Estimating the average time for inter-continental transport of air pollutants

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[1] We estimate the average time required for inter-continental transport of atmospheric tracers based on simulations with the global chemical tracer model MOZART-2 driven with NCEP meteorology. We represent the average transport time by a ratio of the concentration of two tracers with different lifetimes. We find that average transport times increase with tracer lifetimes. With tracers of 1- and 2-week lifetimes the average transport time from East Asia (EA) to the surface of western North America (NA) in April is 2–3 weeks, approximately a half week longer than transport from NA to western Europe (EU) and from EU to EA. We develop an 'equivalent circulation' method to estimate a timescale which has little dependence on tracer lifetimes and obtain similar results to those obtained with short-lived tracers. Our findings show that average inter-continental transport times, even for tracers with short lifetimes, are on average 1–2 weeks longer than rapid transport observed in plumes. **Citation:** Liu, J., and D. L. Mauzerall (2005), Estimating the average time for inter-continental transport of air pollutants, *Geophys. Res. Lett.*, *32*, L11814, doi:10.1029/2005GL022619.

1. Introduction

[2] Policy makers are increasingly interested in the impact that emissions from other continents have on air quality in the United States. Rapid episodic inter-continental transport under exceptional meteorological conditions of chemical constituents and aerosols have been reported with transport times of between 1 and 10 days [*Bailey et al.*, 2000; *Husar et al.*, 2001; *Jaffe et al.*, 1999; *Stohl et al.*, 2003]. Methods used to estimate episodic transport times include: (1) evaluation of satellite observations [*Husar et al.*, 2001]; (2) isentropic back-trajectory models [*Jaffe et al.*, 2003]; (3) transit-time probability density functions (pdf) [*Holzer et al.*, 2003]; (4) passive tracers with fixed lifetimes [*Stohl et al.*, 2002]; (5) ratios of concentrations of hydrocarbons with different lifetimes in a plume (allowing for dilution) to derive the plume age [*Mauzerall et al.*, 1998; *McKeen and Liu*, 1993; *McKeen et al.*, 1996].

[3] Relatively little research has focused on deriving the average time for inter-continental transport of background levels of air pollution despite the fact that significant quantities of pollutants may be transported outside of plumes. We focus on short-lived tracers because short-lived air pollutants (i.e. SO_2, aerosols, NO_x, O_3) with lifetimes less than a few months have more direct health impacts than long-lived air pollutants (i.e. CH_4, CO_2, CFCs). It is therefore important to establish the average inter-continental transport time of short-lived pollutants. In this study we derive a method to estimate the average intercontinental transport times for short-lived tracers. Our mean transport time represents the average age of air parcels, weighted by the concentration of tracer remaining in the parcel when it arrives at a destination D. Here the transport times we calculate are based on averaged concentrations of tracers (including both plumes and background air) from multi-year model simulations and are intended to approximate the general transport properties experienced by air pollutants with first-order decay properties.

2. Methods

[4] We start by constructing a heuristic model in which two chemical tracers *a* and *b*, identical except for different first-order lifetimes τ_a and τ_b, respectively, are continuously emitted in a series of small air parcels from the source region S and pass a destination D. The properties of these parcels are similar to those of *Plumb and McConalogue* [1988] (i.e. small enough to keep their integrity during transport). Assuming all transport properties are the same (i.e. same emission rates, initial conditions, advection and diffusion), the ratio of the tracers' concentrations in any such parcel represents the age of the parcel. Our analysis is distinct from the work of *McKeen et al.* [1996] and *Mauzerall et al.* [1998] who examined plume age where concentrations inside of plumes were elevated relative to local background concentrations. Here we assume continuous uniform emissions of tracers from each continental region and assume that dilution with surrounding air is included with the other physical transport properties. The transport time t for a single parcel to travel from S to D is:

$$t = \frac{\tau_a \tau_b}{\tau_a - \tau_b} \ln\left(\frac{c_a(t)}{c_b(t)}\right) \quad (1)$$

where τ_a and τ_b are tracer lifetimes, and $c_a(t)$ and $c_b(t)$ are tracer concentrations inside the parcel.

