RC1 Comments:

The manuscript is well-written, and conclusions are well-supported by the experimental data. I recommend this work for publication, and some minor comments can be considered.

Q1-1. *In the introduction, the authors justify the use of squalene as a model system. However, a brief statement for selecting ethylamine as the model amine, compared to more commonly studied amines (e.g., ammonia or dimethylamine), can be helpful. Is it due to its atmospheric relevance, reactivity, or as a simpler model for primary amines?*

Reply1-1: Ethylamine is abundant in the atmosphere (Atmos. Environ. 2013, 71, 95-103). It exhibits higher heterogeneous reactivity towards the aerosols studied in this work. Its simple structure also makes it a good model for studying primary amination mechanisms, avoiding the complexity of secondary/tertiary amine groups. We have added a justification at Line 67 on Page 3 of the revised manuscript as follows.

"As a representative atmospheric amine (Lee and Wexler, 2013), ethylamine was selected for its remarkable heterogeneous reactivity and simple structure."

Q1-2. *The use of APPI-HRMS is critical to the identification of intermediates and products. To facilitate a better assessment of the methodology, it is suggested to provide key instrumental parameters in the supplementary information. These should include, but not be limited to: the ionization mode used (positive/negative), mass resolution, mass range scanned, and vaporizer temperature.*

Reply1-2: We have added more details about the APPI-HRMS on Page S5 in the revised *Supplementary Information* as follows.

"The mass resolution of HRMS (Orbitrap Fusion, Thermo Scientific) is 500,000 at *m/z* 200. During the experiments, the MS mode was set to positive polarity. Meanwhile, the ion transfer tube was maintained at 300 °C. The mass range from *m/z* 75 to 800 was scanned with a rate of 30 spectra/min, and the resulting data were collected using Xcalibur 4.0 software."

Q1-3. *The mass spectrum in Figure 3b would be more clearly distinguishable if the different product classes (adducts, -H_2O, -H_2O_2) were labeled with different colors or symbols directly on the spectrum.*

Reply1-3: Figure 3b along with its corresponding caption has been revised in the manuscript as follows.

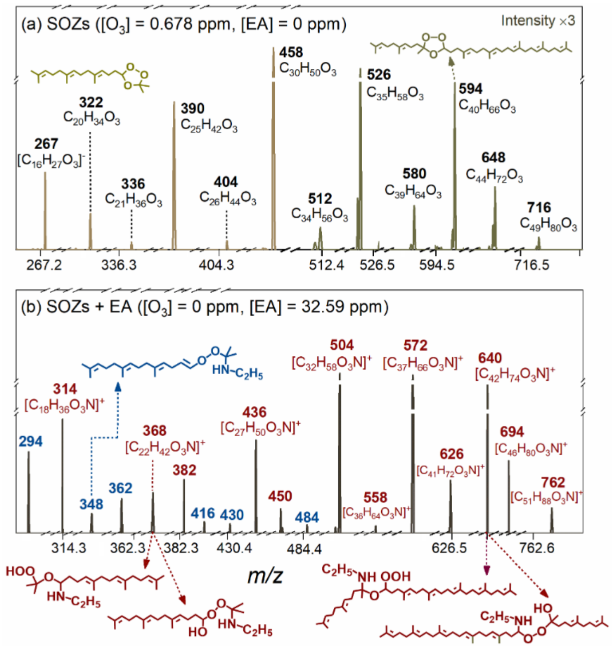

Figure 3: Mass spectra of (a) representative SOZs formed during the ozonolysis of Sqe in the first flowtube reactor, and (b) their amination products (adducts of SOZ + EA are in red and dehydration products are in blue) upon exposure to ethylamine (EA) in the secondary flowtube reactor.

***Q1-4**. The identification of the four key intermediates is a core finding of this work. For clarity, it is suggested to add a simplified schematic flowchart, either in Figure 4 or as a supplementary figure, to summarize the competing pathways and subsequent consumption pathways. This would facilitate visualization of the complex reaction network.*

Reply1-4: A simplified schematic flowchart has been added as Fig. S15 in the revised *Supplementary Information*. Meanwhile, an introduction for this Fig. S15 has been added at Line 282 on Page 15 in the revised manuscript.

"Figure S15 summarizes a simplified mechanism involving four key intermediates, including hydroxyl peroxyamines, peroxyamines, amino hydroperoxides, and amino ethers."

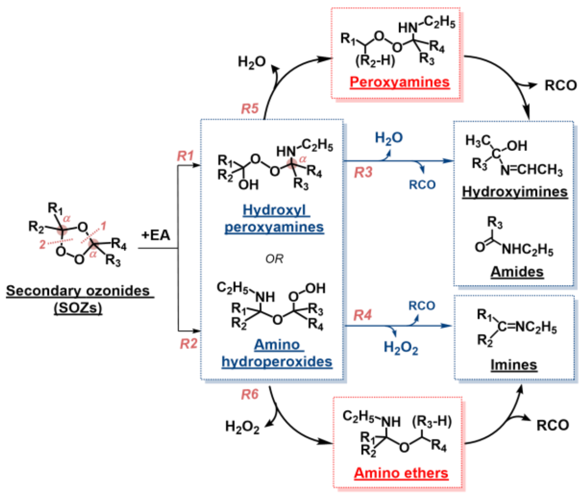

Figure S15. A simplified mechanism of SOZs upon EA exposure.

Q1-5. *For the newly proposed intermediates (peroxyamines and amino ethers), it is suggested to provide their exact mass and compare them with the experimentally observed m/z values in a supplementary table. Listing representative intermediates and products observed for each SOZ in this table can provide additional evidence for their identification.*

Reply1-5: A new Table S1 has been added to the revised *Supplementary Information*, which lists the representative products, including their molecular formulas, representative structures, theoretical exact mass, and experimentally observed *m/z*.

Technical corrections:

There are inconsistencies in the terminology used for differential effective uptake coefficients, which are sometimes written as "$\Delta\gamma_{eff}$", and sometimes abbreviated as "$\Delta\gamma$".

Reply1-6: To ensure consistency, we have represented the differential effective uptake coefficients as $\Delta\gamma_{\mathrm{eff}}$ in the revised manuscript and *Supplementary Information*.

Line 203, "see Sect. 3.2 for mechanistic details," but the mechanistic details are in Sect. 3.3. Please correct it.

Reply1-7: This word "Sect. 3.2" has been revised to "Sect. 3.3" on Page 9 of the revised manuscript.

Line 254, "has be demonstrated", this should be correct to "has been demonstrated".

Reply1-8: We have corrected this.

Line 268, "$C_{32}O_{55}O_2N$ (peroxyamine)" should be corrected to $C_{32}H_{55}O_2N$.

Reply1-9: We have corrected this to "$C_{32}H_{55}O_2N$".

Line 269, "$C_{32}O_{57}O_3N$" should be corrected to "$C_{32}H_{57}O_3N$".

Reply1-10: We have corrected this to "$C_{32}H_{57}O_3N$".