Jet Stream-surface tracer relationships: Mechanism and sensitivity to source region

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Abstract

The upper-level jet stream impacts surface-level trace gas variability, yet the cause of this relationship remains unclear. We investigate the mechanism(s) responsible for the relationship using idealized tracers with different source regions within a chemical transport model. All tracers' daily variabilities are correlated with the meridional position of the jet in the midlatitudes, but tracers emitted south (north) of the jet increase (decrease) in the mid-latitudes when the jet is shifted poleward. The jet stream regulates the near-surface meridional wind, and this coupling together with the meridional tracer gradient robustly predicts where the jet stream and tracers are in and out of phase. Our study elucidates a major driver of trace gas variability and links it to the location of the jet stream and emissions. These results are useful for understanding changes in trace gas variability if the jet stream's position or major emission source regions change in the future.

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Key Points:

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10	٠	The daily variability of near-surface tracer mixing ratios in the mid-latitudes is
11		correlated with the latitude of the jet stream
12	•	The sign of the jet-tracer relationships depends on the tracer source region and

- the resulting meridional tracer gradients
- The meridional movement of the jet stream alters the near-surface meridional flow, which changes tracer mixing ratios

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16 Abstract

The upper-level jet stream impacts surface-level trace gas variability, yet the cause of 17 this relationship remains unclear. We investigate the mechanism(s) responsible for the 18 relationship using idealized tracers with different source regions within a chemical trans-19 port model. All tracers' daily variabilities are correlated with the meridional position 20 of the jet stream in the mid-latitudes, but tracers emitted south (north) of the jet in-21 crease (decrease) in the mid-latitudes when the jet is shifted poleward. The jet stream 22 regulates the near-surface meridional wind, and this coupling together with the merid-23 ional tracer gradient robustly predicts where the jet stream and tracers are in and out 24 of phase. Our study elucidates a major driver of trace gas variability and links it to the 25 location of the jet stream and emissions. These results are useful for understanding changes 26 in trace gas variability if the jet stream's position or major emission source regions change 27 in the future. 28

²⁹ Plain Language Summary

Previous studies have shown a connection between greenhouse gases or air pollu-30 tants and the jet stream, a narrow band of strong winds aloft that encircle the mid-latitudes. 31 The mechanisms that link the jet stream to changes in greenhouse gases and air pollu-32 tants at earth's surface and how they are connected to the source regions of emissions 33 are not well understood. To address this, we use computer models of the atmosphere that 34 include "tracers," artificial particles that track fluid motion within the atmosphere. Trac-35 ers are emitted from different latitudes in the Northern Hemisphere, ranging from the 36 equator to the pole. All tracers are impacted by the position of the jet stream, but whether 37 a particular tracer increases or decreases when the jet is in a poleward position is a strong 38 function of where it was emitted. We show that the jet stream affects variations in the 39 north-south wind at the surface, and changes in this wind lead to the advection of air 40 with higher or lower tracer concentrations, depending on the latitudinal tracer gradient. 41 Our findings may help interpret other atmospheric models that simulate pollution and 42 greenhouse gases and the impacts of climate change on these species. 43

44 1 Introduction and Motivation

⁴⁵ Concentrations of near-surface air pollutants and greenhouse gases exhibit large
⁴⁶ day-to-day variations, driven by a combination of variations in emissions, chemistry, and
⁴⁷ transport. Understanding the cause of this variability is paramount for interpreting mea⁴⁸ surements and trends in pollutants (e.g., Cooper et al., 2014; Dawson et al., 2014; Kerr
⁴⁹ et al., 2019) and greenhouse gases (e.g., Keppel-Aleks et al., 2011; Miller et al., 2013, 2015;
⁵⁰ Randazzo et al., 2020).

Several studies have highlighted the importance of transport in explaining the daily 51 variability of near-surface composition. For example, daily variations of ozone (O_3) have 52 been linked to transport-related phenomena such as horizontal and vertical advection 53 and frontal systems (Jacob et al., 1993; Kerr et al., 2019; Porter & Heald, 2019; Kerr et 54 al., 2020), while Keppel-Aleks et al. (2011) and Torres et al. (2019) have shown that the 55 variability of carbon dioxide (CO₂) attributed to the prevailing synoptic- and mesoscale 56 weather is of similar magnitude to the variability from local diurnal fluxes. Moreover, 57 variations in the meridional, or north-south, position of the upper-level jet stream and 58 its effect on transient atmospheric eddies and frontal zones have been linked to variabil-59 ity in near-surface particulate matter (Ordóñez et al., 2019), CO_2 (Randazzo et al., 2020; 60 Pal et al., 2020), methane (Guha et al., 2018), and O_3 (Barnes & Fiore, 2013; Shen et 61 al., 2015; Kerr et al., 2020). 62