[5] If n parcels taking n different pathways meet at D simultaneously with parcel i ($1 \le i \le n$) having been released from S_i (with initial concentration C_i^o), traveled path i over time t_i to reach D, the total concentration of tracer *a* or *b* at D will be the sum of the concentrations in the n parcels. The 'average' transport time based on the ages of the n parcels would be:

$$t = \frac{\tau_a \tau_b}{\tau_a - \tau_b} \ln\left(\frac{\sum_{i=1}^{n} f_i^o \exp(-t_i/\tau_a)}{\sum_{i=1}^{n} f_i^o \exp(-t_i/\tau_b)}\right) \quad (2)$$

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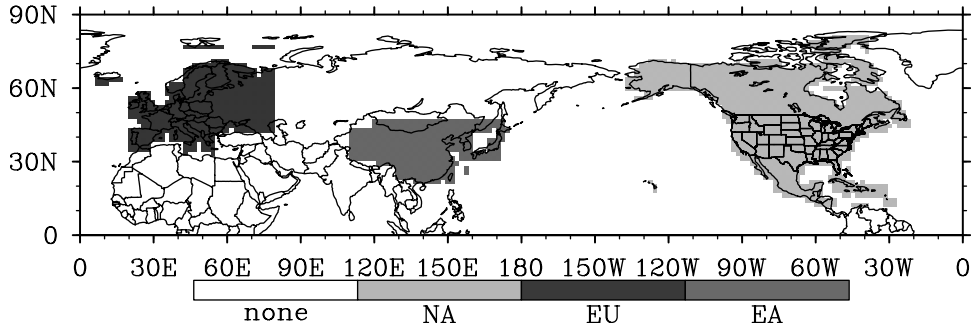

Figure 1. Three tagged continental regions. See color version of this figure in the HTML.

where i indicates a specific air parcel. $f_i^o = C_i^o/(\sum_{i=1}^{n} C_i^o)$ is the initial fraction of each tracer concentration at S in each air parcel. Equation (2) implies that the average transport time calculated by equation (1) would depend both on the meteorological conditions along each path and on the lifetimes of the tracers.

[6] Now consider three limiting situations, namely both tracers' lifetimes are very small, very large or similar. By applying L'Hospital's rule to equation (2), we obtain:

$$t_{\min} = \min(t_1, t_2, \ldots) \quad (\text{when both } \tau_a, \tau_b \to 0) \tag{3a}$$

$$t = \sum_i f_i^D t_i \quad (\text{when } \tau_a \to \tau_b) \tag{3b}$$

$$t_{\max} = \sum_i f_i^o t_i \quad (\text{when both } \tau_a, \tau_b \to \infty) \tag{3c}$$

where $f_i^D = C_i^D/(\sum_{i=1}^{n} C_i^D)$ is the fraction of tracer concentration at D arriving in each air parcel (indicating the age spectrum of parcels at D). Equations (3a)–(3c) indicate that when τ_a and τ_b are very small, the average transport time calculated by equation (1) represents the transport time via the fastest pathway. When tracer lifetimes are similar, the average transport time follows a Green function [*Hall and Plumb*, 1994]. When both τ_a and τ_b are sufficiently large, the transport time reaches its maximum.

[7] To calculate the average time for inter-continental transport, we use the Model of Ozone and Related Tracers, version 2 (MOZART-2) driven with assimilated NCEP meteorology from 1991 to 2001 at a horizontal resolution of approximately 1.9° latitude × 1.9° longitude with 28 sigma vertical levels from the surface to 2.7mb. A description of this version of the model, including initialization and tracer definitions, is available in *Liu et al.* [2005]; a full description can be found in *Horowitz et al.* [2003]. We prescribe uniform emissions of chemical tracers from three continental regions, North America (NA), Europe (EU), and East Asia (EA) (see Figure 1) at 10^{11} molecules $cm^{-2}s^{-1}$and for each region use four chemical tracers with first-order lifetimes of 1, 2, 4 and 8 weeks. We calculate the 11-year monthly mean concentrations for each tracer from the results of the simulation. Within each grid cell we assume that in the 11-year monthly mean tracer concentration distribution chemical loss of the tracer is balanced by transport of the tracer into the grid cell in order to maintain the average tracer distribution. Our approach can approximate the average transport of radon, individual hydrocarbons or CO (by choosing tracers of appropriate lifetimes but without accounting for variability in OH fields), but can not represent transport of pollutants such as O_3 or aerosols (because our model configuration does not include photochemistry or deposition) nor of individual parcels or plumes.

