.04 | 0.45 | 0.44 | 1.04 | 1.00 |
| 20 | SORPES | Y. Liu et al. (2019b) | 6-8/2018 | 118.95 | 32.12 | 0.66 | 0.31 | 0.46 | 0.86 | 1.30 |
| 21 | Tung Chung | Z. Xu et al. (2015) | 8/3-9/7/2011 | 113.93 | 22.30 | 0.68 | 0.10 | 0.14 | 0.43 | 0.64 |
| 22 | IEECAS | Huang et al. (2017) | 7/24-8/6/2015 | 108.87 | 34.22 | 1.11 | 0.06 | 0.06 | 0.31 | 0.28 |
| Autumn | | | | | | | | | | |
| 23 | EESCAS | J. Wang et al. (2017b) | 9/22-10/21/2016 | 116.34 | 40.01 | 2.27 | 0.31 | 0.13 | 0.62 | 0.27 |
| 24 | ICCAS | Tong et al. (2015) | 10/28-11/2/2014 | 116.32 | 39.99 | 1.45 | 0.37 | 0.26 | 0.64 | 0.44 |
| 25 | UCAS | Tong et al. (2015) | 10/28-11/2/2014 | 116.60 | 40.40 | 0.75 | 0.17 | 0.23 | 0.27 | 0.36 |
| 26 | ICCAS | Jia et al. (2020) | 8/23-9/15/2018 | 116.32 | 39.99 | 1.24 | 0.21 | 0.17 | 0.66 | 0.53 |
| 27 | Jinan | D. Li et al. (2018) | 9/1-11/30/2015 | 117.05 | 36.67 | 1.75 | 0.80 | 0.46 | 1.51 | 0.86 |
| 28 | Tung Chung | Z. Xu et al. (2015) | 11/1-12/3/2011 | 113.93 | 22.30 | 0.92 | 0.08 | 0.09 | 0.29 | 0.32 |

**Table S4.** Summary of the locations, sampling information, and the maxima in the diurnal profiles of the hourly mean concentrations from 10 surface OH and $HO_2$ measurements in China as well as the corresponding model results in the Sp_base and Sp_R1+2 simulations.

| Site # | Site Name/ Region | References | Sampling Period | Lon. | Lat. | OH ($10^6$ $cm^{-3}$) | | | $HO_2$ ($10^8$ $cm^{-3}$) | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | Obs | Sp_base | Sp_R1+2 | Obs | Sp_base | Sp_R1+2 |
| Winter | | | | | | | | | | | |
| 1 | Huairou/ NCP | Tan et al. (2018) | 1/6-2/2/2016; 2/20-3/5/2016 | 116.68 | 40.41 | 3.1 | 1.1 | 2.8 | 0.8 | 0.2 | 0.5 |
| 2 | IAP/ NCP | Slater et al. (2020) | 11/16-11/20/2016; 12/2-12/8/2016 | 116.37 | 39.97 | 2.7 | 1.3 | 3.0 | 0.4 | 0.2 | 0.6 |
| 3 | IAP/ NCP | Ma (2019) | 11/15-12/9/2016 | 116.37 | 39.97 | 2.3 | 1.1 | 2.8 | 0.6 | 0.2 | 0.5 |
| 4 | PKU/ NCP | X. Ma et al. (2019) | 11/16-12/24/2017 | 116.31 | 40.00 | 1.8 | 1.2 | 2.7 | 0.4 | 0.2 | 0.5 |
| Summer | | | | | | | | | | | |
| 5 | Wangdu/ NCP | Tan et al. (2017) | 6/7-7/8/2014 | 115.2 | 39.5 | 8.3 | 6.3 | 12.9 | 8.3 | 2.6 | 5.2 |
| 6 | IAP/ NCP | Whalley et al. (2021) | 5/21-6/26/2017 | 116.37 | 39.97 | 8.6 | 3.5 | 7.6 | 2.9 | 3.5 | 5.9 |
| 7 | Tiazhou/ YRD | Yang (2021) | 5/23-6/17/2018 | 119.9 | 32.3 | 11.1 | 9.0 | 14.2 | 11.4 | 4.4 | 6.6 |
| 8 | Chengdu/ Southwest | Yang et al. (2021) | 8/10-8/25/2019 | 103.8 | 30.4 | 10.0 | 9.8 | 19.7 | 10.1 | 4.0 | 7.6 |
| Autumn | | | | | | | | | | | |
| 9 | Heshen/ PRD | Tan et al. (2019) | 10/19-11/22/2014 | 112.9 | 22.7 | 4.4 | 7.0 | 18.4 | 3.2 | 1.8 | 5.6 |
| 10 | Shenzhen/ PRD | Yang (2021) | 10/5-10/28/2018 | 113.9 | 22.6 | 4.5 | 7.6 | 11.8 | 4.4 | 5.4 | 7.8 |

