Simulation of water isotopes in combustion-derived vapor emissions in winter

Yan Yang¹ and Kei Yoshimura¹

¹University of Tokyo

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Abstract

With urbanization, anthropogenic water vapor emissions have become a significant component of the urban atmosphere. Fossil fuel combustion-derived vapor (CDV) is a primary source of these emissions. Owing to the notably low CDV d-excess, stable hydrogen and oxygen isotopes are promising for distinguishing CDV from natural sources. Considering the limitations of in situ observations, this study aims to explore the feasibility of using IsoRSM, an isotopically enabled regional atmospheric model, to simulate CDV emissions in urban areas in winter. Two experiments were conducted: one in Salt Lake City in January 2017 and another in Beijing in January 2007. The simulation results showed that the CDV addition significantly reduced the water vapor d-excess, particularly when the boundary layer was stable. The simulation with CDV emissions aligned better with the time series of in situ observations in Salt Lake City. The modification led to a more pronounced positive correlation between vapor d-excess and specific humidity, which was similar to the observation of Salt Lake City. The CDV inclusion significantly increased the vapor d-excess variability with varying wind directions in both sites. However, in Beijing, the underestimation of d-excess variation from natural sources caused a bigger discrepancy between the observed and simulated d-excess and CDV fraction. Thus, though there were still biases, the inclusion of CDV could improve the accuracy of isotopic simulation in the urban regions where CDV was one of the controlling factors of vapor d-excess.

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5	emissions in winter			
6 7 8 9	Yan Yang ¹ and Kei Yoshimura ²			
10	¹ Graduate School of Frontier Sciences, The University of Tokyo			
11 12 13 14	² Institute of Industrial Science, The University of Tokyo			
15	Key points			
16 17	• The emission of combustion-derived vapor (CDV) was simulated with the isotopic-enable regional spectral model.			
18 19	• The CDV addition reduced the water vapor d-excess and made the simulation align better with the observations in Salt Lake City.			
20 21	• The CDV inclusion significantly increased the vapor d-excess variability with varying wind directions.			
22				

23 Abstract

With urbanization, anthropogenic water vapor emissions have become a significant 24 component of the urban atmosphere. Fossil fuel combustion-derived vapor (CDV) is a 25 primary source of these emissions. Owing to the notably low CDV d-excess, stable hydrogen 26 and oxygen isotopes are promising for distinguishing CDV from natural sources. Considering 27 the limitations of in situ observations, this study aims to explore the feasibility of using 28 IsoRSM, an isotopically enabled regional atmospheric model, to simulate CDV emissions in 29 urban areas in winter. Two experiments were conducted: one in Salt Lake City in January 30 2017 and another in Beijing in January 2007. The simulation results showed that the CDV 31 addition significantly reduced the water vapor d-excess, particularly when the boundary layer 32 was stable. The simulation with CDV emissions aligned better with the time series of in situ 33 observations in Salt Lake City. The modification led to a more pronounced positive 34 correlation between vapor d-excess and specific humidity, which was similar to the 35 observation of Salt Lake City. The CDV inclusion significantly increased the vapor d-excess 36 variability with varying wind directions in both sites. However, in Beijing, the 37 underestimation of d-excess variation from natural sources caused a bigger discrepancy 38 between the observed and simulated d-excess and CDV fraction. Thus, though there were still 39 biases, the inclusion of CDV could improve the accuracy of isotopic simulation in the urban 40 regions where CDV was one of the controlling factors of vapor d-excess. 41

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44 Plain Language Summary

Combustion-derived vapor (CDV) generated from fossil fuel is one of the main sources of 45 the anthropogenic water emission in urban areas. Water d-excess, which is calculated with 46 the water isotopes of oxygen (δ^{18} O) and hydrogen (δ^{2} H), is usually be used to distinguish the 47 water from different sources. Due to the extreme low d-excess of CDV, we can use the water 48 isotopes to partition CDV from naturally evaporating water. Different from previous 49 researched on CDV based on the in-situ observed data, this study aims to simulate the 50 isotopic composition of CDV in urban areas with the atmospheric model. The results of 51 experiments in Salt Lake City and Beijing indicates that CDV can be the one of the 52 controlling factors of the vapor d-excess in some urban regions, and the inclusion of CDV in 53 the model will improve the accuracy of simulation in these regions. Therefore, this approach 54 has great potential for making the simulation of water isotopes on a global scale with the 55 impact of the anthropogenic emissions. 56

