Supplementary A1: International Ozone Metrics (Metrics, section 3)

Table S1 describes the national and regional ozone limit values across the World and includes references to air quality standard web-sites. Figure S1 demonstrates differences between NDGT60 and NDGT70 for present day ozone levels as shown in Figure 4b in section 5.1.

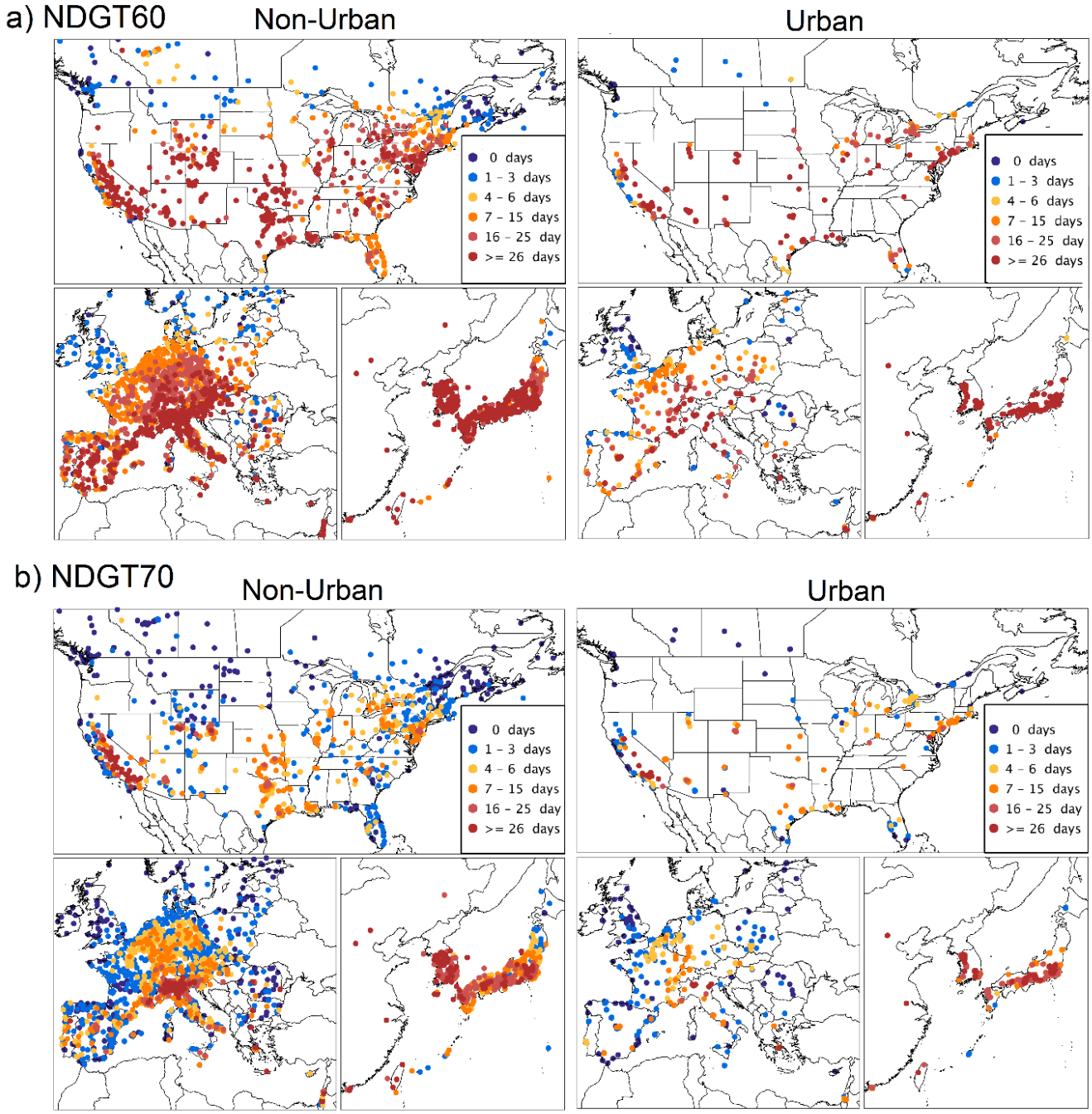


Figure S1. Present-day distributions of NDGT60 compared to NDGT70 for non-urban and urban sites.
Present day (2010-2014) average distributions of NDGT60 and NDGT70 (days/ yr) for non-urban and urban sites across North America, Europe and East Asia.
Full resolution images are available from: https://doi.pangaea.de/10.1594/PANGAEA.876108

Table S1. International, regional, national ozone limit values, averaging periods and references. Units listed in the table are those used in the official reporting.

	Country	**Averaging period**	**Value**	**Source**
WHO	WHO	8 h	100 µg m^{-3} *Interim Target 1 (IT-1):* *160 µg* m^{-3}	– http://apps.who.int/iris/bitstream/10665/69477/1/WHO_SDE_PHE_OEH_06.02_eng.pdf
Europe	EU *Austria, Belgium, Bulgaria, Croatia, Rep. of Cyprus, Czech Republic, Denmark, Estonia, Finland, France, Germany, Greece, Hungary, Ireland, Italy, Latvia, Lithuania, Luxembourg, Malta, Netherlands, Poland, Portugal, Romania, Slovakia, Slovenia, Spain*	8 h	120 µg m^{-3} *not to be exceeded on more than 25 days per calendar year averaged over 3 years* (The maximum daily eight-hour mean concentration shall be selected by examining eight-hour running averages, calculated from hourly data and updated each hour. Each eight -hour average so calculated shall be assigned to the day on which it ends. i.e. the first calculation period for any one day will be the period from 17:00 on the previous day to 01:00 on that day; the last calculation period for any one day will be the period from 16:00 to 24:00 on the day.)	– http://ec.europa.eu/environment/air/quality/standards.htm – http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:32008L0050 Annex VII A.2.B. – (directive 2008/50/EU)
	France (EU, EEA[2])	8 h	120 µg m^{-3} *not to be exceeded on more than 25 days per calendar year* *("Target value")*	– http://www.airparif.asso.fr/_pdf/publications/comparaison-internationale-atmofrance-en.pdf (Figure 5)
			120 µg m^{-3} *No Exceedance* *("Quality objective")*	– http://www.airparif.asso.fr/_pdf/publications/bilan-2013-anglais.pdf

	UK (EU, EEA)	8 h	100 µg m^{-3} *Not to be exceeded more than 10 times per year*	– http://uk-air.defra.gov.uk/assets/documents/reports/aqeg/aqeg-ozone-report.pdf
	Norway (EEA)	8 h	80 µg m^{-3}	– https://www.fhi.no/ml/miljo/luftforurensninger/luftkvalitetskriterier/
		1 h	100 µg m^{-3}, *180 µg* m^{-3}*– for informing the public*	– https://www.fhi.no/en/op/public-health-report-2014/risk--protective-factors/air-pollution-in-norway---public-he/#ozone
	Switzerland	1 h	120 µg m^{-3} *1 exceedance per year*	– http://acm.eionet.europa.eu/docs/ETCACM_TP_2011_3_evaluation_AQ_LT_TV.pdf – http://www2.dmu.dk/atmosphericenvironment/expost/database/docs/aq_limit_values.pdf
		½ h	100 µg m^{-3} *For each month, 98% values should be < 100 µg* m^{-3}	
	Macedonia	8 h	120 µg m^{-3} *not to be exceeded on more than 25 days per calendar year averaged over 3 years*	– http://airquality.moepp.gov.mk/?page_id=1557&lang=en
	Montenegro	8 h	120 µg m^{-3} *Adopting EU standards*	– http://www.unep.org/Transport/Airquality/Montenegro.pdf
	Albania			– http://www.unece.org/fileadmin/DAM/thepep/en/workplan/ia4pi/presentations/Shortcut%20to%20berlin__marieta_workshop-Presentation.ppt.lnk.pdf – https://www.eea.europa.eu/soer/countries/al/air-pollution-state-and-impacts-albania
	Bosnia and Herzegovina	-	-	– http://www.unep.org/Transport/Airquality/Bosnia_Herzegovina.pdf
North America	US	8 h	70 ppb *Annual 4th-highest daily maximum 8-h mole fraction,*	– https://www.epa.gov/ozone-pollution/setting-and-reviewing-standards-control-ozone-pollution#standards – https://www.epa.gov/criteria-air-pollutants/naaqs-table