**Table S5.** Descriptions of the implemented new sources of HONO.

| New sources | Description | Reference |
|---|---|---|
| Traffic emission | $E_{\mathrm{HONO,traffic}} = 0.017 \times E_{\mathrm{NO}_x,\mathrm{traffic}}$ | Rappengluck et al. (2013) |
| Soil emission | $E_{\mathrm{HONO,soil}} = \mathrm{HONO}_f \times E_{\mathrm{NO_x,soil}} \times f_{\mathrm{SWC}} \times \mathrm{CRF(LAI, meteology, biome)}$<br>$E_{\mathrm{NO}_x,\mathrm{soil}}$ and canopy reduction factor (CRF): Hudman et al. (2012)<br>The soil water content adjustment factor ($f_{\mathrm{SWC}}$) and the proportions of the emissions of HONO to $NO_x$ ($\mathrm{HONO}_f$): Rasool et al. (2019) | Hudman et al. (2012)<br>Rasool et al. (2019) |
| Biomass burning emission | Burned area data: GFED4<br>$EF_{\mathrm{HONO,bb}}$: Andreae (2019) | Giglio et al. (2013)<br>Andreae (2019) |
| Heterogeneous reaction of $NO_2$ on the ground | $\mathrm{NO_2} \xrightarrow{surface} \mathrm{HONO} \quad (k_g)$<br>$k_{\mathrm{g}} = \frac{1}{8} \times v_{\mathrm{NO_2}} \times \frac{S_{\mathrm{g}}}{V} \times \gamma_{\mathrm{g-NO_2}}$<br>$\gamma_{\mathrm{g-NO_2}} = \begin{cases} 1 \times 10^{-6} & \text{(night)} \\ 2 \times 10^{-5} \times \frac{J(\mathrm{NO_2})}{2\times10^{-3}} & \text{(day)} \end{cases}$<br>$\frac{S_{\mathrm{g}}}{V} = \begin{cases} 0.1 & \text{(urban area)} \\ \frac{2\times\mathrm{LAI}}{\mathrm{H}} & \text{(other areas)} \end{cases}$<br>Land cover type: MCD12C1 v6 | Li et al. (2010);<br>Zheng et al. (2020) |
| Photolysis of nitrate | $\mathrm{Nitrate} \xrightarrow{hv} 0.67\mathrm{HONO} + 0.33\mathrm{NO_2} \quad (J_{Nitrate})$<br>$J_{Nitrate} = 100 \times J_{HNO_3}$ | Kasibhatla et al. (2018) |

**Table S6.** The ratios of IVOCs to NMVOCs emissions from different subsectors.

| Subsectors | IVOCs/NMVOCs | Reference |
|---|---|---|
| Gasoline | 0.032 | Lu et al. (2018) |
| Diesel | 0.527 | Lu et al. (2018) |
| Coal | 0.352 | Cai et al. (2019) |
| Biomass burning | 0.164 | Lim et al. (2019) |
| Solvent use | 0.240 | Khare and Gentner (2018) |

**Table S7.** Annual mean emissions of IVOC6, IVOC5, and IVOC4 in China in 2014 (unit: Tg year$^{-1}$). The values in parentheses are the IVOCs emissions when the residential emission is multiplied by a factor of 7.

| Sectors | Corresponding subsectors | IVOC6 | IVOC5 | IVOC4 | Total IVOCs |
|---|---|---|---|---|---|
| Power | Coal | 0.01 | 0.01 | 0.01 | 0.03 |
| Industry | Coal<br>Solvent | 2.51 | 1.43 | 0.83 | 4.77 |
| Residential | Coal<br>Biofuel | 0.60 (3.10) | 0.44 (1.72) | 0.17 (0.83) | 1.21 (5.64) |
| Transportation | Gasoline<br>Diesel | 0.23 | 0.18 | 0.15 | 0.56 |
| Total | - | 3.35 (5.85) | 2.05 (3.33) | 1.15 (1.81) | 6.56 (11.00) |