3. Results and Discussion

[8] Using equation (1) and results from MOZART-2 we calculate the average time in April (the month associated with strongest trans-Pacific transport) required to transport the EA tracer to the surface at Cheeka Peak (CPO), WA, USA, the NA tracer to the surface at Paris, France, and the EU tracer to the surface at Beijing, China. Table 1 summarizes the results based on the transport of two shorter-lived (1 and 2 weeks) and two longer-lived (4 and 8 weeks) tracers. We find that the mean 11-year transport time is 2–2.5 weeks when using the two shorter-lived tracers, similar to *Stohl et al.* [2002] and *Holzer et al.* [2003], but almost doubles when using the two longer-lived tracers. Figure 2 shows the relationship between the logarithm of a tracer concentration ln(c) and the reciprocal of that tracer lifetime ($1/\tau$). The slope of the curve in Figure 2 represents the average transport time (see equation 1). However, depend-

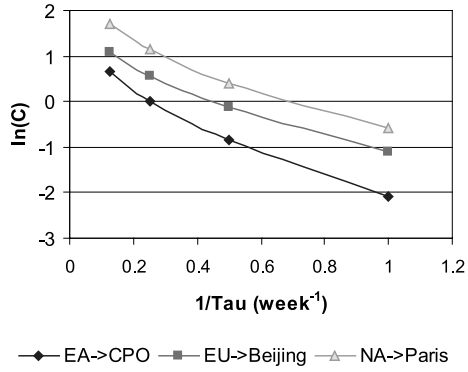

Figure 2. The relationship between the logarithm of the 11-year mean tracer concentrations in April ln(c) at the surface and the reciprocal of tracer lifetimes ($1/\tau$) of (1) the EA tracer at CPO, WA, USA, (2) the EU tracer at Beijing, China, and (3) NA tracer at Paris, France. See color version of this figure in the HTML.

Table 1. 11-Year Average Inter-Continental Transport Times for Two Sets of Tracers in April (Unit: Weeks)

Tracer Lifetime	EA->CPO	EU->Beijing	NA->Paris
1–2 weeks	2.5	2.0	2.0
4–8 weeks	5.1	4.1	4.5

ing on which tracers are used to evaluate the slope, different apparent transport times are obtained. When tracer lifetimes (τ) are short, the slope approaches a constant which is the shortest possible transport time (see equation 3a). As τ increases, the absolute value of the slope, and hence the transport time, increases as well.

[9] In reality, both short and long pathways exist. Tracers with lifetimes significantly longer than average may travel very long (or slow) pathways before intercepting D. This increases the frequency of older parcels arriving at D relative to what occurs with short-lived tracers (see equations 3b and 3c). To account for both short and long pathways and simplify the age spectrum, we introduce a concept of 'equivalent circulation'. We assume that a newly released parcel will first take an average of t^* weeks to travel from S to D. It may then travel beyond D and revisit D on average every T^* weeks. If parcels are continuously emitted, the ages of the parcels at D include t^*, $t^* + T^*$, $t^* + 2T^*$ (where T^* determines the shape of the age spectrum of the tracer at D). The total tracer concentration at D is:

$$C_D = \sum_{i=0}^{\infty} C_0^* \exp\left(-\frac{t^* + iT^*}{\tau}\right) = \frac{C_0^* \exp(-t^*/\tau)}{1 - \exp(-T^*/\tau)} \quad (4)$$

where C_0^* is the initial tracer concentration at S, and i is the number of times the parcel revisits D. Therefore, by using equation (4) to adjust tracer concentrations at D and assuming t^* has little dependence on tracer lifetimes, we obtain both t^* and T^*. Figure 3 indicates that the slope (which represents the short transport time t^*) of $\ln c'$ (where c' is the tracers concentrations adjusted by equation 4) and $1/\tau$ is almost constant when T^* for EA tracer to CPO, EU tracer to Beijing, and NA tracer to Paris is 6, 8 and 7 weeks respectively. These times (t^*) are similar to those obtained using only the shorter-lived tracers (i.e. τ = 1 and 2 weeks) without adjustment. We find that for tracers with lifetimes of 2-weeks or less, at least 95% of tracers are transported from NA to Paris France, from EU to East Asia and from EA to CPO, Washington in t* time.

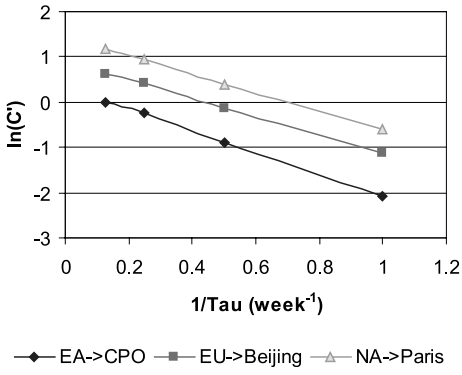

Figure 3. Same as Figure 2, but for adjusted tracer concentrations based on equation 4 where T^* are 6, 8, 7 weeks for EA->CPO, EU->Beijing, and NA->Paris, respectively. See color version of this figure in the HTML.