			averaged over 3 years.	– https://fas.org/sgp/crs/misc/R43092.pdf
		1 h	0.12 ppm *a) N. of days per year with max. hourly average mole fractions above 0.12 ppm ≤ 1* *b) 1 h standard applicable to all areas notwithstanding the* *promulgation of 8-h ozone standards (EPA proposed several options for when the 1-h standard would no longer apply)*	– https://www.epa.gov/green-book/green-book-1-hour-ozone-1979-area-information-naaqs-revoked – NAAQS Revoked on June 15, 2005 for all areas except the 8 h ozone nonattainment Early Action Compact Areas (EAC) areas
	Canada	8 h	65 ppb *3-year average of the annual 4th highest daily max 8-hour average mole fractions*	– http://www.ccme.ca/files/Resources/air/pm_ozone/PN_1526_2012_CWS_for_PM_and_Ozone_Final_Report.pdf – http://www.ec.gc.ca/default.asp?lang=En&n=56D4043B-1&news=A4B2C28A-2DFB-4BF4-8777-ADF29B4360BD –
			2015: 63 ppb 2020: 62 ppb	
Central America	Mexico	1 h	216 μg m^{-3}	– http://www.cleanairinstitute.org/calidaddelaireamericalatina/summary-airquality-la.pdf
		8 h	157 μg m^{-3}	
	Costa Rica	1 h	160 μg m^{-3}	
	Dominican R.	1 h	250 μg m^{-3}	
		8 h	160 μg m^{-3}	
	El Salvador	1 h	235 μg m^{-3}	
		8h	120 μg m^{-3}	
		Annual	60 μg m^{-3}	
	Jamaica	1 h	235 μg m^{-3}	
	Nicaragua	1 h	235 μg m^{-3}	

		8 h	160 µg m^{-3}	
	Panama	1 h	235 µg m^{-3}	
		8 h	157 µg m^{-3}	
	Puerto Rico	1 h	235 µg m^{-3}	
		8 h	147 µg m^{-3}	
	Belize, Honduras, Haiti, Cuba, Guatemala	-	-	
South America	Brazil	1 h	160 µg m^{-3} *Not to be exceeded more than 1 time per year*	– http://www.mma.gov.br/images/arquivo/80060/tabela%20padroes%20qualidade.pdf
	Argentina	1 h	196 µg m^{-3}; Buenos Aires: 235 µg/m3	– http://www.cleanairinstitute.org/calidaddelaireamericalatina/summary-airquality-la.pdf
		8 h	Buenos Aires: 157 µg m^{-3}	
	Bolivia	1 h	236 µg m^{-3}	
		8 h	La Paz: 100 µg m^{-3}	
		Annual	La Paz: 60 µg m^{-3}	
	Colombia	1 h	120 µg m^{-3}	– https://www.minambiente.gov.co/images/normativa/app/resoluciones/bf-Resoluci%C3%B3n%20610%20de%202010%20-%20Calidad%20del%20Aire.pdf
		8 h	80 µg m^{-3}	
	Chile	8 h	61 ppb *Annual 4th-highest daily maximum 8-h mole fraction, averaged over 3 years.*	– http://www.leychile.cl/Navegar?idNorma=208198
	Ecuador	1 h	160 µg m^{-3}	– http://www.cleanairinstitute.org/calidaddelaireamericalatina/summary-airquality-la.pdf
		8 h	120 µg m^{-3}	
	Peru	8 h	120 µg m^{-3}	
	Venezuela	1 h	200 µg m^{-3}	
		8 h	160 µg m^{-3}	
	Paraguay, Uruguay	-	-	

Asia	China **I: residential* *II: commercial* *III: industrial* *** I: special regions e.g. national parks* *II: Urban/* *Industrial*	1 h	* China I: 120 µg m^{-3} China II: 160 µg m^{-3} China III: 200 µg m^{-3} *based on the Chinese standard T=273K, P=101.325kPa:* * I: 47 ppb II: 75 ppb III: 93 ppb	– http://www2.dmu.dk/atmosphericenvironment/expost/database/docs/aq_limit_values.pdf – http://acm.eionet.europa.eu/docs/ETCACM_TP_2011_3_evaluation_AQ_LT_TV.pdf *(Categories I and II)* – List of Chinese NAAQS: Qiao, X., Jaffe, D., Tang, Y., Bresnahan, M., Song, J., 2015. *Evaluation of air quality in Chengdu, Sichuan Basin, China: are China's air quality standards sufficient yet?* Environ. Monit. Assess. 187, 11.
			China I: 160 µg m^{-3} China II: 200 µg m^{-3} *After 2016***	– http://transportpolicy.net/index.php?title=China:_Air_Quality_Standards
		8 h	China I: 100 µg m^{-3} China II: 160 µg m^{-3} *After 2016***	
	Japan	1 h	Should not exceed 0.06 ppm *(60 ppb)* photochemical oxidants (1973) 0.12 ppm *(warning level)* 0.24 ppm *(alert level)*	– http://www.env.go.jp/en/air/aq/aq.html
		-	-	– http://www2.dmu.dk/atmosphericenvironment/expost/database/docs/aq_limit_values.pdf
	India	1 h	180 µg m^{-3}*	– http://transportpolicy.net/index.php?title=India:_Air_Quality_Standards (*) – http://www.indiaenvironmentportal.org.in/files/826.pdf
		8 h	100 µg m^{-3}*	

			**24 h, 8 h or 1 h monitored values, shall be compiled with 98 % of the time in a year. 2 % of the time, they may exceed the limits but not on 2 consecutive days of monitoring.*	– http://acm.eionet.europa.eu/docs/ETCACM_TP_2011_3_evaluation_AQ_LT_TV.pdf – http://www.cpcb.nic.in/upload/NewItems/NewItem_192_NAAQSTI.pdf
	Russia	1 time max *Average: 20-30 min*	100 µg m^{-3} (50 ppb) *workers*	– http://pgia.ru:81/seminar/archive/2006/6_atm/V_I_demin2.pdf
		1 time max	160 µg m^{-3} (80 ppb) *population*	
		24 h	30 µg m^{-3} (15 ppb) *population*	
	South Korea	8 h	≤ 0.06 ppm	– http://transportpolicy.net/index.php?title=South_Korea:_Air_Quality_Standards
		1 h	≤ 0.1 ppm	
	Thailand	8 h	0.14 mg m^{-3} (0.07 ppm) *= 140 µg* m^{-3}	– http://transportpolicy.net/index.php?title=Thailand:_Air_Quality_Standards – http://www.pcd.go.th/info_serv/reg_std_airsnd01.html
		1 h	0.2 mg m^{-3} (0.10 ppm) = *200 µg* m^{-3}	
	Turkey		*Looking to match EU guidelines?*	– http://www.eea.europa.eu/publications/air-pollution-by-ozone-across-EU-2012 – http://www.sisef.it/iforest/contents/?id=ifor0585-004