57

58 1 Introduction

The impact of anthropogenic emissions on the hydrological cycle in urban areas has 59 become increasingly significant with urbanization (Moriwaki et al., 2008; Tie et al., 2017; 60 Ueyama et al., 2021). In winter, the specific humidity in the boundary layer over cities is 61 often approximately 10% higher than that over the surrounding rural areas (Salmon et al., 62 2017). This phenomenon can be attributed to various factors, including vapor emissions from 63 anthropogenic sources (Li et al., 2021). One major contributor to anthropogenic water vapor 64 emissions is the combustion of fossil fuels. According to the annual carbon emissions and the 65 emission ratio of CO₂ to H₂O (Gorski et al., 2015), fossil fuel combustion releases 66 approximately 21.1 Pg/a of water into the atmosphere. Although this combustion-derived 67 vapor (CDV) is negligible at the global and annual scales in the hydrologic cycle 68 (Huntington, 2006; Loaiciga A' et al., 1996), it plays a significant role in urban hydrologic 69 cycling and meteorology because of the concentrated spatial and temporal distributions of 70 71 fossil fuel emissions (Bergeron & Strachan, 2012; Sailor, 2011).

CDV can affect the urban air quality and meteorology; it can also directly affect the 72 radiative balance by increasing the water vapor concentrations, influencing the aerosol and 73 cloud properties, and modifying the local or downwind precipitation (Carlton & Turpin, 2013; 74 Fiorella et al., 2018; McCarthy et al., 2010; Rosenfeld et al., 2008). Previous studies have 75 demonstrated that CDV contributes on average an additional 4.6 µg m⁻³ of PM2.5 during 76 severely polluted conditions in the Guanzhong Basin, China. This additional CDV-induced 77 PM2.5 corresponds on average to 5.1% of the local anthropogenic PM2.5 (Xing et al., 2020). 78 The observation in Beijing also revealed that the CDV increases with aggravation of haze 79 (Wu et al., 2022). Temporal variations of the fraction of CDV showed an obvious correlation 80 with the intensity of human activities (Liu et al., 2022). 81

Conducting standard meteorological measurements is difficult to isolate the anthropogenic 82 vapor from natural sources. However, during inversion periods in winter, there is a notable 83 negative correlation between vapor d-excess ($d = \delta^2 H - 8 \times \delta^{18} O$) and atmospheric CO₂ 84 concentration (Gorski et al., 2015). Laboratory combustion experiments have demonstrated 85 that the CDV d-excess ranges from -470‰ to -180‰, thereby being significantly lower than 86 that of other natural sources (Blossey et al., 2010; Dansgaard, 1964; Fiorella et al., 2015). 87 Due to this distinct difference, stable water vapor isotopes offer a promising approach for 88 differentiating between observed water vapor originating from combustion and advection 89 90 sources.

In 2008, the Isotope-Enabled Global Spectral Model (IsoGSM) was introduced as a 91 three-dimensional stable water isotope model that operates at the global scale and covers 92 multiple decades of data (Kei Yoshimura & Kanamitsu, 2008). This model considers both 93 kinetic and equilibrium fractionation processes to determine the isotopic ratios in vapor, 94 while the complex atmospheric processes are considered by incorporating a spectral nudging 95 96 technique. A regional version of IsoGSM called Isotope-Enabled Regional Spectral Model 97 (IsoRSM) was also developed, offering higher temporal and spatial resolutions (Kei Yoshimura et al., 2010); however, water emitted from fossil fuel combustion is not explicitly 98 considered in this model. Therefore, the influence of anthropogenic vapor emissions on the 99 model results has not been verified or validated, with the model focusing primarily on natural 100 101 processes and not considering the impact of human-induced emissions on stable water 102 isotopes.

Most current studies on CDV focus on in situ measurements and observations; however, 103 conducting continuous in situ observations over a long-time scale often involves various 104 limitations (Wei et al., 2019). To propose a new approach to study on the impact of CDV and 105 106 make further analysis on CDV and other meteorological factors, this study aimed to develop 107 a CDV emission model within the IsoRSM and investigate whether incorporating CDV emissions improved the accuracy of isotopic simulations. The results were compared with in 108 109 situ observational data to assess the performance of the model. Furthermore, the fractionation of CDV in the atmosphere was calculated to examine whether the boundary layer conditions 110 affected the CDV dispersion. 111

112 2 Methodology

113 2.1 Model simulation

Yoshimura et al. (2010) introduced the IsoRSM, which is an enhanced version of the 114 regional spectral model of the Scripps Experimental Climate Prediction Center (Kanamitsu et 115 al., 2005). The IsoRSM incorporates the isotopic species of water vapor as tracers. In this 116 model, no isotopic fractionation occurs during dynamic advection, ice and snow sublimation, 117 terrestrial evapotranspiration (where 100% transpiration is assumed), and runoff processes. 118 thermodynamic equilibrium fractionation occurs during 119 However, most phase 120 transitions-such as condensation and evaporation-among the gaseous, liquid, and solid phases of water under saturated conditions (Majoube, 1971a,b.). In addition, kinetic 121 fractionation is considered for evaporation and isotopic exchange between open water and 122 123 liquid raindrops with ambient air under unsaturated conditions, as well as condensation from 124 vapor to ice at temperatures below -20 °C under supersaturated conditions (Xiaoyang Li et al., 125 2021: Merlivat & Jouzel, 1979).