**Table S8.** The SOA yield parameterization for IVOCs under low and high $NO_x$ conditions in this study compared with the literature parameterization. The saturation concentration ($C^*$) is in unit of μg $m^{-3}$. Only one-step oxidation is considered in the parameterizations.

| Parameterization | Category | mass-based stoichiometric coefficients ($\alpha$) for $C^*$ | | | | | | SOA yield at 10 μg $m^{-3}$ | Reference |
|---|---|---|---|---|---|---|---|---|---|
| | | nonvolatile | 0.1 | 1 | 10 | 100 | 1000 | | |
| low $NO_x$ condition | | | | | | | | | |
| Pye and Seinfeld (2010) (Base) | IVOCs | 0.73 | - | - | - | - | - | 0.73 | Chan et al. (2009) |
| high $NO_x$ condition | | | | | | | | | |
| Pye and Seinfeld (2010) (Base) | IVOCs | - | 0 | 0.039 | 0.296 | 0.235 | - | 0.21 | Chan et al. (2009) |
| Koo et al. (2014) | IVOCs | - | - | 0.030 | 0.194 | 0.264 | 0.376 | 0.15 | Murphy and Pandis (2009) |
| Jathar et al. (2014) | *n*-$C_{13}$ proxy | - | 0.014 | 0.059 | 0.220 | 0.400 | - | 0.21 | Presto et al. (2010) |
| | *n*-$C_{15}$ proxy | - | 0.044 | 0.071 | 0.410 | 0.300 | - | 0.34 | |
| Lu et al. (2020) | IVOC6-ALK | - | 0.009 | 0.045 | 0.118 | 0.470 | - | 0.15 | Presto et al. (2010); Zhao et al. (2015); Zhao et al. (2016) |
| | IVOC5-ALK | - | 0.051 | 0.061 | 0.394 | 0.494 | - | 0.35 | |
| | IVOC4-ALK | - | 0.068 | 0.083 | 0.523 | 0.239 | - | 0.43 | |
| | IVOC3-ALK | - | 0.067 | 0.086 | 0.544 | 0.198 | - | 0.43 | |
| | IVOC6-ARO | - | 0.022 | 0.109 | 0.251 | 0.005 | - | 0.25 | |
| | IVOC5-ARO | - | 0.143 | 0.021 | 0.329 | 0.358 | - | 0.36 | |
| This study | IVOC6 | - | 0.011 | 0.052 | 0.201 | 0.296 | - | 0.19 | Presto et al. (2010) |
| | IVOC5 | - | 0.049 | 0.078 | 0.439 | 0.271 | - | 0.36 | |
| | IVOC4 | - | 0.063 | 0.089 | 0.550 | 0.200 | - | 0.44 | |

**Table S9.** The observed campaign-average mass concentrations of primary IVOCs at the SAES site and the corresponding model results in the Cp_R1+2 and Cp_R1+2+3 simulations.

| Site | Reference | Period | Observation ($\mu g\ m^{-3}$) | Cp_R1+2 simulation | | Cp_R1+2+3 simulation | |
|---|---|---|---|---|---|---|---|
| | | | | Sim ($\mu g\ m^{-3}$) | Sim/ Obs | Sim ($\mu g\ m^{-3}$) | Sim/ Obs |
| SAES (Shanghai) | Y. Li et al. (2019) | December 5, 2016 to January 3, 2017 | 35.1 | 9.3 | 0.26 | 15.6 | 0.44 |
| | | July 16 to August 8, 2017 | 4 | 1.9 | 0.48 | - | - |

**Table S10.** The values of $EF_{SOAP}/EF_{CO}$ used in models and the maximum of ΔOA/ΔCO obtained in the experiments based on oxidation flow reactor (OFR). The values in parentheses are the ΔOA/ΔCO uncorrected for losses of low volatility vapors in the experiments.

