The authors thank the reviewer for the careful review of our manuscript and the helpful comments and suggestions. All the comments (in black) are addressed point by point, with our response in blue, and the corresponding revisions to the manuscript in red.

Review # 1

This article provides a detailed review of existing methods and introduces a new analytical method called WALL-E, which is designed to measure the chemical composition of atmospheric particles in real time. The work is significant and innovative, but the readability of the article needs improvement. Here are the specific details:

1. Introduction: The Introduction compares many methods, but it doesn't clearly state that the main contribution is adding a device that integrates TD (thermal desorption) and other functions in front of the Br-CIMS. This might be confusing for users of the instrument, especially those who are not developers, as they may struggle to quickly understand the research goal. In addition, the introduction of VIA-NO_3-CIMS is too simplistic and needs to be strengthened.

We added and modified the last paragraph of the introduction to clarify the research goal of this manuscript:

Lines 106-109: *"Currently, there is no technique based on thermal evaporation able to prevent thermal fragmentations, and suitable for the on-line measurement of moderate oxygenated (e.g., molecular oxygen atoms <6) organic species."*

Lines 123 – 126: "*In this work, we designed a newly designed wall-free particle evaporator (WALL-E) to perform online organic particle characterization while preventing ionization-induced fragmentation and minimizing thermal decomposition effects. WALL-E is coupled to a chemical ionization inlet attached to a CIMS to achieve real-time measurements of aerosol particles at a molecular level."*

We also strengthened the introduction of VIA-NO3-CIMS:

Lines 98-103: *"..., which mainly consists of a sulfinert-coated stainless steel as TD unit and a following cold dilution flow of N_2. The evaporation tube of VIA is bonded with an insert silica layer into the surface; a dilution flow is used to cool down the sampling flow and minimize the recondensation of the evaporated compounds. The parameters of the dilution unit are critical factors that affect the final sensitivity of the entire system."*

2. Abstract: The phrase "suffers from different artifacts" in the Abstract is too vague. It should specify what kind of artifacts WALL-E addresses.

We clarify it by mentioning specific artifacts.

Line 25-26: *"suffers from different artifacts (i.e., thermal decomposition, fragmentation, wall loss)."*

3. Undefined Term in Abstract: T_{50} is not defined in the Abstract.

T_{50} is now defined in the Abstract.

Line 40-41: *"In addition, the measured T_{50} (the temperature at which 50% of a compound evaporates), for α-pinene-derived SOA …"*

4. Figures 1 and 2: Combining Figures 1 and 2 would make it easier to see both the appearance of the TD and the airflow simulation inside. It would also be helpful to label the "Dilution/Cooling Unit" and "Gas-phase Denuder."

We appreciate the reviewer's suggestion. In response, we have revised Figure 1 to include a new panel (Figure 1B) that clearly defines all key regions of the system, including the "Dilution/Cooling Unit" and the "Gas-phase Denuder," as well as the direction and entry points of the different flows. This provides a clearer visual understanding of the setup and complements the airflow simulation shown in Figure 2.

Lines 144-149:

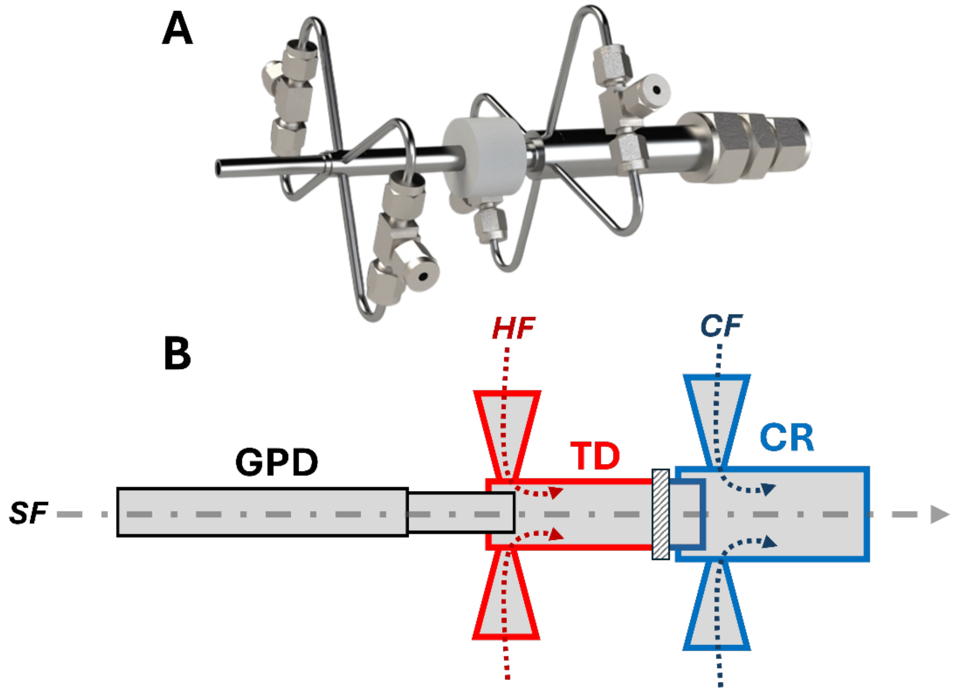

Figure 1: (A) Design of the WALL-E interface. (B) Schematic of WALL-E with a gas-phase denuder (GPD) connected to the inlet. The thermal desorption region (TD) is shown in red, where the hot flow (HF) is mixed with the sample flow (SF). A ceramic spacer is indicated by the dashed rectangle. The cooling region (CR), shown in blue, is where the cooling flow (CF) is introduced.