The IsoRSM utilizes a spectral nudging technique to enhance the simulation of 126 atmospheric circulation and improve the realism of the model (Kanamaru & Kanamitsu, 127 2007). This technique involves forcing large-scale temperature and wind fields in the model 128 towards the reanalysis forcing fields. Thus, the model captures broader atmospheric 129 circulation patterns and their interactions with the global climate system. On the contrary, 130 131 small-scale details, which are influenced by local geographic features, such as topography, land-sea distribution, and land use, are considered in the IsoRSM. This allows the model to 132 maintain the local characteristics and capture the interplay between atmospheric circulation 133 134 and the specific geographic features of the region, thereby contributing to a more accurate representation of the regional climate (Kanamitsu et al., 2010; Kie Yoshimura & Kanamitsu, 135 136 2009).

To address the Gibbs phenomenon, the Non-iteration Dimensional-Split Semi-Lagrangian (NDSL) advection scheme was implemented in IsoRSM. The Gibbs phenomenon arises when positive-definite quantities, such as moisture and tracers, generate negative values owing to the spectral space transformation employed in spectral model systems. To overcome this issue, the spectral prognostic specific humidity and radioactive tracer advection schemes in the IsoRSM were replaced with the NDSL advection scheme. Unlike spectral space transformations, the NDSL scheme considers the advection of tracers within a grid system without spectral calculations. By employing the NDSL scheme, negative specific humidity
values produced during spectral calculations can be eliminated without sacrificing the
detailed features and accuracy of the model. This helps to alleviate the Gibbs phenomenon
and ensures more reliable simulations using the IsoRSM (Chang & Yoshimura, 2015).

148 2.2 Observational data

Observational isotopic data from Salt Lake City (SLC) were collected by (Fiorella et al., 149 2019) between December 2016 and February 2017. To maintain data continuity, only data 150 from January 2017 were used in this experiment. The isotopic ratio of vapor in SLC was 151 measured using a Picarro L2130-i analyzer.Observational isotopic data for Beijing were 152 obtained from the Stable Water Vapor Isotopes Database (SWVID) and covered the period 153 from December 2006 to December 2007 (Wen et al., 2010). The isotopic ratio of vapor in 154 155 Beijing was measured using a Campbell TGA-100A Trace Gas Analyzer. Meteorological data 156 of these two sites, such as 2-m air temperature and 2-m specific humidity, were obtained from the ERA5 reanalysis dataset (Hersbach et al., 2020). 157

158 2.3 Experiment design

In this study, an additional quantity of vapor characterized by a fixed isotopic ratio was introduced into the evaporation process of the IsoRSM to simulate the CDV emissions. To simulate the urban area, CDV was added within a $2^{\circ} \times 2^{\circ}$ domain, whereas the rest of the domain represented natural sources without CDV emissions. This allowed to focus specifically on the CDV impact on the urban environment while maintaining a comparison with natural sources in the surrounding area.

Two experiments were conducted, one in SLC, USA (40.77° N, 111.85° W), and one in 165 Beijing, China (40.00° N, 116.38° E). Table 1 presents an overview of the key experimental 166 details. SLC is situated in Salt Lake Valley and is characterized by a basin topography on a 167 168 plateau. Previous studies have indicated that during winter, the boundary layer in this region 169 can become highly stable due to temperature inversions, resulting in the formation of "persistent cold air pools" (PCAPs). During this period, elevated levels of CO₂, vapor, and 170 pollutants are evident, leading to significantly lower d-excess values. Conversely, Beijing is 171 172 located in the North China Plain, which is significantly influenced by the summer and winter 173 monsoons. These two sites are relatively close in latitude, allowing for a comparative analysis of how the CDV impact is influenced by these distinct topographies and climates. 174 Considering the greater CDV influence in winter and the limitations of the observation period, 175 the experiments in SLC and Beijing were conducted in January 2017 and January 2007, 176 177 respectively.

The isotopic composition of the added vapor was determined based on laboratory combustion experiments conducted in Xi'an, China (Xing et al., 2020). Specifically, the δ^2 H and δ^{18} O values of the added vapor were set to -134.4‰ and 9‰, respectively. To estimate the order of magnitude of the CDV emission rate to be added, the annual CO₂ emission rate in urban areas and the emission ratio of CO₂ to CDV were considered (Hirano et al., 2015; Mckain et al., 2012; Moriwaki et al., 2008; Pisso et al., 2019). In this experiment, the CDV emission rates were set to six different values, from 0.000 to 0.003 g/m²·s, to check which rate exhibited the highest correlation with the observations. The selected emission rate aftervalidation would be used in the subsequent analysis.

