

Interactive comment on “Formation of Nighttime Sulfuric Acid from the Ozonolysis of Alkenes in Beijing” by Yishuo Guo et al.

Anonymous Referee #2

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This paper presents gas phase sulfuric acid measurements from Beijing during winter and summer with CIMS and show some nighttime formation of sulfuric acid, possibly from sCI formed from VOCs ozonolysis reactions. Sulfuric acid measurements are valuable –considering roles of sulfuric acid on new particle formation especially in urban environments. But data analysis is not effective – please see comments below. The other conclusion is nighttime sulfuric acid is responsible for sub-3 nm particles. How did the authors exclude HOMs from sub-3 nm particle formation?

Section 4.3. I have never seen a kinetic expression like this: $k_{app} \cdot [\text{Alkene}] \cdot [\text{O}_3] \cdot [\text{SO}_2]$. This does not make sense to me. Please show how k_{app} is the same as $k_1 \cdot k_2 \cdot \varphi$? And what about k_3 , k_4 and k_5 then? Does this mean nighttime sulfuric acid is not from $\text{OH} + \text{SO}_2$ reaction (OH from VOCs ozonolysis)? The authors assume

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$\varphi \cdot f$ to be 0.05 – these are random numbers. The dimer formation rate β is $3.46 \times 10^{-3} \text{ cm}^3 \text{ s}^{-1}$ – isn't it too high? And what about the sink due to formation sulfuric-amines or sulfuric acid-ammonia clusters? And if this VOCs does not include monoterpenes and isoprene (emitted from biogenic emissions in summer and from volatile personal products year around), then how is the source-sink discussion really useful? I suggest the authors make steady state calculations of sCI and OH (or use a box model) to simulate nighttime sulfuric acid concentrations.

Please change Figure 1 in linear scale (Y axis) – as opposed to log scale, as a more common practice in the field (or adding inset in log scale). If I look at Figure s1 (move this to the main text or replace Figure 1 with this), it seems that nighttime sulfuric acid is quite minor. So I am not sure with the conclusion of nighttime sulfuric acid formation. Did authors measure only gas phase monomer sulfuric acid? Does this also include the sulfuric acid-amine cluster? Do they have measurements of dimer, trimer, tetramer of sulfuric acid, ammonia and amines during this period to show that sub-3 nm particles are really from sulfuric acid clusters? Please also include size distributions to show if there are new particle formation or not during the night.

Figure 5. Please also show HOMs in the same way as sulfuric acid, and include daytime data as well (vs. nighttime). Did sub-3 nm particles grow further? If they did not grow larger, then what are the possible explanations? Can you calculate J from PSM? What is p (power dependence) of J or sub-3 nm particles on nighttime sulfuric acid (vs. daytime)? Is p different during day and night?

Line 40: Needs refs., e.g., [Lee et al., 2019].

Line 43: Please include [Yu et al., 2012].

Line 51: please include [Erupe et al., 2010] and [Yu et al., 2013]

Line 54: needs refs. Is this statement true? I hardly see 7×10^{-3} level of sulfuric acid.

Line 60: please include [Yu et al., 2013].

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Line 64: Change “frequent and noticeable” to “noticeable nighttime sulfuric acid sometimes”.

Line 105: This is a very long inlet. What is the residence time, and radius? Wall loss rate in the inlet?

Line 134: Calibration once a month? This is really infrequent! How frequently did they make background measurements?

Line 143/165: Regardless, SO₂ seemed to me always at the ppb in average, so in high SO₂ conditions year around.

Figure 2. Please show amines and ammonia.

Line 191-192: This does not make sense to me.

Figure s1: move this to the main text – including spring measurements.

Figure s3: the same as Figure 3?

Erupe, M. E., et al. (2010), Correlation of aerosol nucleation rate with sulfuric acid and ammonia in Kent Ohio: an atmospheric observation, *J. Geophys. Res.*, 115, Doi:10.1029/2010JD013942, doi:10.1029/2010JD013942.

Lee, S.-H., H. Gordon, H. Yu, K. Lehtipalo, R. Haley, Y. Li, and R. Zhang (2019), New Particle Formation in the Atmosphere: From Molecular Clusters to Global Climate, *Journal of Geophysical Research: Atmospheres*, 124(13), 7098-7146, doi:10.1029/2018JD029356.

Yu, H., et al. (2013), Sub-3 nm particles observed at the coastal and continental sites in the United States, *J. Geophys. Res.*, 119, Doi: 10.1029/2013JD020841, doi:10.1029/2013JD020841.

Yu, H., R. McGraw, and S. H. Lee (2012), Effects of amines on formation of sub-3 nm particles and their subsequent growth, *Geophys. Res. Lett.*, 39, Doi:

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10.1029/2011gl050099, doi:10.1029/2011gl050099.

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