5. Lines 346-363: While it's possible to guess what DC and dV_{50} mean, it would be clearer if their full names were provided.

The full names of DC (direct current) and dV_{50} (voltage difference at half signal maximum intensity) are now added to the main text.

Lines 370 – 375: *"By utilizing the in-source collision ion dissociation feature (Riva et al., 2019), which corresponds to an increase in the direct current (DC) offset voltages between two ion optics within the flatapole, the binding energy of the $[M\text{-}Br^-]$ adducts can be probed (Figure*

S7). The voltage difference at half signal maximum intensity (dV_{50}) broadly ranges from 5.1 to 20.2 Volts, indicating differences in binding energies and varying clustering strengths to Br^-."

6. Lines 377-381: The explanation here is not clear or intuitive enough in the following.

The sentence is rewritten.

Line 402-406: *"Among all calibrated compounds, two outliers exist (i.e., shikimic acid and glucose), which might be due to partial evaporation leading to an underestimation of the sensitivity. When excluding these two species, the estimate of total particle mass concentration is closer (with 36% overestimation) to the total particle mass concentration measured by the SMPS as depicted in Figure 5."*

7. Lines 407-418, Lines 430-438: This part repeats information from the Introduction. It should be shortened and focus on the core issues. Some part is better for Introduction

To improve readability and reduce redundancy, we have made the following changes:

A shorter version of the general discussion on volatility determination was added to better introduce the challenges and context of our work:

Lines 119-122: *"Additionally, for highly oxygenated and multifunctional compounds, volatility determination remains particularly uncertain, as isomerism and intermolecular interactions can significantly influence evaporation behavior (Lee et al., 2014; Bannan et al., 2019)."*

Section 3.3.1 (Thermograms and T_{max} Determination):

The general discussion on volatility determination was removed to avoid repeating content already presented in the introduction. This section now focuses directly on the experimental results and the specific improvements provided by WALL-E.

Lines 433-446: *"The determination of volatility represents one of the greatest analytical challenges when characterizing aerosol particles, as it depends on multiple factors, including molecular composition, intermolecular interactions, and experimental conditions (Compernolle et al., 2011). Various experimental and theoretical techniques have been developed over the last decades to retrieve volatility information, each with advantages and limitations.*

While FIGAERO, VIA have been widely used, their design constraints introduce inherent limitations, can introduce artifacts such as recondensation, analyte interactions, and fragmentation. The prolonged residence time on the FIGAERO may also lead to early desorption of volatile species or chemical reactions between co-deposited compounds, impacting the accuracy of volatility estimates (Stark et al., 2017; Schobesberger et al., 2018; Buchholz et al., 2020). VIA thermograms, on the other hand, show evidence of fragmentation and thermal decomposition at high temperatures (Zhao et al., 2024b). WALL-E introduces a new approach, optimizing the balance between thermal residence time and evaporation efficiency, allowing for precise volatility determination with reduced wall interactions."

Section 3.3.2 (From T_{max} to T_{50}):

The Title was changed

Line 447: “T_{50} as a Robust Volatility Metric”

No changes were made in the paragraph. We believe this section is essential to clearly explain and justify the introduction of T_{50} as a key innovation in this work.

8. Line 474: The statement “The corrected values align well with those reported for VIA and FIGAERO” is too general. It should include references and specific results.

The corresponding values and references for the VIA and FIGAERO data are provided in Figure S14 of the Supplementary Information, the sentence was modified as follow:

Lines 500-503: *“The corrected values align well with those reported for VIA and FIGAERO, maintaining a consistent trend across different molecular weights (Figure S14).”*

9. Section 3.3: The main innovation is the use of T_{50}, but the subsection titles are confusing. T_{max} is discussed in Sections 3.3.1 and 3.3.3, while T_{50} is in Sections 3.3.2 and 3.3.4. This arrangement makes it hard for readers to follow.

To clarify the structure and maintain consistency with previous studies, we have added a transition sentence at the beginning of the Temperature Correction and Comparison section. This explains why T_{max} is used for the correction and inter-comparison before introducing the improvements provided by T_{50}.

Lines 490-493: *“Although T_{50} is introduced as a more robust metric in this study, temperature correction and inter-comparison with other techniques have traditionally been performed using T_{max} values. Therefore, we present the correction and comparison based on T_{max} to ensure consistency with previous studies before applying the T_{50} approach.”*