Africa	South Africa	8 h	120 µg m^{-3} *No more than 11 exceedances*	– https://law.resource.org/pub/za/ibr/za.sans.1929.2011.pdf – https://www.environment.gov.za/sites/default/files/docs/stateofair_executive_iaiquality_standardsonjectives.pdf – http://www.saaqis.org.za/Downloads.aspx?type=AQ (List of Air Quality Related Documents)
		1 h	200 µg m^{-3}	– https://www.environment.gov.za/sites/default/files/docs/stateofair_executive_iaiquality_standardsonjectives.pdf
	Egypt	1 h	200 µg m^{-3}	– http://www.eeaa.gov.eg/eimp/limit%20values.html
		8 h	120 µg m^{-3}	
Oceania	Australia	1 h	0.1 ppm *1 day/year exceedance*	– https://www.environment.gov.au/protection/air-quality/air-quality-standards – http://transportpolicy.net/index.php?title=Australia:_Air_Quality_Standards
		4 h	0.08 ppm *1 day/year exceedance*	
	New Zealand	1 h	150 µg m^{-3} *No exceedances*	– http://www.mfe.govt.nz/sites/default/files/2011-user-guide-nes-air-quality.pdf

1) Conversion factor: µg/m^3 = ppb*(12.187)*(48) / (273.15 + °C) at standard pressure (1013.25hPa), where 12.187=p/R from the ideal gas law. The conversion factor at 20 ºC is 1 ppb = 2 µg m^{-3} (at 25 ºC, 1 ppb = 1.96 µg m^{-3} ; at 0 ºC, 1 ppb = 2.242, and at 40 ºC 1 ppb = 1.868 µg/m^3). WHO (2008)
2) EEA is the European Environment Agency

WHO 2008. Health risks of ozone from long-range transboundary air pollution. World Health Organization Regional Office for Europe, ISBN 978 92 890 42895

Supplementary A2: Cluster analysis (Station classification, section 4.2)

Further independent classification of the sites based on these proxy data, in addition to tropospheric NO_2 column data at 0.1° resolution from the OMI satellite instrument (Krotkov et al. 2016), was carried out. Ward's method hierarchical cluster analysis was used to group the sites based on the similarity of the proxy variables (as well as station altitude) (Kaufman and Rouseeuw, 1990; Ward, 1963). Prior to application of the clustering algorithm, the proxy data were normalised so that variation across sites in each proxy variable had a mean of zero, and a standard deviation of 1. Using Ward's method hierarchical cluster analysis, each 'object', in this case each site represented by the normalised proxy variables, initially constitutes its own cluster. The two clusters were then merged that give rise to the smallest increase in within cluster variance. Within cluster variance (WCV) was quantified as the squared Euclidean distance between an object in the cluster (x_j), and the mean of that cluster (x_m), summed over all objects in that cluster (n) (Eq. 1).

$$WCV = \sum_{j=1}^{n}(x_j - x_m)^2 \qquad \text{Eq. 1}$$

The process of merging the two clusters which resulted in the smallest increase in WCV was repeated until all sites were contained within a single cluster. This process is visualised in a dendrogram, as shown in Figure S2. The dendrogram can 'but' at an appropriate height to group sites into a particular cluster set. The aim of this division is to maximise the variability across the sites explained by the cluster results while maintaining a small number of clusters. In this case six distinct clusters (termed elevated, rural, intermediate – rural, intermediate – urban, urban, most urban) were produced. An example of the increasing urbanicity of the sites in each of these classifications is shown in Figure S3, with the value of the NO_2 column and the population density increasing for each group defined as being more urban.

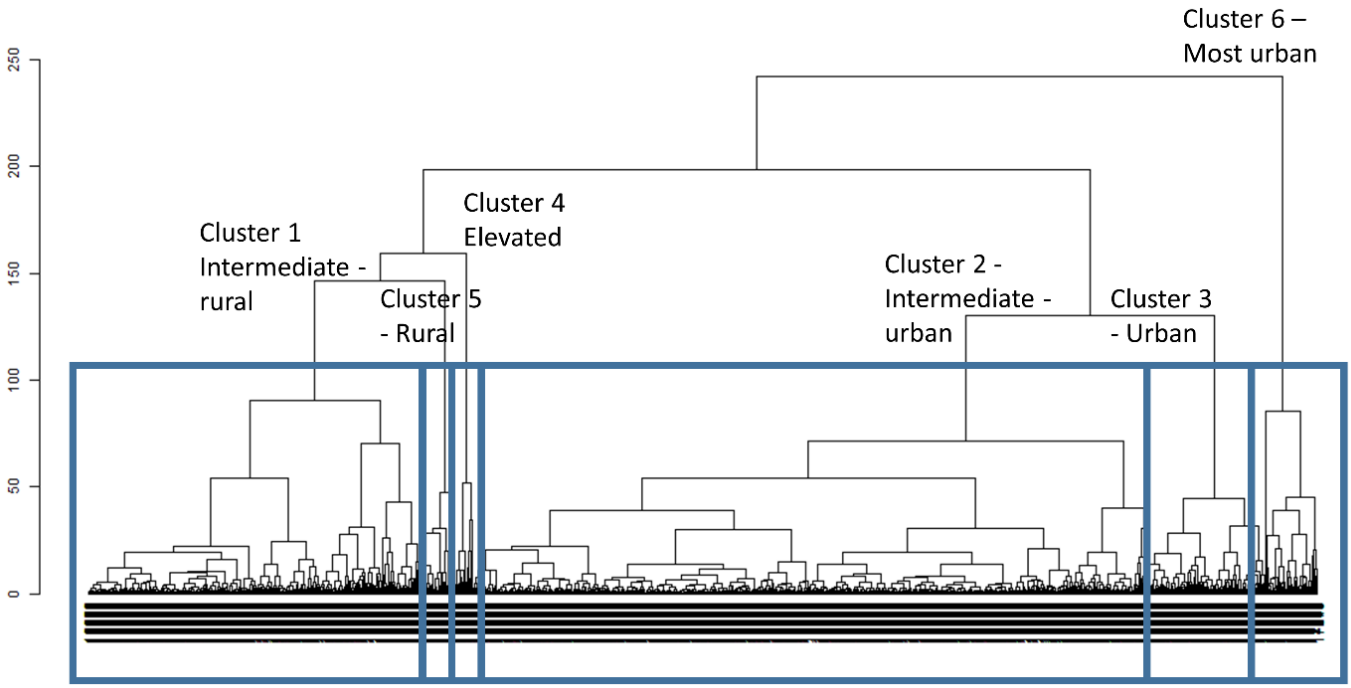


Figure S2. Dendrogram of site classifications using cluster analysis.

Site classifications are based on a cluster analysis of proxy datasets: NO_x emissions from OMI satellite, population density, nighttime lights. Cluster 1 Intermediate - Rural; Cluster 2 Intermediate - Urban; Cluster 3 Urban; Cluster 4 Elevated, Cluster 5 Rural; Cluster 6 Most Urban. There are 4812 sites.