⁶³ A recent study by Kerr et al. (2020) provided further support for a link between ⁶⁴ variability in the upper-level jet and surface-level O_3 but also showed substantial spatial variations in the relationship. They showed that the daily variability in surface-level O₃ during boreal summer (JJA) is significantly correlated with the meridional position of the jet across the Northern Hemisphere mid-latitudes, but the sign of the relationship differed between land and ocean (with O₃ increasing over land but decreasing over the oceans when the jet is in a poleward position). Furthermore, the jet-O₃ relationship is weak or non-existent at high and low latitudes.

The findings from the aforementioned studies raise several important questions: What 71 mechanisms connect flow aloft to near-surface composition and variability? Why does 72 73 the jet- O_3 relationship vary with latitude and between land and ocean? How do species' lifetimes and source regions affect the relationship? The last question is important when 74 considering the jet's role in the variability of greenhouse gases and surface-level partic-75 ular matter whose lifetimes and source regions differ. Increases in anthropogenic green-76 house gas emissions will likely shift the mean jet latitude poleward and modulate jet speed 77 later in the twenty-first century (Barnes & Polvani, 2013). These projected changes war-78 rant an improved understanding of how flow aloft impacts near-surface composition, which 79 could improve our projections of how future pollutant distributions could change. 80

We address these questions by performing chemical transport model (CTM) sim-81 ulations of a suite of idealized tracers with differing source regions. The simulations en-82 able us to examine how the Northern Hemisphere tracer-jet relationships vary with source 83 region and under what condition(s) there are land-ocean or seasonal variations. Ideal-84 ized tracers can aid in understanding and interpreting the impact of the jet stream on 85 near-surface composition while avoiding the complex interplay of non-linear gas- and particle-86 phase chemistry and temporally- and spatially-varying precursor emissions (e.g. Orbe 87 et al., 2016). 88

In Section 2, we describe the CTM simulations, reanalysis, and methodology used in this study. We document the relationship of the tracers with the jet in Section 3.1 and the impact of the jet on near-surface meridional wind in Section 3.2. We find simple balances that relate the connection of the jet stream with near-surface meridional wind to the meridional tracer gradient give a satisfying physical explanation to differences in the sign of the tracer-jet relationships (Sections 3.2-4).

95 **2** Data and Methodology

We use the GEOS-Chem CTM (version 12.0.2) to perform our tracer simulations 96 (Bey et al., 2001; The International GEOS-Chem User Community, 2018, October 10). 97 GEOS-Chem is driven by assimilated meteorology from the Modern Era-Retrospective 98 Analysis for Research and Analysis, Version 2 (MERRA-2). Three-dimensional MERRA-99 2 fields are input to the CTM every three hours, while surface quantities and mixing depths 100 are provided every hour. Specifically, our configuration of GEOS-Chem follows a pas-101 sive simulation described in Liu et al. (2001). We perform this simulation at a resolu-102 tion of 2° latitude x 2.5° longitude with 72 vertical levels (~ 15 hPa spacing below 800 103 hPa) for 2007 - 2010, and we discard the first year (2007) for spin up. 104

Previous studies have demonstrated the accuracy of transport in GEOS-Chem and 105 the assimilated meteorological product, MERRA-2, driving the CTM. Bosilovich et al. 106 (2015) showed that magnitude of MERRA-2 zonal and meridional wind fields as well as 107 the location of wind maxima are well-constrained by observations and other reanalyses. 108 GEOS-Chem yields realistic mixing ratios and seasonal and latitudinal variations of other 109 tracers such as lead and beryllium with no significant global bias (Liu et al., 2001). How-110 ever, Yu et al. (2018) recently pointed out that the use of offline CTMs, such as GEOS-111 Chem, together with an archived assimilated meteorological product can lead to verti-112 cal transport errors due, in part, to loss of transient advection (resolved convection). While 113 potential biases and errors are important to keep in mind, the extensive body of liter-114



Figure 1. (a) Zonally-averaged tracer mixing ratios in JJA. (b) JJA-averaged mixing ratios of (b) χ_{70-80} , (c) χ_{40-50} , and (d) χ_{10-20} . Scatter points and vertical bars in (b)-(d) represent the mean position and variability of the jet stream in JJA, respectively. Note that the thicker lines in (a) correspond to the tracers featured in (b)-(d).

ature on the reliability of GEOS-Chem supports its suitability as the framework to ad dress our research questions.