[10] Figure 4 shows the horizontal distribution of the 11-year average surface transport time in April for NA, EU and EA tracers by two short-lived tracers (τ = 1 and 2 weeks). The time for transport from EA to the surface of western NA is 2–3 weeks, about a half week longer than the transport time from NA to western EU or from EU to EA, and is consistent with the findings of *Stohl et al.* [2002].

[11] Using tracers with 1 and 2 week lifetimes, which reduces the contribution from long pathways, we approximate the cumulative probability distribution of inter-continental transport times in April. We calculate the daily average transport times in April between 1991 and 2001 for transport of EA tracer to Cheeka Peak (CPO), WA, USA, of NA tracer to Paris, France, and of EU tracer to Beijing, China (Figure 5). At CPO, 90% of the tracer coming from EA arrives in 12–18 days at 500 hPa and 13–24 days at the surface (Figure 5a). The most rapid transport occurs in 8 days at 500 hPa and 11 days at the surface. The fastest trans-Atlantic transport of the NA tracer to Paris is 8 days and occurs at both the surface and at 500 hPa (Figure 5b). Trans-Atlantic transport exhibits a larger distribution of transport times at the surface (10–25 days) than that at 500 hPa (10–17 days) indicating that once emissions reach the free troposphere they are relatively rapidly transported across the Atlantic. The age distribution of the EU tracer arriving in Beijing at the surface (12–22 days) is parallel to that at 500 hPa (10–20 days) (Figure 5c) indicating very similarly shaped distributions with only slightly slower transport at the surface. This is

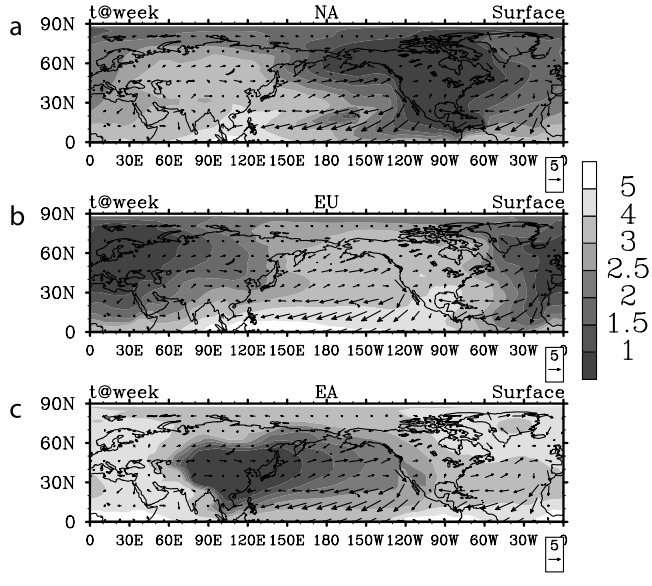

Figure 4. Horizontal distribution of average transport times for 1- and 2-week lifetime tracers, in April (1991 through 2001) of (a) NA, (b) EU, and (c) EA tracer at the surface (Units: color is time in weeks; arrows indicate wind velocity in m/sec). See color version of this figure in the HTML.

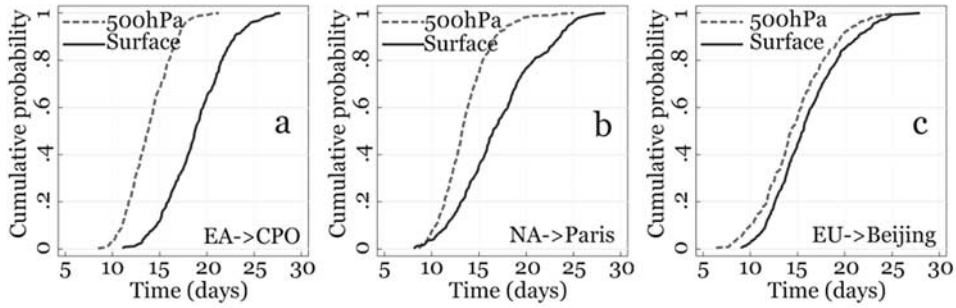

Figure 5. The cumulative probability distribution of the daily averaged transport times for 1- and 2-week lifetime tracers, in April (1991 through 2001) of tracer emissions transported (a) from EA to Cheeka Peak WA, USA, (b) from NA to Paris, France, and (c) from EU to Beijing, China. See color version of this figure in the HTML.

likely because, due to its high-latitude mid-continental emissions and therefore weak vertical transport in spring, the major transport pathway of the EU tracer is in the lower troposphere [*Liu et al.*, 2005].