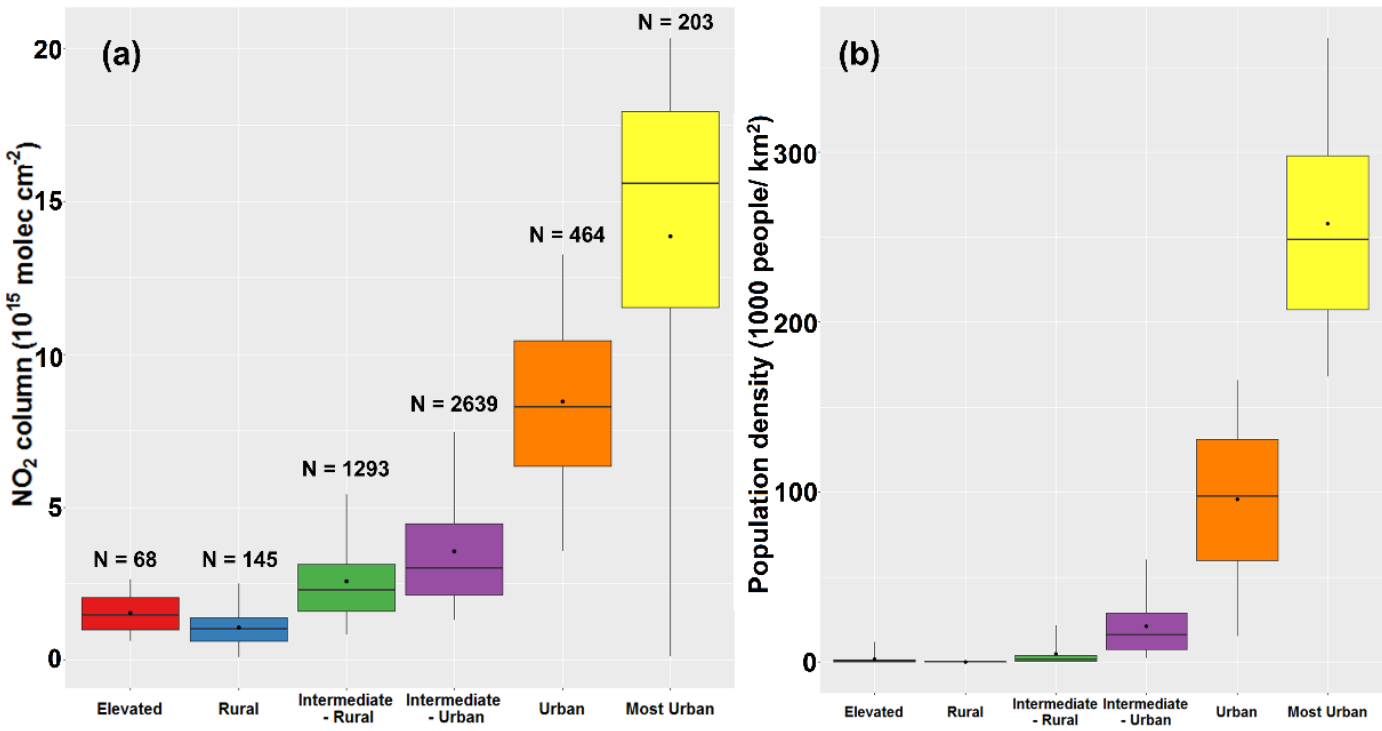


Figure S3. Box and whisker plots showing NO_2 and population density within each cluster.
NO_2 and population density within each of the 6 clusters produced from a cluster analysis of proxy variables (i.e. altitude, night-time lights, population density, NO_x emissions) at 4812 sites. a) Distribution of NO_2 column values across the sites in each cluster. b) Distribution of population density in each cluster.

Supplementary A3: Population exposed to NDGT60 > 25 days per year (section 5.2)

Further information for the population exposure analysis (percentage of the population within 5 km of a monitoring station exposed to NDGT60> 25 days) as carried out in 5.2 as shown in Figure 7 is provided. The number of stations used to calculate the percentage of the population exposed to NDGT60> 25 days is shown in Figure S4 and the percentage of the state or national population within 5 km of the stations is shown in Figure S5.

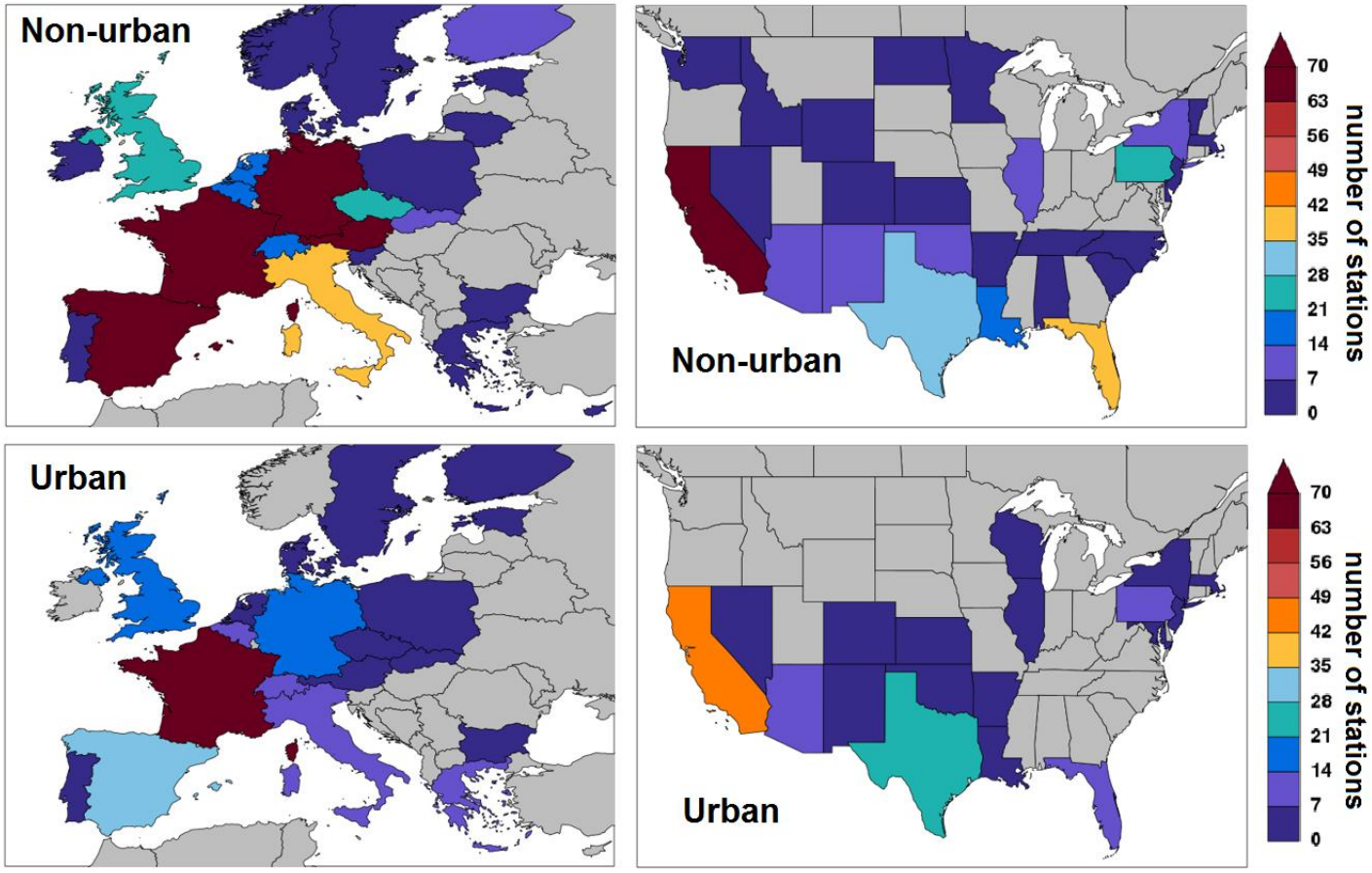


Figure S4. Number of non-urban and urban stations used to calculate NDGT60> 25 days per year for EU countries and US states.
Total stations by EU country or state of the US used for population exposure analysis. The colour scale has a maximum of 70 stations but there were 78, 126, 165, and 72 non-urban stations included in the analysis for Austria, France, Germany and Spain, respectively, and 79 urban stations in France. States and countries in grey have no data. Although whole EU countries/US states are coloured in the result, these pertain to locations within 5km radius of ozone monitoring locations.