Within GEOS-Chem, we implement a suite of nine passive tracers that differs only 117 in their source regions, which are prescribed as constant flux boundary conditions (i.e., 118 emissions) in zonally-symmetric 10° latitudinal bands. Tracers are herein denoted $\chi_{\phi_1-\phi_2}$, 119 where ϕ_1 is the latitude corresponding to the southern boundary of the source region and 120 ϕ_2 is the northern boundary. All tracers decay uniformly at a loss rate of $\tau = 50 \text{ days}^{-1}$. 121 Tracers with the same loss have been used in prior studies (e.g., Shindell et al., 2008; Orbe 122 et al., 2017, 2018; Yang et al., 2019). Although not the primary focus of our analysis, 123 we also explore how the lifetime of tracers impacts their relationship with the jet by sim-124 ulating χ_{40-50} with loss rates of $\tau = 5, 25, 100$ and 150 days⁻¹. Unless indicated, all 125 analyses use daily mean near-surface (1000 - 800 hPa) tracer mixing ratios. 126

In addition to driving the GEOS-Chem simulations, we use MERRA-2 to charac-127 terize the meteorology responsible for tracer variability (McCarty et al., 2016; Gelaro 128 et al., 2017). MERRA-2 is output on a global $0.5^{\circ} \ge 0.625^{\circ}$ grid with 72 vertical levels. 129 Specifically, we obtain 3-hourly 1000-800 hPa meridional wind (V) and 500 hPa zonal 130 wind (U) from MERRA-2 and average these data to daily mean values, consistent with 131 our treatment of tracers from GEOS-Chem. The horizontal resolution differs between 132 GEOS-Chem and MERRA-2, and we degrade the resolution of MERRA-2 to match that 133 of GEOS-Chem using xESMF, a universal regridder for geospatial data (Zhuang et al., 134 2020).135

We locate the latitudinal position of the jet stream (ϕ_{jet}) daily at each longitude by finding the latitude (restricted to 20-70°N) of maximum 500 hPa U. A simple convolutionbased smoothing is applied in longitudinal space to address potential longitudinal discontinuities in the jet's position (i.e., "jumps" in ϕ_{jet}) using a box-shaped function with a width of ~ 10° longitude. Identifying ϕ_{jet} using 500 hPa winds follows previous work by Barnes and Fiore (2013) and Kerr et al. (2020).

The temporal correlation between ϕ_{jet} and near-surface tracer mixing ratios or V 142 is quantified with the Pearson product-moment correlation coefficient, indicated by r(X, Y), 143 where X and Y are the time series of interest. We assess the significance of the corre-144 lation coefficient using the non-parametric moving block bootstrapping method, which 145 preserves much of the temporal correlation in the time series and makes no *a priori* as-146 sumptions about the time series' distributions. In essence, time series X and Y are ran-147 domly reordered by sampling continuous blocks of data with length = 10 days, and r(X, Y)148 is thereafter recalculated. We conduct 10000 realizations of this reordering, and signif-149 icance is determined with a two-tailed percentile confidence interval method at the 0.05 150 significance level (Wilks, 1997; Mudelsee, 2003; Wilks, 2011). 151

¹⁵² We also generate composites of tracer mixing ratios and V on days when the jet ¹⁵³ stream is poleward (PW) and equatorward (EW). The PW (EW) composite is defined ¹⁵⁴ locally (i.e., at each longitude) as the average value of the field of interest for days where ¹⁵⁵ ϕ_{jet} exceeds (is less than) the 70th (30th) percentile. We define a "positive" relation-¹⁵⁶ ship to mean that the PW (EW) movement of the jet is associated with increased (de-¹⁵⁷ creased) mixing ratios or V. The opposite is true for a "negative" tracer-jet relationship.