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Table 1 Information and data sources of two experiments
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	SLC	Beijing	
Starting & ending time	2017/1/1 - 2017/1/31	2007/1/1 - 2007/1/31	
Domain area	39 - 41°N	39 - 41°N	
Topography	Basin	Plain	
Climate type	Continental	Monsoon	
Output time step	1 h	iour	
Boundary condition of simulation	IsoGSM		
Observed isotopic data source	(Fiorella et al., 2019)	SWVID (Wen <i>et al.</i> , 2010)	
Meteorological data source for validation	ERA5 reanalysis dataset		

189

190 2.4 CDV fraction

The CDV fraction in the total moisture is also an important variable for studying the CDV 191 impact on the urban atmosphere. In this study, two different methods were used to calculate 192 the CDV fraction. The first approach was the moisture tracer approach. In the IsoRSM, it is 193 194 possible to tag the moisture in a certain domain or amount and trace its diffusion and transportation. This approach is typically used to analyze the moisture sources of a domain or 195 event. Using this method, the CDV that was added to the model was tagged, so that the 196 specific humidity caused by the CDV could be simulated and the CDV fraction could be 197 198 directly calculated by the proportion of the CDV amount and the total humidity.

The second approach involved the use of a two-end-member mixing equation to calculate the CDV fraction in the atmosphere. By comparing the simulation results before and after the modification, it was possible to estimate the CDV contribution to atmospheric humidity. The moistening of the lower troposphere by CDV can be modeled as a mixing process between the CDV and background natural water vapor. The equation for this mixing process is as follows:

$$d_{\rm mix} = \frac{d_{\rm CDV}q_{\rm CDV} + d_{\rm nat}q_{\rm nat}}{q_{\rm mix}}.$$

Here, the subscripts CDV, nat, and mix represent the properties of the CDV, atmospheric moisture in the absence of CDV, and the values of the mixed parcel, respectively. In this study, the mixed parcel can be considered as the result after the CDV addition, whereas the atmospheric moisture without CDV can be regarded as the result before the modification. q_{mix} represents the total specific humidity in the mixed parcel. Using the constraint that $q_{mix} = q_{CDV} + q_{nat}$, the CDV fraction can be calculated as follows:

$$\frac{q_{\rm CDV}}{q_{\rm mix}} = \frac{d_{\rm mix} - d_{\rm nat}}{d_{\rm CDV} - d_{\rm nat}}.$$

 d_{CDV} was set to -206.4‰ due to the isotopic ratio of the CDV added into the model. Using this method, the maximum contribution of the CDV to the atmospheric humidity could be estimated.

Besides the second method, the CDV fraction was also calculated based on isotopic 214 observations. The approach was similar to that of a previous method, albeit with some 215 modifications. Due to the lack of d_{nat} (isotopic ratio of atmospheric moisture in the absence of 216 CDV), a different approach was employed. First, 10% of the observed data with the highest 217 218 d-excess values were selected and assumed to have no CDV impact. These data points represented the natural background without CDV influence. Subsequently, a regression line 219 was established in the ²H/¹⁸O plot representing the natural background line. More details 220 regarding this method can be found in the Appendix. It is important to note that this method 221 222 assumes that all d-excess decreases can be attributed to the CDV impact, which may not be 223 entirely realistic. Therefore, the estimation obtained using this method serves as a reference for comparison with the simulation results. 224

225 **3 Results**

226 3.1 Time series

Figure 1 presents the time series and diurnal variations of various parameters, including 227 228 the planetary boundary layer height, 2-m air temperature, 2-m specific humidity, and 2-m 229 vapor d-excess in both SLC and Beijing, and shows that the CDV addition had a negligible effect on the simulated air temperature. In SLC, the monthly mean temperature decreased 230 slightly from 269.75 to 269.49 K, whereas in Beijing, the temperature increased marginally 231 from 266.96 to 266.99 K. Conversely, the simulation indicated a more noticeable increase in 232 specific humidity after adding CDV, particularly in Beijing with an increase of 0.1230 g/kg 233 compared to the smaller increase of 0.0160 g/kg at SLC. 234

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Figure 1. Time series and diurnal variation of boundary layer height (m), 2m air temperature (K), 2m specific humidity (g/kg), and 2m vapor d-excess (‰) in SLC (a-h) and Beijing (i-p). The black lines are ERA5 reanalysis data or in-situ observation, the blue dashed lines are the simulation without CDV, and the red lines are the simulation with CDV.