| Site/campaign name | Site/campaign condition | Simulating/Sampling period | $EF_{SOAP}/EF_{CO}$ (g $kg^{-1}$) or Maximum ΔOA/ΔCO (μg $m^{-3}$ $ppmv^{-1}$) | Reference |
|---|---|---|---|---|
| | | Model | | |
| | China | 2014 | 69 in Sp_base, R1, R1+2<br>80 in Sp_R1+2+3 | This study |
| | World | 2013 | 69 | Pai et al. (2020) |
| MILAGRO | Mexico City region | 15 to 31 March 2006 | 80 | Hodzic and Jimenez (2011) |
| CalNex | Urban outflow from Los Angeles | 15 May to 15 June 2010 | 69 | Hayes et al. (2015); Woody et al. (2016) |
| $SEAC^4RS$ | Summertime aircraft in the southeast US | 1 August to 31 October 2013 | 69 | Kim et al. (2015) |
| WINTER | Wintertime aircraft campaign in the northeastern US | 1 February to 15 March 2015 | 80 | Shah et al. (2019) |
| WINTER | Wintertime aircraft campaign in the northeastern US | 1 February to 15 March 2015 | 69 | Schroder et al. (2018) |
| | | OFR experiment | | |
| CalNex | Urban outflow from Los Angeles | 29 May to 10 June 2010 | 45-68 (43) | Ortega et al. (2016) |
| KORUS-AQ | Aircraft campaign over South Korea and the Yellow Sea | May to June 2016 | 129 ± 16.5 | Nault et al. (2018) |
| Changping | Suburban near Beijing | 12 to 21 June 2016 | 59 | J. Li et al. (2019b) |
| Hong Kong | The east coast of Hong Kong | 11 to 21 October 2016 | 22 | |
| Pittsburgh | A highway tunnel | May 2013 | 91 | Tkacik et al. (2014) |
| Hong Kong | 1 m from a major road | 24 Dec 2017 to 15 Jan 2018 | 48 (38 ± 65) | T. Liu et al. (2019) |
| Durham | 10 m from the edge of Interstate 40 | 1 to 15 July 2015 | 180 (62 ± 18) | Saha et al. (2018) |
| | | 5 to 25 February 2016 | 110 (20) | |
| Beijing | The 4th Ring Road | November 2018 | 44 | Liao et al. (2021) |

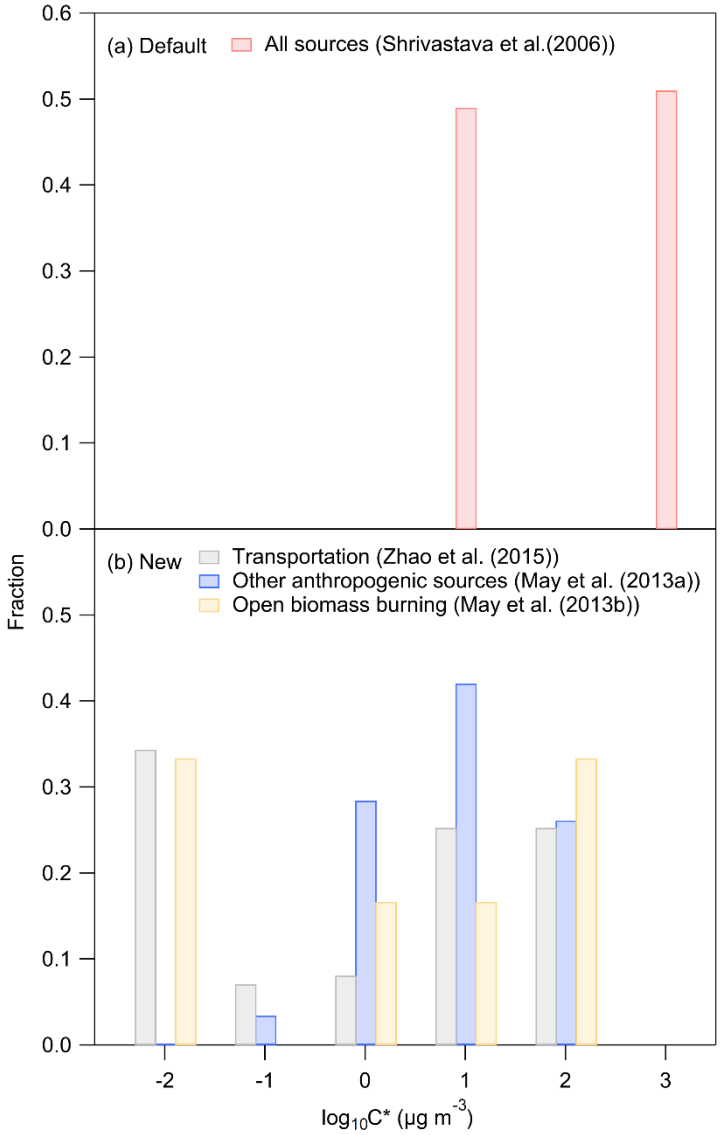


**Figure S1.** The (a) default and (b) new volatility distributions of primary SVOCs that used in the Cp_base and Cp_R1 simulations, respectively.