158 **3 Results**

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3.1 Relationship between the jet stream and tracers

Before we examine the tracers' relationship with ϕ_{jet} we briefly discuss the mean tracer distributions and their daily variability. Zonally-averaged tracer mixing ratios peak within their source regions and diminish to roughly half of their maximum value $\pm 5^{\circ}$ outside their source regions (Figure 1a). Tracers with source regions at latitudes (ϕ) north of 60°N have higher mixing ratios within their source regions compared with tracers emitted at lower latitudes (Figure 1a), supporting an isolated Arctic lower troposphere and the "polar dome" as a barrier to transport (Law & Stohl, 2007).

¹⁶⁷ Despite zonally-symmetric emissions, there are zonal variations in tracer mixing ¹⁶⁸ ratios (Figure 1b-d). The latitudinal range with high tracer mixing ratios (> 0.8 ppm) ¹⁶⁹ is larger over the ocean basins for tracers with high and mid-latitude sources (e.g., χ_{70-80} , ¹⁷⁰ χ_{40-50} ; Figure 1b-c). These ocean regions coincide with the Atlantic and Pacific storm ¹⁷¹ tracks. High mixing ratios of tracers with source regions in the tropics (e.g., χ_{10-20}) are ¹⁷² more diffuse over land and more restricted over the tropical ocean (Figure 1d).

173 Spatial variations in the tracers' daily variability (as measured by the standard de-174 viation) are similar to spatial variations in their mean distribution, with highest vari-175 ability near the tracer source region and decreasing to the north and south (not shown). 176 Furthermore, the ratio of each tracer's standard deviation to its mean is $\sim 50\%$ near 177 the source region and diminishes to $\sim 20\%$ well outside the source region (not shown).

To assess the impact of the meridional movement of the jet on daily tracer vari-178 ability, we examine composites of tracer mixing ratios when the jet is PW and EW (see 179 Section 2). As is shown in Figure 2, there is a significant tracer-jet relationship for all 180 tracers during JJA and boreal winter (DJF) within the mid-latitudinal range over which 181 the jet traverses. However, the sign of the relationship hinges on the meridional gradi-182 ents of the tracers $(\partial \chi / \partial \phi)$. Tracers with source regions at low latitudes ($\phi < 40^{\circ}$ N) 183 have a negative gradient $(\partial \chi / \partial \phi < 0)$ within the latitudinal range of the jet and in-184 crease in the mid-latitudes when the jet is PW (Figure 2a-b). Tracers emitted around 185 the latitude of the jet $(40^{\circ} < \phi < 60^{\circ} \text{N})$ have a spatially-varied gradient and relation-186 ship with the jet in the mid-latitudes. In particular, we note the land-ocean differences 187 in the JJA χ_{40-50} -jet relationship (Figure 2c). Tracers with source regions at high lat-188



correlations that are not statistically significant. Scatter points and vertical bars represent the mean position and variability of the jet stream in JJA, respectively. The difference in composites of JJA (a) χ_{70-80} , (c) χ_{40-50} , and (e) χ_{10-20} for days with a PW versus EW jet stream. Hatching denotes tracer-jet (b), (d), and (f) are the same as (a), (c), and (f) but for DJF. Figure 2.

itudes ($\phi > 60^{\circ}$ N) are characterized by $\partial \chi / \partial \phi > 0$ in the mid-latitudes and decrease in the mid-latitudes when the jet is PW (Figure 2a-b).

Beyond the mid-latitudes and these three tracers, impact of source region on the 191 tracer-jet relationships for all the GEOS-Chem tracers can be easily seen in the zonal 192 mean (Figure 3a-b). The tracer-jet relationships all exhibit an oscillatory pattern, but 193 tracers with source regions south of the range of the jet are positively correlated with 194 the jet in the mid-latitudes and are flanked by negative correlations (although generally 195 not significant) outside the mid-latitudes. Tracers with source regions north of the jet 196 197 have a negative correlation with the jet in the mid-latitudes and a positive, but non-statistically significant, correlation outside the mid-latitudes (Figure 3a-b). 198

The variations in tracer mixing ratios related to the meridional oscillations of the jet are a sizable fraction of the overall daily tracer variability discussed earlier in this section. For example, the ratios of the jet-associated variations in χ_{10-20} , χ_{40-50} , and χ_{70-80} to the overall variability (standard deviation) zonally-averaged over the mid-latitudes ($40^{\circ} < \phi < 60^{\circ}$ N) are 58%, 35%, and 47%, respectively.

In a gross sense, the relationship between the jet stream and our tracers does not change in DJF compared to JJA, but further inspection indicates that there are nuanced differences in the relationships (Figure 2). For example, the change in mid-latitude mixing ratios of χ_{40-50} due to the meridional movement of the jet is varied in sign and strength during JJA, while the DJF change is largely negative (Figure 3b-c).