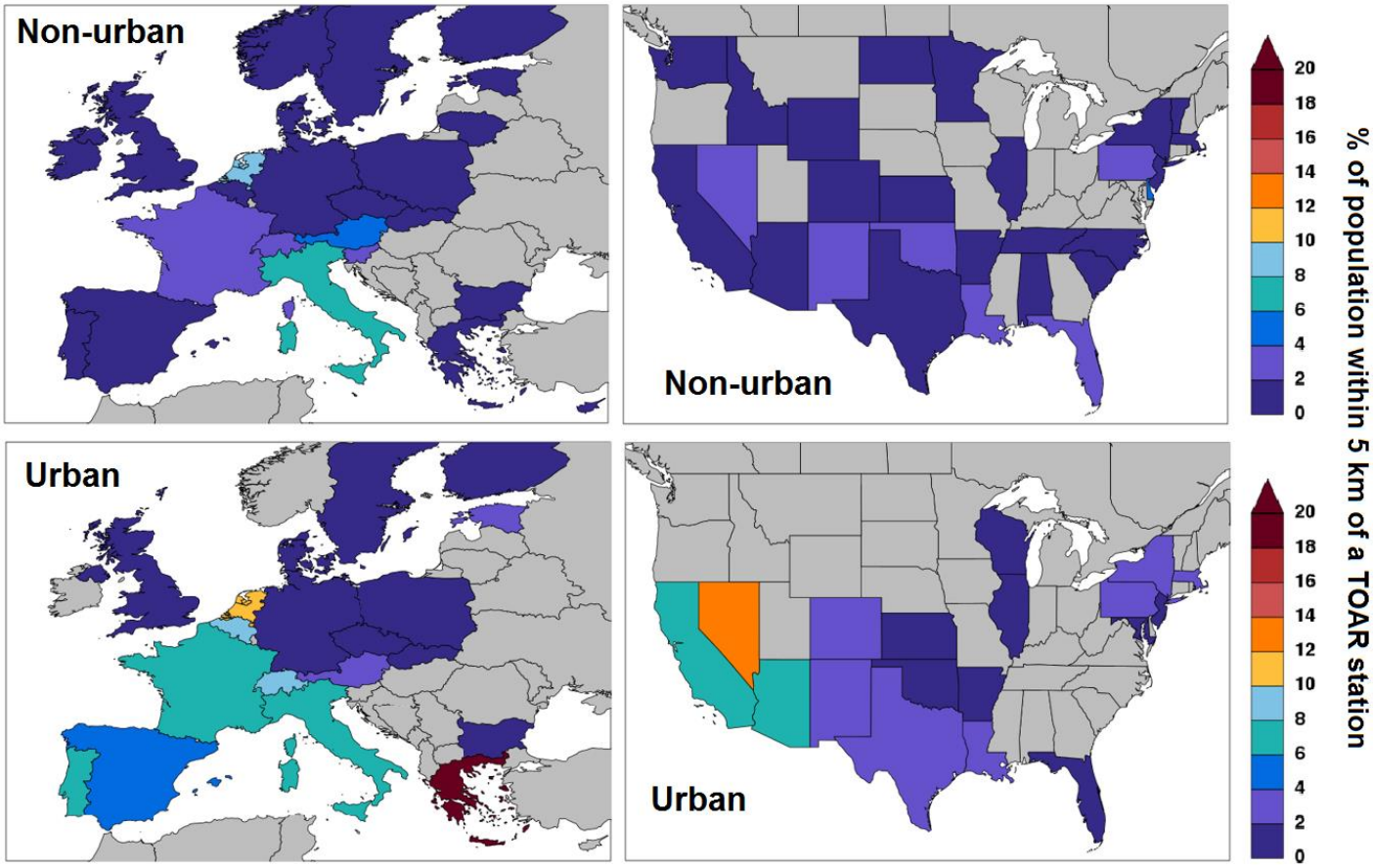


Figure S5. Percentage of the total state or national population within 5 km of TOAR stations. For urban and non-urban stations used in the population exposure analysis for European countries and US states. States and countries in grey have no data. Although whole EU countries/US states are coloured in the result, these pertain to locations within 5km radius of ozone monitoring locations.

Supplementary A4: Metric Sensitivity to alternative trend periods (section 6)

The averaging period (annual or warm season average) of the MDA8 calculation is investigated in Figure S6, to complement the regional trend analysis (with p value breakdown) shown for AVGMDA8 in Figure 9e in section 6.1.

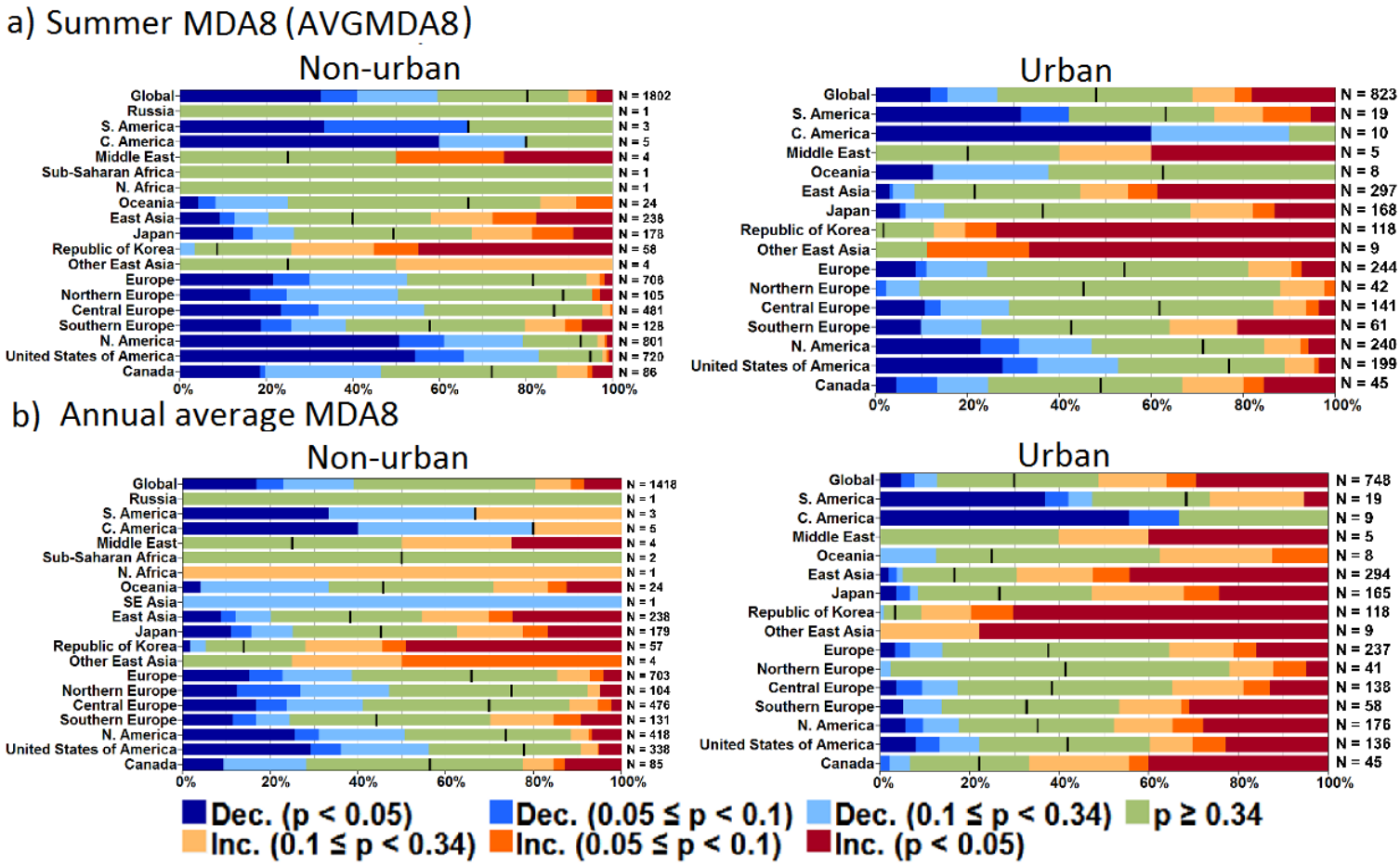
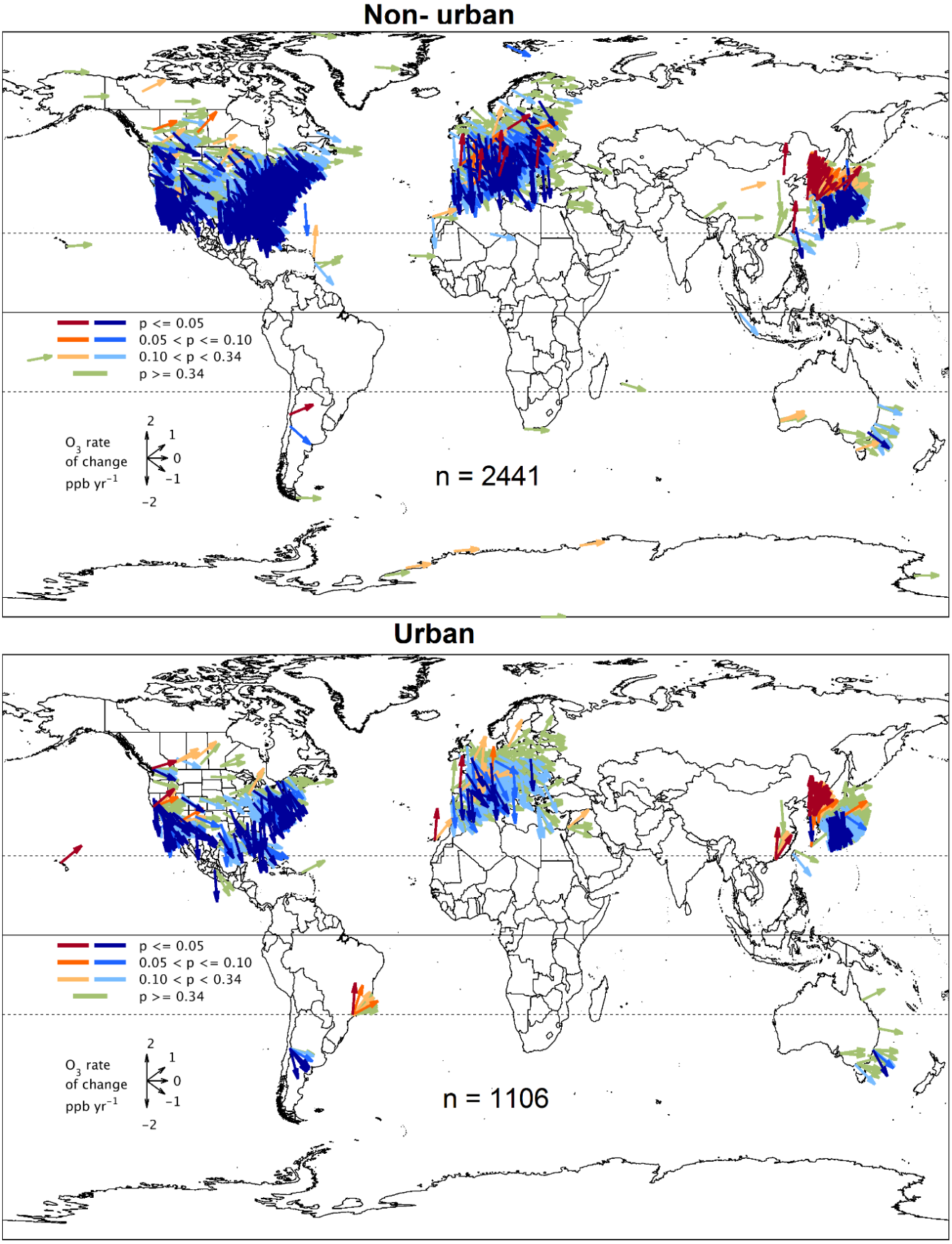