Site	Experiment	Correlation coefficient	RMSE	Residual standard deviation
	Ctrl	-0.2207	20.4743	6.9503
	0.0005	0.0796	18.2682	6.3959
	0.0010	0.2690	16.3777	6.3583
Salt Lake City	0.0015	0.3537	14.8922	6.9751
	0.0020	0.4058	13.7362	7.9219
	0.0030	0.4559	12.9047	10.6402
	Ctrl	-0.0569	21.0852	20.5319
	0.0005	0.0380	30.0539	22.2266
Dettine	0.0010	0.0502	45.2486	27.8657
Beijing	0.0015	0.0536	62.4340	35.7632
	0.0020	0.0577	80.2537	44.5966
	0.0030	0.0613	116.8704	63.7893

238 Table 2 Correlation coefficient, RMSE and Residual standard deviation of observed 2m vapor d-excess and each experiment

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Table 2 lists the correlation coefficient, root mean square error (RMSE), and residual 240 standard deviations for the different experiments. In SLC, all four experiments exhibited 241 d-excess overestimations in most periods compared to the observed data. The observed 242 monthly mean d-excess value was -7.52‰, i.e., significantly lower than the simulated values 243 (from 11.75% for the control to -0.53% for $0.003 \text{ g/m}^2 \cdot \text{s}$). The CDV impact was more 244 pronounced when the planetary boundary layer height was lower, indicating a more stable 245 atmosphere. With an increase in the CDV emission rate, the correlation coefficient increased 246 owing to the increasingly significant variation. However, the residual standard deviation 247 suggested that a higher emission rate could result in a variation range higher than that 248 observed, thereby worsening the simulation performance. Overall, the 0.0015 $g/m^2 \cdot s$ 249 emission rate had the best simulating result in SLC, and was therefore used in the subsequent 250 251 analysis part. At 9:00 on January 6, 2017, the d-excess difference between the control and 252 CDV experiment peaked at approximately 33%. This finding suggests that the CDV impact was greater under conditions of low boundary layer height, low temperature, and low 253 humidity. The d-excess reduction resulted in a better fit with the observations, as evidenced 254 by the correlation coefficient increasing from -0.22 to 0.35. 255

In Beijing, prior to the CDV addition, the d-excess values exhibited a relatively stable trend compared to that of the observed data. However, the CDV introduction led to a significant fluctuation increase. Interestingly, the observed monthly mean vapor (19.19‰) was higher than that of the control simulation (14.42‰), thereby contradicting the CDV impact. Following the modification with CDV, the monthly mean d-excess sharply decreased



Figure 2. Monthly mean 2m vapor d-excess (‰) and the differences between modified experiments and control one in SLC (a-c) and Beijing (d-f). The yellow boxes show the domain that CDV was added.

261 to -16.00^{\%}. The CDV inclusion slightly improved the correlation coefficient between the simulation and observations. However, the highest correlation coefficient was only 0.0613 for 262 the case with an emission rate of 0.003 g/m² \cdot s, thereby still lacking significance. Furthermore, 263 the RMSE noticeably increased after the CDV addition, owing to the lower d-excess values; 264 265 and the residual standard deviation increased significantly when the emission rate was above 0.001 g/m² s, indicating that the fluctuation of simulation became higher than observation. 266 Therefore, the CDV emission rate of 0.001 $g/m^2 \cdot s$ was selected to represent the best 267 simulation in Beijing and was therefore used in the subsequent analysis part. 268

The diurnal variation of Δd -excess along with the mean value is shown in **Figure 1(h)** and 269 1(p). The diurnal SLC variation range was lower than that in Beijing, making the variation in 270 271 the simulations without CDV more pronounced. However, these results without CDV impact displayed an opposite trend compared to the observations. The CDV inclusion in the 272 simulations partially reduced this opposite variation by decreasing the d-excess during 273 nighttime and increasing it during daytime. This effect was more prominent in Beijing. The 274 diurnal variation of the observed data was much higher than that of the simulation without 275 CDV. The CDV addition improved dramatically the variation, bringing it closer to the range 276 of the observations. This improvement was attributed to the difference in humidity between 277 day and night. With lower humidity during nighttime, the proportion of the fixed CDV 278 amount would be higher, resulting in lower d-excess values. Overall, these findings indicated 279 that the addition of CDV could significantly enhance the simulation of the diurnal variation 280 281 of vapor d-excess.

282 3.2 Spatial distribution

Figure 2 displays the monthly average distribution of 2-m vapor d-excess in the four experiments, as well as the differences between the three modified experiments and the control experiment. The results indicated that the CDV inclusion led to a significant vapor d-excess reduction within the urban domains. Additionally, owing to moisture diffusion, a



Figure 3. Monthly mean specific humidity (g/kg) caused by the CDV emission and CDV fraction (%) in the total 2m moisture in SLC (a-c) and Beijing (d-f). Monthly mean 10m wind fields are also shown in (a) and (d).