4. Conclusions

[12] We present a method by which average inter-continental transport times can be estimated. The average transport times are approximated by the ratio of the concentrations of two chemical tracers with different first-order lifetimes which are emitted from the same source region and evaluated at a destination D. Tracers may travel by a myriad of different pathways. We find that average transport times increase with tracer lifetimes. This is because short-lived tracers decay more rapidly than long-lived tracers resulting in less short-lived than long-lived tracer arriving at a destination D via a long (or slow) pathway. Contributions from long pathways (or slow transport) fall in the right tail of the air parcel age spectrum at D.

[13] We develop an 'equivalent circulation' method to estimate a timescale which has little dependence on tracer lifetimes. We find the 'equivalent circulation method' gives results that are very similar to those obtained with short-lived tracers. After the influence of older air parcels are removed, the average transport times of EA tracer to the surface of CPO, WA, of NA tracer to Paris, France, and of EU tracers to Beijing, China in April (the month of rapid trans-Pacific transport) 1991–2001 range from 2 to 2.4 weeks.

[14] We use tracers with 1- and 2-week lifetimes to approximate the distribution of intercontinental transport times in April. We find that although the shape of the age spectrum of tracer transport varies, average tracer transport times in April over the 11-year period range from approximately 10 to 25 days at both the surface and 500hPa for transport between NA and Paris France, EU and Beijing China, and EA and CPO, Washington USA. Our findings show that average inter-continental transport times, even for tracers with short lifetimes, are on average approximately 1–2 weeks longer than inter-continental transport times that have been observed in rapidly transport in plumes.

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References

Bailey, R., L. A. Barrie, C. J. Halsall, P. Fellin, and D. C. G. Muir (2000), Atmospheric organochlorine pesticides in the western Canadian Arctic: Evidence of transpacific transport, *J. Geophys. Res.*, *105*(D9), 11,805–11,811.

Hall, T. M., and R. A. Plumb (1994), Age as a diagnostic of stratospheric transport, *J. Geophys. Res.*, *99*(D1), 1059–1070.

Holzer, M., I. G. McKendry, and D. A. Jaffe (2003), Springtime trans-Pacific atmospheric transport from east Asia: A transit-time probability density function approach, *J. Geophys. Res.*, *108*(D22), 4708, doi:10.1029/2003JD003558.

Horowitz, L. W., et al. (2003), A global simulation of tropospheric ozone and related tracers: Description and evaluation of MOZART, version 2, *J. Geophys. Res.*, *108*(D24), 4784, doi:10.1029/2002JD002853.

Husar, R. B., et al. (2001), Asian dust events of April 1998, *J. Geophys. Res.*, *106*(D16), 18,317–18,330.

Jaffe, D., et al. (1999), Transport of Asian air pollution to North America, *Geophys. Res. Lett.*, *26*(6), 711–714.

Jaffe, D., I. McKendry, T. Anderson, and H. Price (2003), Six 'new' episodes of trans-Pacific transport of air pollutants, *Atmos. Environ.*, *37*(3), 391–404.

Liu, J., D. L. Mauzerall, and L. W. Horowitz (2005), Analysis of seasonal and inter-annual variability in trans-Pacific transport, *J. Geophys. Res.*, *110*, D04302, doi:10.1029/2004JD005207.

Mauzerall, D. L., J. A. Logan, D. J. Jacob, B. E. Anderson, D. R. Blake, J. D. Bradshaw, B. Heikes, G. W. Sachse, H. Singh, and B. Talbot (1998), Photochemistry in biomass burning plumes and implications for tropospheric ozone over the tropical South Atlantic, *J. Geophys. Res.*, *103*(D7), 8401–8423.

McKeen, S. A., and S. C. Liu (1993), Hydrocarbon ratios and photochemical history of air masses, *Geophys. Res. Lett.*, *20*(21), 2363–2366.

McKeen, S. A., S. C. Liu, E. Y. Hsie, X. Lin, J. D. Bradshaw, S. Smyth, G. L. Gregory, and D. R. Blake (1996), Hydrocarbon ratios during PEM-WEST A: A model perspective, *J. Geophys. Res.*, *101*(D1), 2087–2109.

Plumb, R. A., and D. D. McConalogue (1988), On the meridional structure of long-lived tropospheric constituents, *J. Geophys. Res.*, *93*(D12), 15,897–15,913.

Stohl, A., S. Eckhardt, C. Forster, P. James, and N. Spichtinger (2002), On the pathways and timescales of intercontinental air pollution transport, *J. Geophys. Res.*, *107*(D23), 4684, doi:10.1029/2001JD001396.

Stohl, A., et al. (2003), Rapid intercontinental air pollution transport associated with a meteorological bomb, *Atmos. Chem. Phys.*, *3*, 969–985.

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