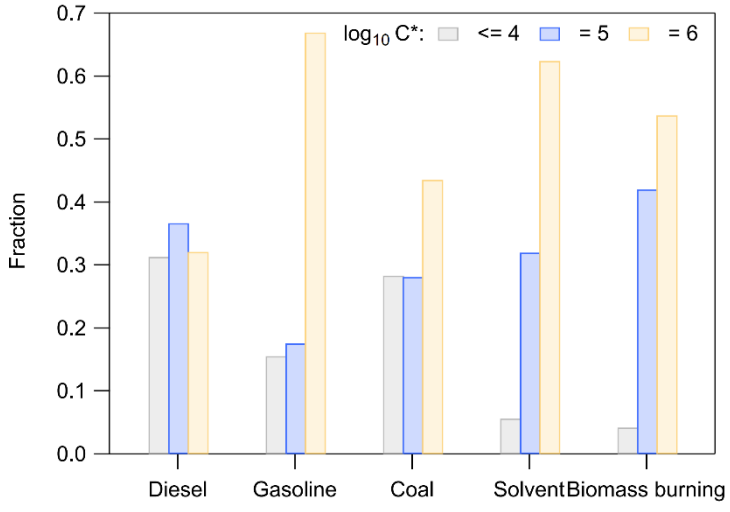


**Figure S2.** The volatility distributions of primary IVOCs from diesel, gasoline, coal combustion, solvent uses, and biomass burning. The volatility distributions from diesel and gasoline are adopted from Lu et al. (2018). For coal combustion, solvent uses, and biomass burning, the volatility distributions are provided by Cai et al. (2019), Khare and Gentner (2018), and Lim et al. (2019), respectively.