Figure 4. Time series and diurnal variation of CDV fraction (%) in SLC (a,b) and Beijing (c,d). Black lines are calculated from in-situ observation. Orange lines are calculated from the moisture tracer method. Purple lines are calculated from the mixing equation and simulated d-excess.

slight d-excess decrease was observed in the surrounding areas, whereas the other domains
remained unaffected. When the same CDV emission rate was applied, the d-excess decrease
in SLC was approximately 5‰, which was notably lower than the decrease observed in
Beijing. This difference could be attributed to the higher specific humidity in SLC during
January, resulting in a lower CDV proportion in the total moisture content, and consequently,
a weaker impact on vapor d-excess.

293 3.3 CDV fraction

As shown in **Figure 3(a)** and **3(d)**, there was no significant difference in the monthly mean specific humidity caused by CDV between SLC and Beijing. However, owing to the higher specific humidity, the CDV fraction was significantly lower in SLC. The monthly 10-m wind field explained the direction of the CDV diffusion. The higher wind speed in Beijing could

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also cause a greater kinetic fractionation effect on the land surface in this domain.

The time series and diurnal variation of the CDV fraction are presented in **Figure 4**. In SLC, the monthly mean CDV fraction calculated using the moisture tracer method (fCDV1) was 4.9%, i.e., 2% higher than that obtained using the mixing equation method (fCDV2). In Beijing, fCDV1 was estimated to be approximately 12.3%, i.e., lower than fCDV2 (13.8%).

Table 3 provides the correlation coefficients and RMSEs between these CDV fraction values and the estimates derived from the observed data. SLC exhibited significantly higher correlation coefficients and lower RMSEs than those of Beijing, indicating that impact of a fixed emission rate of CDV can be used to explain the variation of d-excess in the observed data of SLC, but fails to simulate the similar pattern in the observation of Beijing.

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 Table 3 Correlation coefficient and RMSE of the fraction of CDV from observation and each method

Site	Method	Correlation coefficient	RMSE
SI C	fCDV1	0.6524	3.8030
SLC	fCDV2	0.5329	2.5706
D	fCDV1	0.1505	9.5450
Beijing	fCDV2	0.0683	11.4817

309 4 Discussion

310 4.1 Relationship between humidity and vapor d-excess

The relationship between humidity and vapor d-excess is shown in Figure 5. The control 311 experiment in the original IsoRSM simulation revealed no significant correlation between 312 specific humidity and vapor d-excess. However, the CDV addition had a stronger impact on 313 the vapor d-excess reduction when humidity was lower. This effect aligns with the observed 314 315 results in SLC, suggesting that the CDV inclusion can improve the relationship between 316 humidity and vapor d-excess at this location. This relationship can be explained by the CDV fraction in the total atmospheric moisture, as demonstrated by the earlier diurnal variation 317 analysis. With a fixed CDV amount, lower humidity from natural sources would result in a 318 higher CDV impact, and this change caused by CDV would be more pronounced when the 319 atmosphere was relatively stable. However, the observations from Beijing showed a different 320 correlation, i.e., a low specific humidity corresponded to higher observed d-excess values. 321 322 This phenomenon indicated the presence of other moisture sources with higher d-excess 323 values contributing to the distinctive correlation between humidity and vapor d-excess in SLC and Beijing. Overall, these findings suggest that the relationship between humidity and 324 vapor d-excess is influenced by the presence of CDV and other moisture sources, 325 highlighting the complexity of the isotopic composition in different environments. 326



Figure 5. Relationship between 2m specific humidity (q, kg/kg) and 2m vapor d-excess (d, %), and the relationship between q and d*q (%·kg/kg) in SLC (a-f) and Beijing(g-i). The color of the dots shows the temperature differences between 2m and 800hPa for SLC and 1000hPa for Beijing, which can be regarded as an indicator for describing atmospheric stability.

To figure out the source of high d-excess moisture in Beijing, an individual experiment 327 which considered the kinetic fractionation effect in the evaporating process on the land 328 surface was made. Due to this experiment was not particularly relevant to the topic of this 329 study, the results were not displayed in this article. Based on the experiment, the simulation 330 in SLC showed a d-excess reduction when humidity was high, whereas in Beijing, the 331 simulation indicated that d-excess was increased when humidity was low. Therefore, it is 332 333 possible that the kinetic fractionation effect on the land surface is one of the factors 334 contributing to the higher d-excess in Beijing (Lee et al., 2009; Kei Yoshimura et al., 2006). 335 The specific mechanisms and processes involved would require further investigation to better understand the observed relationship between humidity and d-excess in Beijing and the 336 337 influence of the kinetic fractionation effect.



4.2 Humidity increase caused by CDV and the CDV fraction 338

Figure 6. Relationship between specific humidity (kg/kg) and CDV fraction (%) in SLC (a-d) and Beijing (e-h). The color of dots indicates the same meaning in Figure 5.