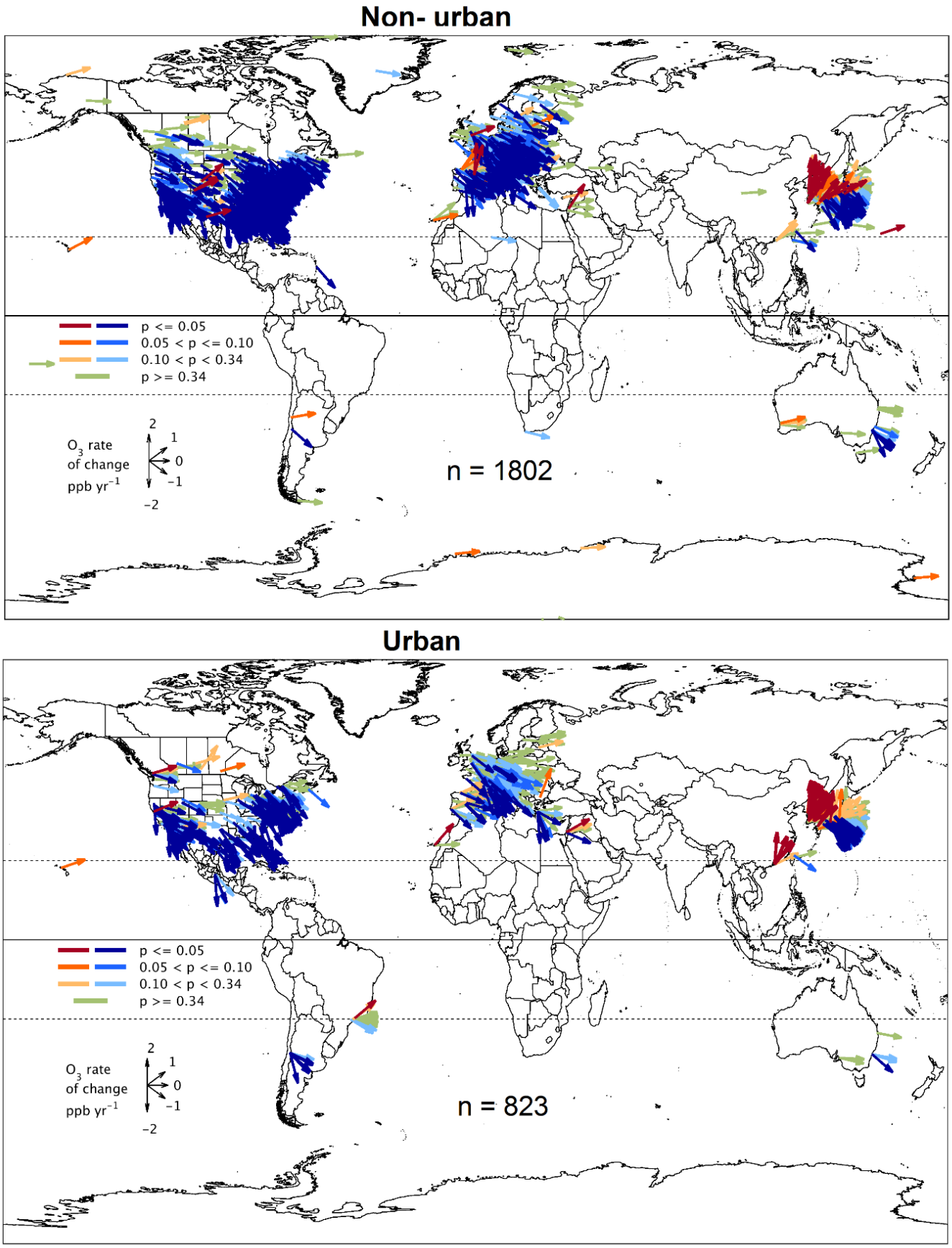
Figure S6. Sensitivity to averaging period used for AVGMDA8 calculation for non-urban and urban sites.
Proportion of monitoring sites in different HTAP regions (and sub-regions) with increasing and decreasing changes and trends over 2000-2014 for alternative ways of calculating average MDA8. Shown is the percentage of increasing and decreasing trends at various p values for a) summer AVGMDA8 mole fractions at non-urban sites (top left) and, urban (top right) sites, b) annual average MDA8 mole fractions at non-urban (bottom left) sites and urban (bottom right) sites.

Figures S7 to S9 provide the reader with three alternative trend periods (2005-2014, 1995-2014 and 1970-2014) to the 2000-2014 shown in Figures 8-10 in section 6. Figure S7 shows the trends for 4MDA8 for the alternative trend periods compared to 2000-2014 in Figure 8a. Figure S8 shows box and whisker plots for 4MDA8 for the alternate trend periods compared to Figure 10a. Figure S9 shows SOMO35 for the alternate trend periods compared to 2000-2014 in Figure 8c.