The jet is an important source of variability for near-surface trace species spanning 209 a range of lifetimes. The relationship of the jet with χ_{40-50} for loss rates spanning 5 to 210 150 days⁻¹ is similar in sign and significance to χ_{40-50} with the 50 days⁻¹ loss rate dis-211 cussed elsewhere in this study, although the precise magnitude of the variability asso-212 ciated with the jet changes with tracer lifetime (Figure S3). We note that, although these 213 findings hold for tracers with zonally-symmetric emissions, tracers with more realistic 214 emissions (e.g., land-ocean contrasts, urban-rural differences) may have a more compli-215 cated relationship with the jet. With that said, the results presented here indicate a strong 216 relationship between trace gas variability and the jet absent these other confounding fac-217 tors. 218

3.2 Mechanisms

The analysis presented in Section 3.1 has shown that a large fraction of daily tracer variability is related to meridional movement of the jet but does not show the mechanism(s) involved or why the signs of the tracer-jet relationships varies. Kerr et al. (2020) suggested that the jet stream affects surface-level O_3 by altering the near-surface meridional flow (V). We test this hypothesis using our suite of tracers. We first examine the V-jet relationship and then how this impacts the tracers.

Figure 3c indicates that southerly flow increases in the mid-latitudes (around the 226 latitudinal range of the jet stream) when the jet is PW during JJA and DJF; however, 227 it does not show the magnitude. As is shown in Figure 4a-b, V increases over 5 m/s in 228 parts of the mid-latitudes when the jet is PW. This stands in sharp contrast to time-averaged 229 V, which is generally weak (-2 < V < 2 m/s) over the vast majority of the mid-latitudes. 230 It is exceedingly rare for time-averaged V to have the same magnitude changes in V linked 231 to the jet (contours in Figure 3a-b). Outside the mid-latitudes, the relationship between 232 V and ϕ_{jet} is largely non-significant and weak (Figures 3c, 4a-b). 233

The V-jet relationship is not zonally-symmetric (Figure 4a-b). For example, the JJA V-jet relationship is negative over the mid-latitude oceans on the windward shores of the continents but is positive over the mid-latitude continents and the leeward shores (Figure 4a). The spatial extent of regions with a positive V-jet relationship increases in



Figure 3. An illustration of how ϕ_{jet} impacts near-surface V and tracers. (a) The JJA zonally-averaged correlation between ϕ_{jet} and individual tracers (colors) and the mean position and and variability of the jet stream (scatter point and horizontal bars). (b) same as (a) but for DJF. (c) Zonally-averaged $r(V, \phi_{jet})$. Dashed vertical lines in (a)-(b) denote the latitudes where $r(V, \phi_{jet}) = 0$ for each season. Dashed horizontal lines separate positive from negative correlations.

DJF compared to JJA, and the jet has a significant positive relationship with near-surface V over a majority of the Pacific and Atlantic Ocean basins in DJF (Figure 4a-b).

In the zonal mean, the latitudes, or nodes, where $r(\chi, \phi_{jet}) = 0$ are well-aligned with the latitudes where the jet stream and V are not correlated (Figure 3). This result is especially clear for tracers with northern source regions, while tracers with source regions in the mid-latitudes (e.g., $\chi_{30-40}, \chi_{40-50}$) are slightly offset from the latitudes where $r(V, \phi_{jet}) = 0$. The only node where $r(V, \phi_{jet}) = 0$ does not coincide with $r(\chi, \phi_{jet}) =$ 0 occurs during DJF north of the jet (Figure 3b). In this case, the latitude where $r(V, \phi_{jet}) =$ 0 lies north of $r(\chi, \phi_{jet}) = 0$ by ~ 5°, and other processes such as changes in zonal winds or convection could be important for the tracer-jet relationships in this region and season. These results support Kerr et al. (2020) and provide strong evidence linking the tracerjet relationships to (1) the source region of the tracers and (2) the V-jet relationship (Figure 3).