339 As shown in **Figure 1(e)** and **1(m)**, the specific humidity increase caused by the CDV in SLC was not as significant as that in Beijing. Using the moisture tracer method, moisture at 340 these two sites could be divided into local evaporation and remote transportation sources. 341 Figure S2 shows that in SLC, the CDV addition decreased the moisture transported from 342 remote areas. However, this decrease was not observed in the Beijing simulation. Therefore, 343 344 the specific humidity in SLC did not show an obvious increase, which may have influenced 345 the calculation of the CDV fraction using the moisture tracer method.

The findings in the CDV fraction time series were consistent with the relationship between 346 the CDV fraction and humidity, as depicted in Figure 6. All results from the observations 347 348 and simulations demonstrated a negative correlation between specific humidity and the CDV fraction, except for the observational data in Beijing. The discrepancy between the CDV 349 fraction values obtained from observations and simulations indicated that the vapor d-excess 350 351 decrease in SLC could be explained by CDV emissions, whereas the CDV impact alone could not fully account for the d-excess variation in Beijing in winter. 352

To determine the reason for the higher estimated value of the CDV fraction in the moisture 353 tracer method than that in the mixing equation method in SLC, the mixing process of CDV 354 and natural source vapor was theoretically calculated. As the moisture tracer function could 355 directly obtain the specific humidity caused by CDV (here, q_{CDV}), it was possible to 356 calculate the $(q * d)_{mix}'$ using the two end-member mixing equations: 357

 $(q * d)'_{mix} = q_{nat} * d_{nat} + q'_{CDV} * d_{CDV}.$

The $(q * d)_{mix}'$ indicates the q*d mixed by the moisture of natural sources and CDV with 358 the amount obtained from the moisture tracer method. Then the d_{mix}' can be calculated, as 359 360 follows:

$$d'_{mix} = \frac{(q * d)'_{mix}}{q_{nat} + q'_{CDV}}$$





Figure 7. Comparison between d_mix and d_mix', q*d_mix and q*d_mix' in SLC (a,b) and Beijing (c,d). The colorbar showed the same information as Figure 5.

Figure 8. Monthly mean 10m wind speed (m/s), 2m specific humidity (g/kg) and vapor d-excess (‰) anomalies in different wind directions in SLC (a-c) and Beijing (d-f).

Figure 7 shows a comparison of d_{mix} with $(q * d)_{mix}$ from the model simulation and the calculation. The results indicate that the CDV impact on vapor d-excess was slightly underestimated in SLC and overestimated in Beijing, considering the CDV amount in the simulation; and the bias between d_{mix} and d'_{mix} was higher when the boundary condition was more stable in SLC. This phenomenon might have been caused by the different atmospheric conditions at these two sites.

367 4.3 d-excess difference among wind directions

368 Moisture transportation is mainly controlled by wind direction and speed. Therefore, the vapor d-excess difference among the wind directions could reflect different moisture sources. 369 370 Figure 8 shows the monthly anomalies of wind speed, specific humidity, and vapor d-excess 371 for different wind directions. In the case of SLC, the observational data revealed that the 372 easterly wind direction involved vapor d-excess that was about 2‰ lower compared to that of the westerly wind directions. This finding indicated that moisture originating from the east 373 had a greater influence on CDV. The simulations with CDV emissions also reproduced this 374 difference among wind directions, where lower vapor d-excess values were predominantly 375 376 observed with easterly winds. This consistency between the observed and simulated 377 characteristics further supported the notion that CDV emissions play a significant role in the vapor d-excess variation in SLC. 378

The observational data in Beijing revealed a higher vapor d-excess value when the wind 379 was northwesterly and a relatively lower d-excess value when the wind was southerly. 380 Referring to previous research, the northwest of Beijing, which corresponds to the inland area 381 of North China, had a higher vapor d-excess when the isotopic fractionation effect on the land 382 383 surface was considered in the model. This suggests that the higher d-excess with the 384 northwesterly wind could be transported from the inland area. Table 4 shows the correlation coefficients between observed and simulated vapor d-excess for different wind directions, 385 and the linear regression line of the observation and simulation can be seen in Figure S3. 386 When the wind was southerly, the correlation coefficient showed a significant improvement 387 after adding CDV, thereby indicating that during these periods, the CDV impact was likely 388 the dominating factor of the vapor d-excess. In contrast, the correlation coefficient decreased 389 390 when the wind was northeasterly, indicating that the CDV inclusion did not improve the 391 d-excess simulation in this moisture source.