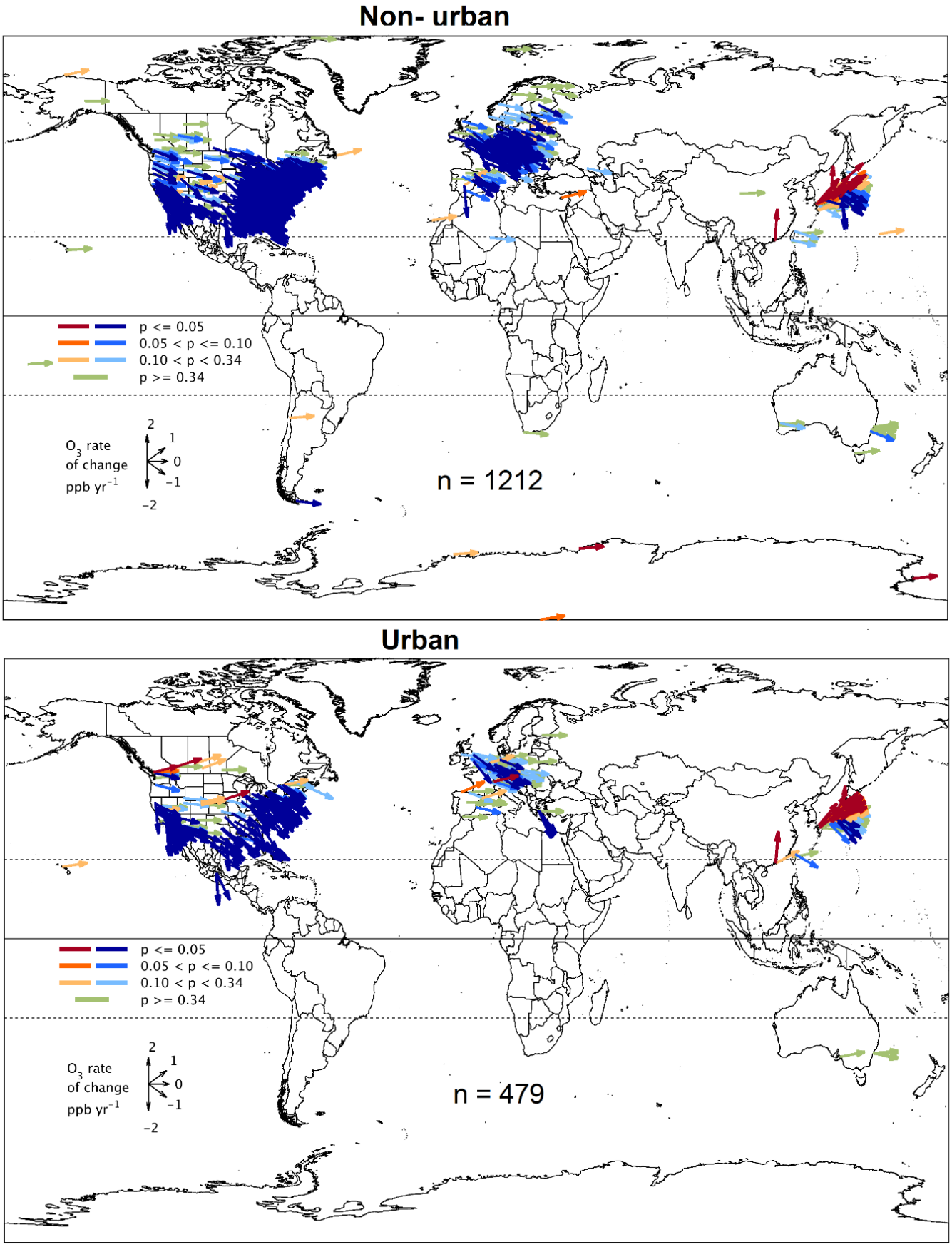
a) 10 yr, (2005-2014)

Figure S7. 4MDA8 trends over four different time periods for non-urban and urban sites.



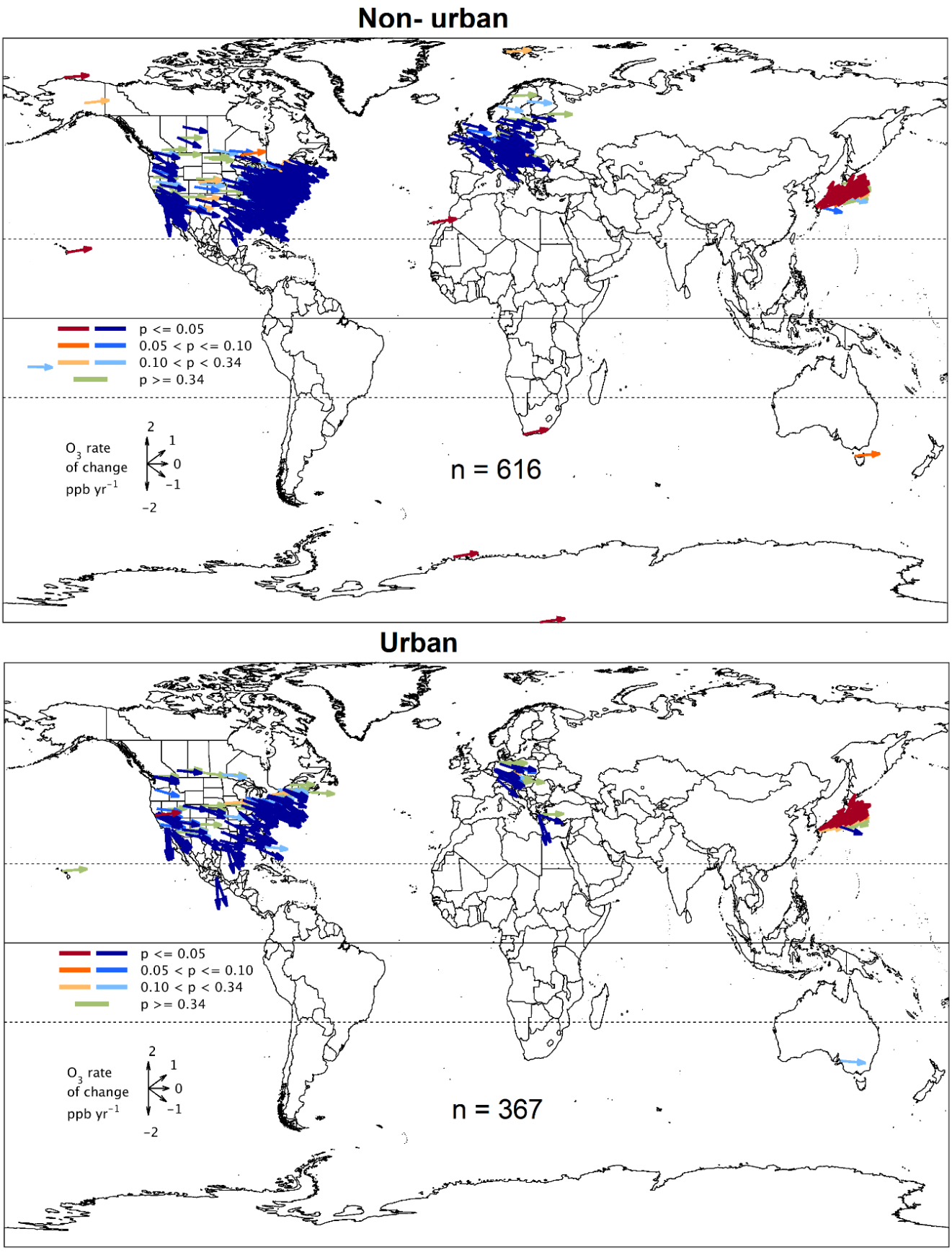
b) 15 yr (2000-2014)

Figure S7. 4MDA8 trends over four different time periods for non-urban and urban sites.



c) 20 yr (1995-2014)

Figure S7. 4MDA8 trends over four different time periods for non-urban and urban sites.



d) long term (at least 25 years) (1970-2014)

Figure S7. 4MDA8 trends over four different time periods for non-urban and urban sites.