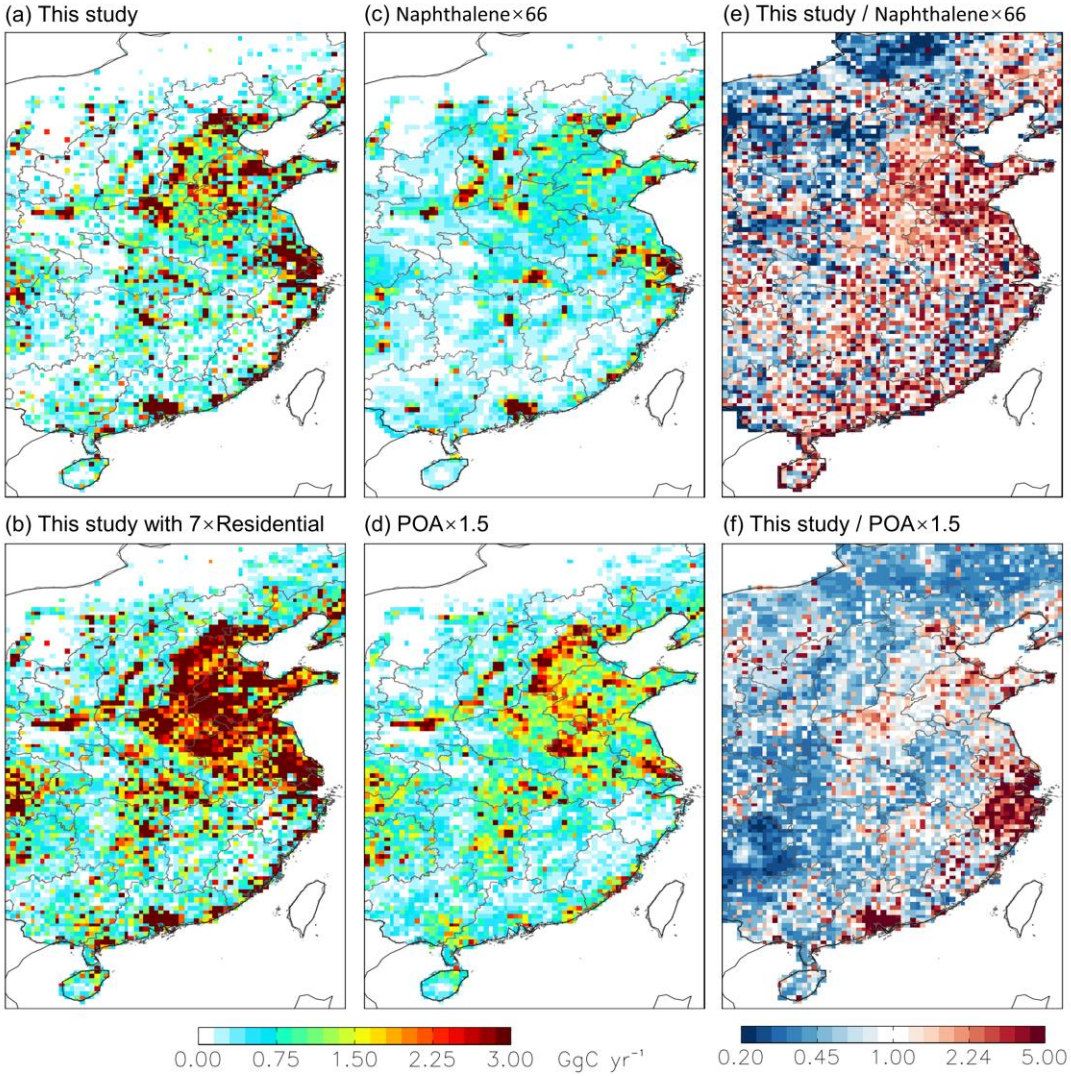


**Figure S3.** Annual emissions of IVOCs in 2014 that are estimated by (a) the new method developed in this study, (b) the new method with 7-fold emissions from the residential sector during the heating season, (c) the naphthalene-based, and (d) the POA-based methods as well as (e-f) the emission ratios of the new method to naphthalene-based and the new method to POA-based estimates.