The jet-induced change in V modifies meridional tracer advection (i.e., $-V \cdot \partial \chi / \partial \phi$). Thus, the impact of a given change in V is expected to depend on the local tracer gradients. If $\partial \chi / \partial \phi$ is weak, then smaller tracer changes are expected compared with locations with stronger $\partial \chi / \partial \phi$. It also follows that the same change in V operating over $\partial \chi / \partial \phi < 0$ versus $\partial \chi / \partial \phi > 0$ would result in changes of tracer mixing ratios with different signs. Given this, we postulate that the expected sign of the tracer-jet relationships $(\text{sgn}[r(\chi, \phi_{jet})])$ shown in Figures 2-3 can be approximated by:

$$\operatorname{sgn}[r(\chi,\phi_{jet})] \sim \operatorname{sgn}(-r(V,\phi_{jet}) \cdot \frac{\partial \chi}{\partial \phi}).$$
(1)

In practice, this balance implies that the anomalous southerly flow in the mid-latitudes that accompanies a PW-shifted jet $(r(V, \phi_{jet}) > 0)$ will advect higher tracer mixing ratios from lower latitudes if $\partial \chi / \partial \phi < 0$, yielding a positive expected tracer-jet relationship (i.e., $\operatorname{sgn}[r(\chi, \phi_{jet})] > 0$).

The simple balance in Equation 1 robustly captures the large-scale differences in the sign of the relationship between the jet and all tracers. We illustrate this for χ_{40-50} in Figure 4c-d. The application of Equation 1 can explain the widespread negative χ_{40-50} jet relationship in mid-latitudes during DJF (Figure 4d) but also the differences in sign on much smaller spatial scales during JJA (Figure 4c). Moreover, we note that Equation 1 captures the land-ocean contrasts present in the JJA χ_{40-50} -jet relationship (Figure 4c).

The application of Equation 1 does not capture the sign of the χ_{40-50} -jet relationship in the vicinity of the Atlantic and Pacific storm tracks (Figure 4c-d), and this is the case for other tracers as well (not shown). Since our tracer mixing ratios are roughly zonallysymmetric (Figure 1b-d), the effect of changes in U are negligible to first order. However, the jet stream exerts an influence on near-surface U (Woollings et al., 2010), especially near the exit region of the these storm tracks. To account for this, future studies could consider the impact of both the V-jet and U-jet relationships.

The zonal variations in the tracer-jet relationships previously discussed could stem 277 from zonal variations in the response of V to the movement of the jet or zonal variations 278 in the tracer gradients. To explore this, we have isolated the terms in Equation 1 by sep-279 arately fixing each to its zonal mean value and thereafter recalculating $\operatorname{sgn}[r(\chi, \phi_{jet})]$ to 280 gauge which exerts a stronger influence on the tracer-jet relationships (not shown). Re-281 calculating Equation 1 with $\partial \chi / \partial \phi$ fixed to its zonal mean value and $r(V, \phi_{iet})$ varying 282 as in Figure 4a-b yields expected tracer-jet relationships with zonal variations that re-283 semble the relationships shown in Figure 4c-d. This sensitivity test together with the anal-284 ysis performed in Figure 4c-d confirm spatiotemporal variations in the V-jet relation-285 ship are the most important factor in explaining the tracer-jet coupling, followed by the 286 latitudinal tracer gradient. 287

The importance of the jet stream and meridional flow on daily tracer variability is not restricted to only near-surface mixing ratios but holds for tropospheric column abundances. To support this, we repeat the analyses shown in Figures 3-4 but with V and mass-weighted tracer mixing ratios from 1000-200 hPa (Figures S1-S2) to show that the V-jet relationship not only explains variations in near-surface mixing ratios but also in tropospheric column tracer mixing ratios.



Figure 4. (a-b) Differences in composites of V for days with a PW versus EW jet stream (colors). Time-averaged V is illustrated for 5 m/s (solid black contour) and -5 m/s (dashed black contour). Hatching denotes statistically non-significant V-jet correlations. (c-d) The correlation coefficient calculated between χ_{40-50} and ϕ_{jet} (colors). As denoted in the legend beneath (c), stippling and hatching show the expected sign of the correlation, $E[r(\chi_{40-50}, \phi_{jet})]$, determined using Equation 1. Scatter points and vertical bars in all subplots represent the mean position of and variability of the jet stream, respectively.