S:40	Wind	Ctrl	CDV	
Site	direction	Ctri	CDV	
	NE	-0.2690	-0.0942	
SI C	SE	-0.3467	0.4791	
SLU	SW	-0.1710	0.0717	
	NW	-0.1806	-0.0641	
	NE	-0.0102	-0.2164	
D	SE	-0.2730	0.2919	
Beijing	SW	-0.1048	0.2770	
	NW	-0.0377	0.0284	

392 Table 4 Correlation coefficient of vapor d-excess in observation and simulation with different wind directions

393

394 5 Summary and Conclusions

In this study, the isotope-enabled regional spectral model (IsoRSM) was used to simulate the combustion-derived vapor (CDV) emissions in urban areas and determine whether anthropogenic emissions influence the isotopic ratio of water in the urban atmosphere. Two separate one-month experiments were conducted: one in Salt Lake City (SLC) in January 2017 and another in Beijing in January 2007. By adding an extra amount of vapor with a fixed isotopic ratio to the $2^{\circ} \times 2^{\circ}$ domains during the evaporation process, the CDV emissions in the urban areas could be roughly simulated.

The simulation results demonstrated that the introduction of CDV emissions significantly 402 reduced the d-excess of water vapor, particularly when the boundary layer was stable. The 403 404 d-excess decrease in Beijing was more pronounced than that in SLC because of higher 405 humidity from natural sources in SLC. The simulation with CDV emissions showed better agreement with the time series of the in-situ observations in SLC than the original model. 406 407 However, in Beijing, the observed vapor d-excess exhibited stronger fluctuations but was occasionally higher than the simulated values, contrary to the expected CDV impact. Despite 408 this discrepancy, the CDV addition improved the simulation by enhancing the diurnal 409 410 variation of d-excess, resulting in a pattern that closely resembled the observations.

The inclusion of a fixed CDV amount in the simulations led to a positive correlation 411 between vapor d-excess and specific humidity, which was consistent with the observations in 412 SLC and inconsistent with the observations in Beijing. The d-excess underestimation at lower 413 humidity levels in Beijing might be caused by the kinetic fractionation effect during the 414 415 evaporation process on the land surface in the inland areas. The moisture with higher 416 d-excess could be transported by the strong winter monsoon in Beijing. This phenomenon indicated that the CDV should not be the only controlling factor of the variation of vapor 417 d-excess in some domains. 418

419 The moisture tracer method and mixing equation were both employed to estimate the CDV 420 fraction in the total 2-m moisture. The moisture tracer method yielded a monthly mean CDV fraction (fCDV1) of 3.3% in SLC and 12.3% in Beijing. The CDV fraction derived from the 421 422 d-excess value and mixing equation (fCDV2) exhibited a similar trend to that of fCDV1, 423 albeit slightly underestimated in SLC and overestimated in Beijing. Both fCDV1 and fCDV2 were negatively correlated with specific humidity, in contrast to the CDV fraction derived 424 from observations in Beijing. These findings suggest that a fixed amount of CDV emissions 425 alone cannot fully account for the vapor d-excess variations observed in Beijing. 426

Dividing the observed and simulated data based on different wind directions allowed the analysis of moisture transportation effects. The vapor d-excess diversity was enhanced with the CDV inclusion and was more similar to those of the observations at both sites. In Beijing, higher d-excess values were observed under northwesterly winds, suggesting the possible transport from inland areas with a stronger kinetic fractionation effect. Conversely, weaker easterly winds resulted in lower d-excess values, indicating a higher impact of CDV with these sources of moisture.

In conclusion, although there were some biases caused by the underestimation of d-excess variation from natural sources, CDV could be one of the controlling factors of atmospheric vapor d-excess in some urban areas such as SLC. For these regions, the inclusion of CDV in the model would significantly improve the bias, temporal and diurnal variations, and enhance the correlation between vapor d-excess and local humidity. Therefore, it is of great potential to use atmospheric models to simulate anthropogenic vapor emissions.

Several aspects of this study can be improved. One important improvement would be the development of a CDV emission map based on the amount of emitted CO₂. This approach should provide a more realistic representation of the CDV emission sources in urban areas. By incorporating such an emission map into the model simulation, the results should become more reliable and accurate, leading to a better understanding of the CDV influence on the isotopic ratio of water vapor in the urban atmosphere. This improvement should enhance the credibility and applicability of the research findings.

447

448 Data Available Statement

The hourly observed vapor isotope data in Salt Lake City are available at 449 https://osf.io/k47ft (Fiorella et al., 2019). And the hourly observed vapor isotope data in 450 Beijing are available at https://vapor-isotope.yale.edu/sites-and-dataset/asia/cn-beijing (Wen 451 452 et al., 2010). ERA5 reanalysis data are available at 453 https://cds.climate.copernicus.eu/cdsapp#!/search?type=dataset%26text=ERA5 (Hersbach et 454 al., 2020). The data analysis and figure construction in this study were performed using the
455 open-source Python language v3.11.4 (<u>https://www.python.org/</u>) and the Grid Analysis and
456 Display System v2 software (<u>http://cola.gmu.edu/grads/</u>).

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