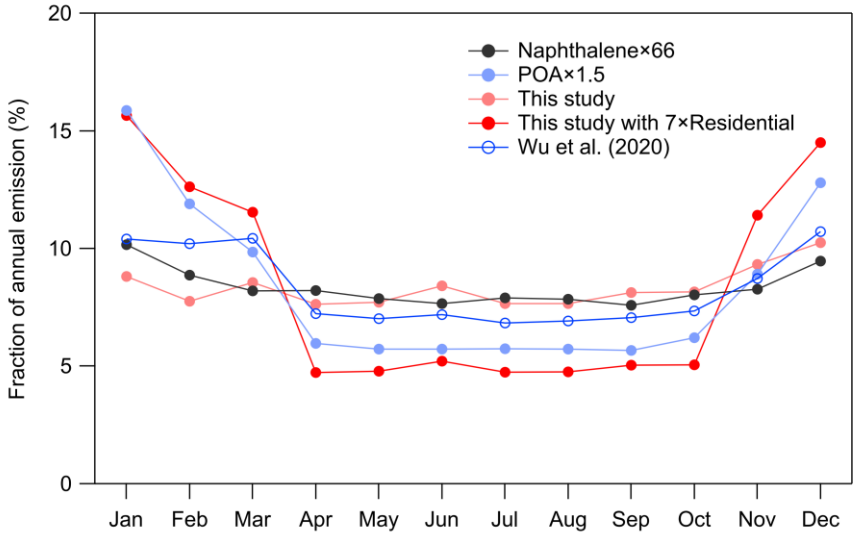


**Figure S4.** Comparisons of monthly profiles of the IVOC emissions in China in 2014.

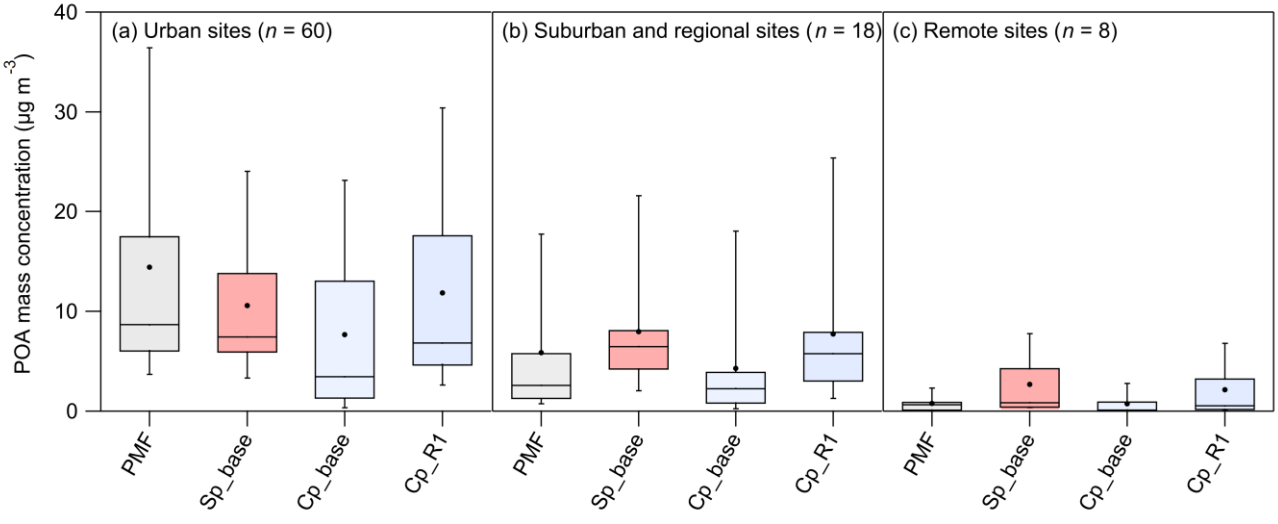


**Figure S5.** The PMF-derived and simulated campaign-average mass concentrations of POA at (a) urban, (b) suburban and regional, and (c) remote sites. The upper and lower edges of the boxes, the whiskers, the middle lines, and the solid dots denote the 25th and 75th percentiles, the 5th and 95th percentiles, the median values, and the mean values of the POA concentrations.