²⁹⁴ 4 Conclusions

This study employs idealized loss tracers within a chemical transport model to show 295 that the daily variability of the position of the jet stream has a strong influence on near-296 surface tracer mixing ratios within the seasonally-dependent latitude range of the jet but 297 a weak relationship outside this range. The sign of the jet-tracer relationship varies with 298 the latitude of tracer source and the resulting meridional tracer gradients (Figures 2, 3a-299 b). Tracers with a negative gradient within the latitudinal range of the jet have posi-300 tive tracer-jet relationships in the mid-latitudes, while the opposite is true for tracers 301 with positive gradients within the jet's range. Tracers whose source regions lie within 302 the latitudinal range of the jet have a zonally-varying meridional gradient and subsequently 303 a zonally-asymmetric relationship with the jet in the mid-latitudes. Strong jet-tracer re-304 lationships are found for both JJA and DJF, but the latitudes with the strongest rela-305 tionships vary with the seasonal movement of the mean jet latitude. 306

Our results help to shed light on whether the variability of an observed near-surface trace species is due to transport. If an observational site does not lie within the seasonal range of the jet, jet-driven variability is likely not a major factor. However, if the observational site is within the seasonally-dependent range of the jet, it is likely that there will be transport-driven variability with the sign of the jet-trace species relationship dependent on the sign of the meridional background gradient and the local V-jet relationship.

We show that the mechanism that connects the upper-level jet to variability near-314 surface composition is changes in near-surface meridional flow that result from the merid-315 ional movement of the jet stream. This mechanism explains (1) the variation in sign of 316 the jet-tracer relationship with tracer meridional gradients, (2) the land-ocean differences 317 in the jet-tracer relationship for tracers with mid-latitude sources, and (3) seasonal dif-318 ferences in the jet-tracer relationship. Furthermore, this mechanism explains both the 319 latitudinal and land-ocean differences in the JJA jet-O₃ relationship reported in Kerr 320 et al. (2020) and also helps explain seasonality in the jet-O₃ relationship. Although not 321

shown in Kerr et al. (2020), the sign of the jet-O₃ relationship over North America and Eurasia changes from positive during JJA to negative in DJF, which is broadly consistent with χ_{40-50} (Figures 2c-d, 4c-d).

The jet-tracer relationships found in our simulations hold for a range of tracer life-325 times (5 to 150 days; Figure S3) and for mass-weighted tropospheric column mixing ra-326 tios. Thus, our results may be useful for interpreting variations in a host of species, in-327 cluding the total column measurements commonplace among satellite products. Con-328 temporaneous studies have found that variations in meteorology can explain a substan-329 330 tial portion of total column observations of greenhouse gases, comparable to the impact of regional variations in surface fluxes (e.g., Keppel-Aleks et al., 2011). Differentiating 331 whether patterns in satellite observations are due to transport versus variations in sur-332 face fluxes may help explain differences in trace gas distributions due to large-scale trans-333 port. Future studies should test this possibility. 334

Our study has documented a major driver of near-surface composition variability 335 (i.e., transport associated with the jet stream) and linked this driver with the location 336 of emissions. This finding is relevant for understanding possible future changes of tracer 337 variability, as models predict that the jet stream will migrate north (e.g., Barnes & Polvani, 338 2013), which will modify the poleward transport of air pollution and greenhouse gases 339 via its regulation of the near-surface meridional flow. Recently there has been a redis-340 tribution of anthropogenic emissions from the mid-latitudes (developed nations) to low 341 latitudes (developing nations) (Zhang et al., 2016), which may change meridional tracer 342 gradients and the daily variations connected to the jet. Further research is needed to quan-343 tify (1) how seasonal variations and non-uniform chemical loss of tracers affect their re-344 lationship with the jet and (2) the impact of changes in the the position of the jet and 345 the source region of emissions on the variability of near-surface trace species. 346

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Jet Stream-surface tracer relationships: Mechanism and sensitivity to source region

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Contents of this file

1. Figures S1 to S3

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Figure S1. Same as Figure 3 in the main text but calculated with mass-weighted 1000 - 200 hPa mixing ratios for χ and 1000 - 200 hPa V.

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Figure S2. Same as Figure 4 in the main text but calculated with mass-weighted 1000 - 200 hPa mixing ratios for χ_{40-50} and 1000 - 200 hPa V.

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Figure S3. Same as Figure 2 in the main text but for χ_{40-50} with loss rates of (a, e) 5 days⁻¹; (b, f) 25 days⁻¹; (c, g) 100 days⁻¹; and (d, h) 150 days⁻¹. Panels (a-d) are for JJA and (e-h) for DJF.

(a) JJA $\chi_{5 d^{-1}; 40-50}$ (PW - EW)

(b) JJA $\chi_{25 d^{-1}; 40-50}$ (PW - EW)