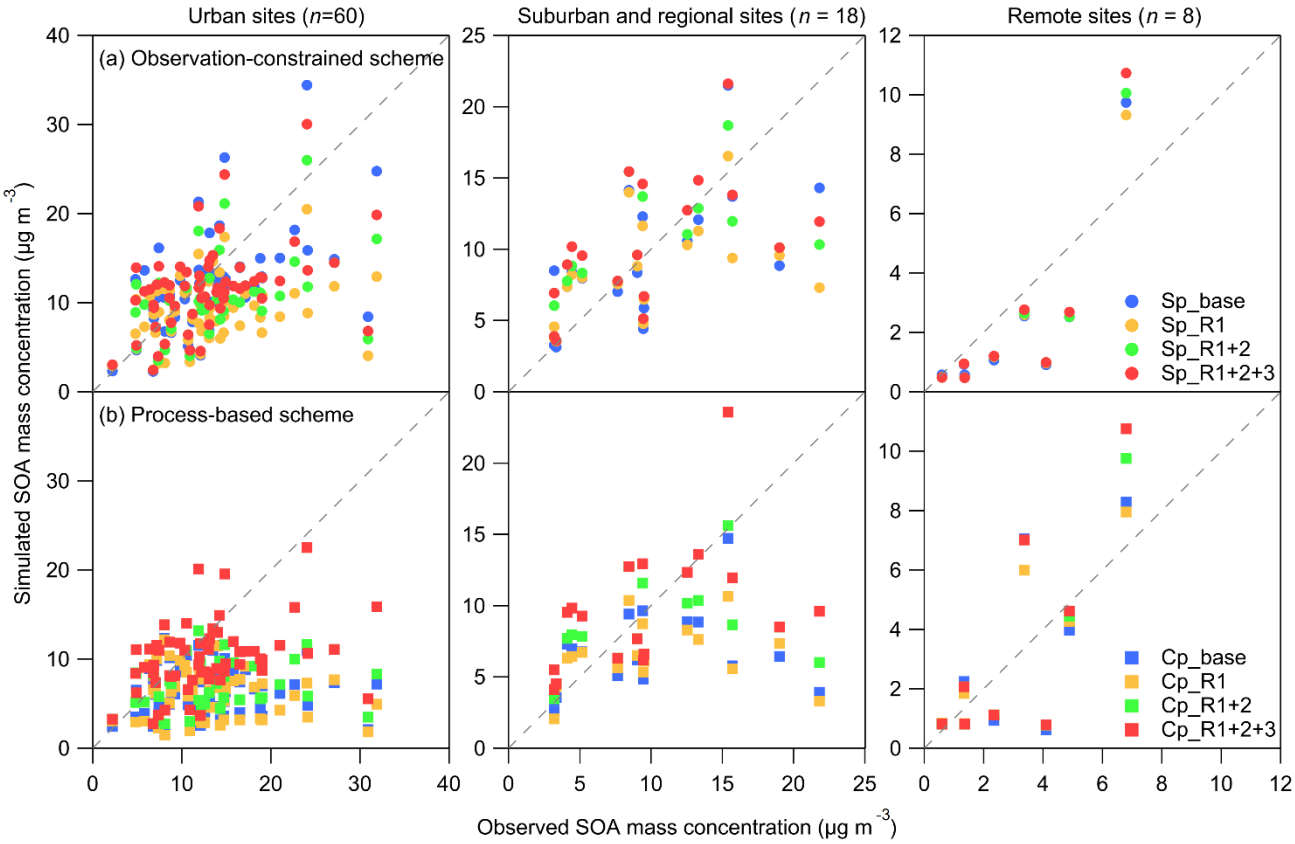


**Figure S6.** Scatter plots of the observed and simulated campaign-average concentrations of SOA by (a) the observational-constrained scheme and (b) the process-based scheme at urban, suburban and regional, and remote sites.

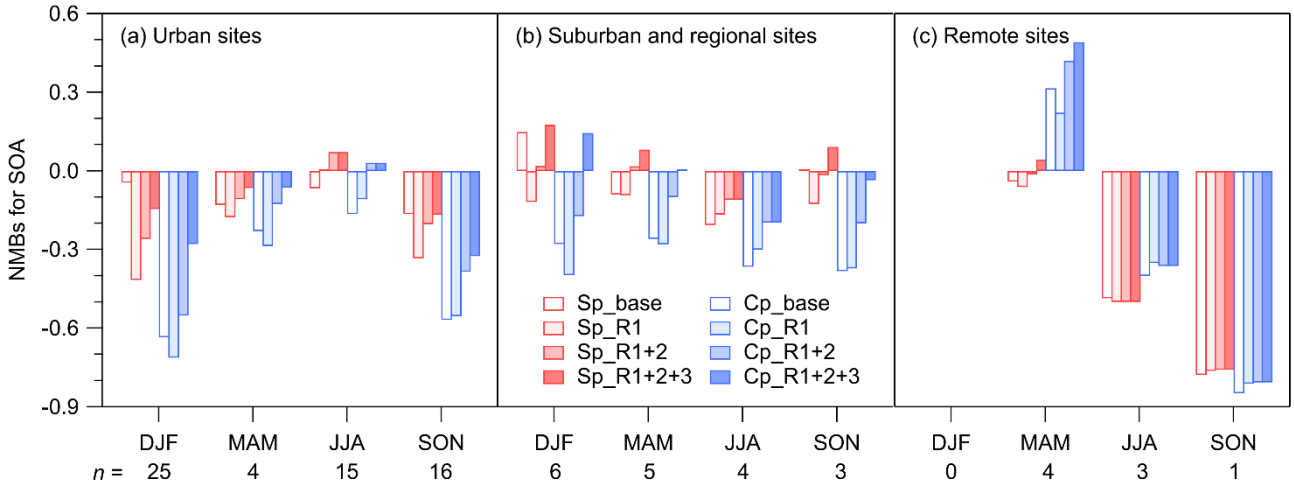


**Figure S7.** NMBs for SOA simulations at (a) urban, (b) suburban and regional, and (c) remote sites for different seasons. The model results were compared to the PMF-derived SOA results in China.

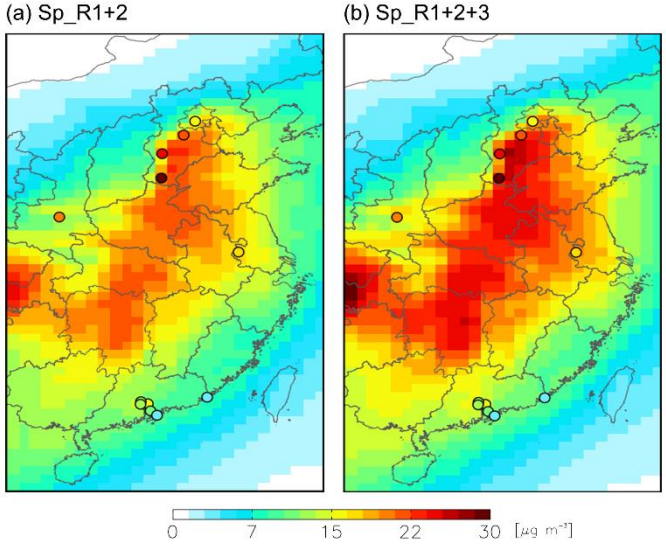


**Figure S8.** Winter-mean mass concentrations of SOA simulated by the Sp_R1+2 and Sp_R1+2+3 simulations. The circles are shown for the PMF-derived SOA concentrations from measurements.

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