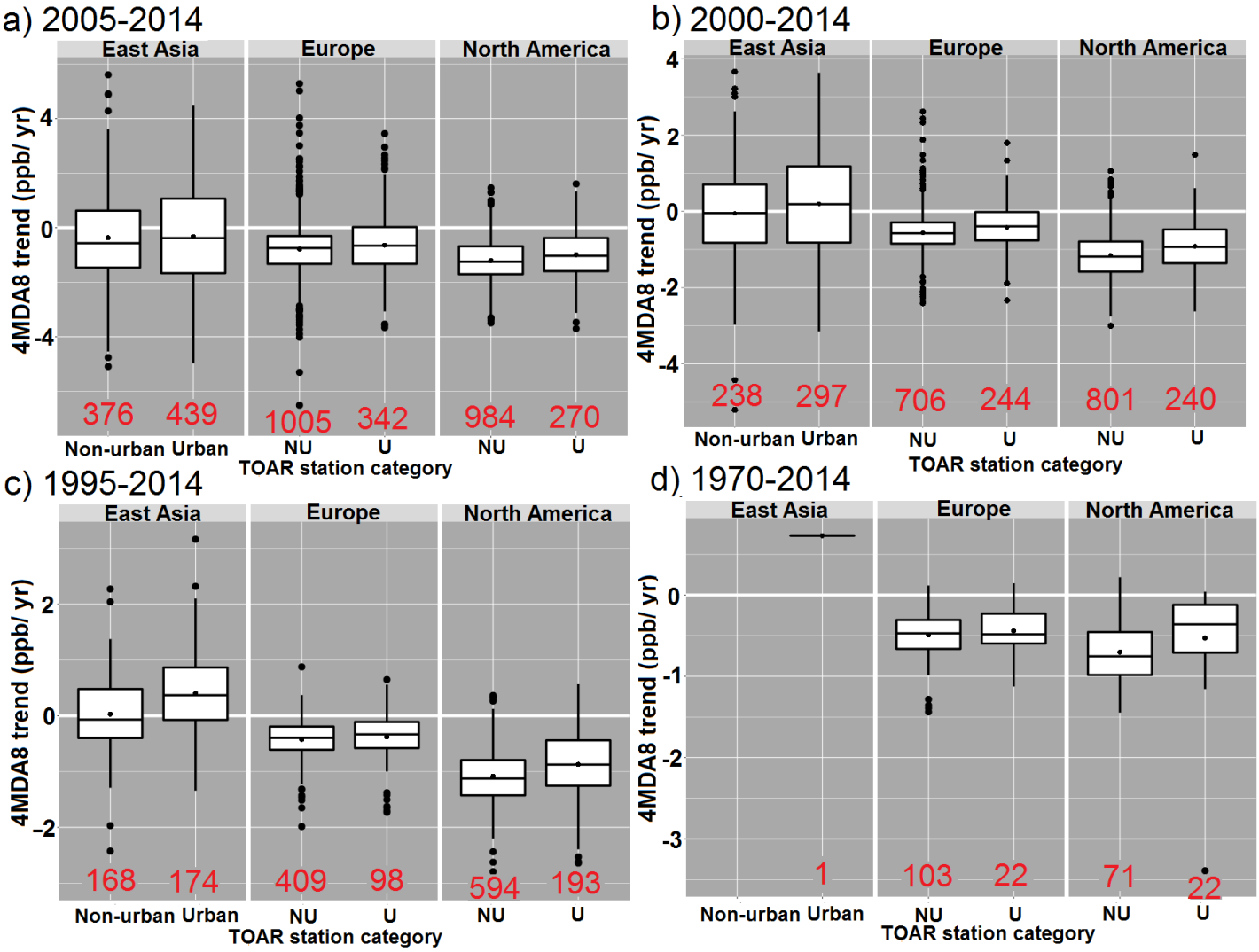
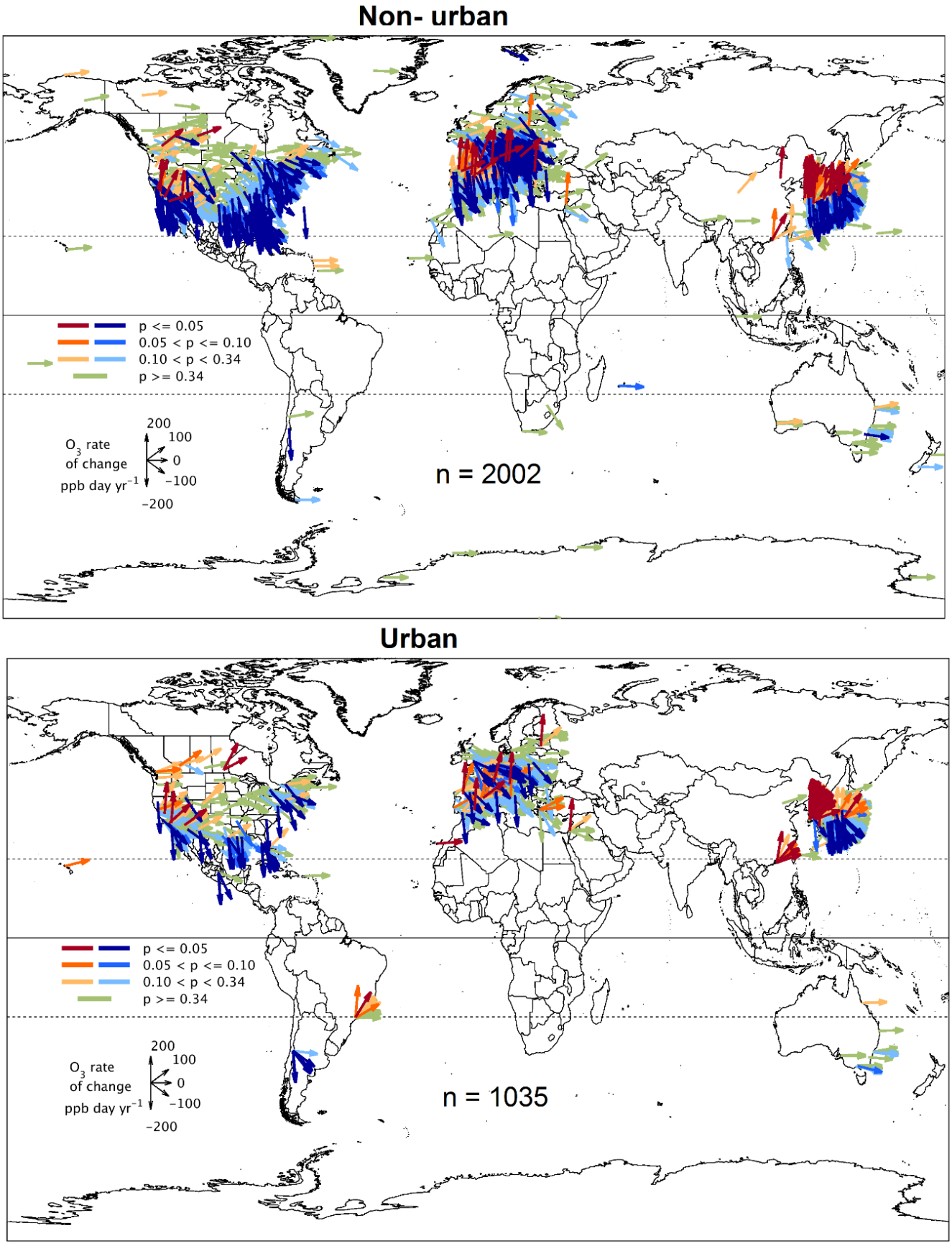
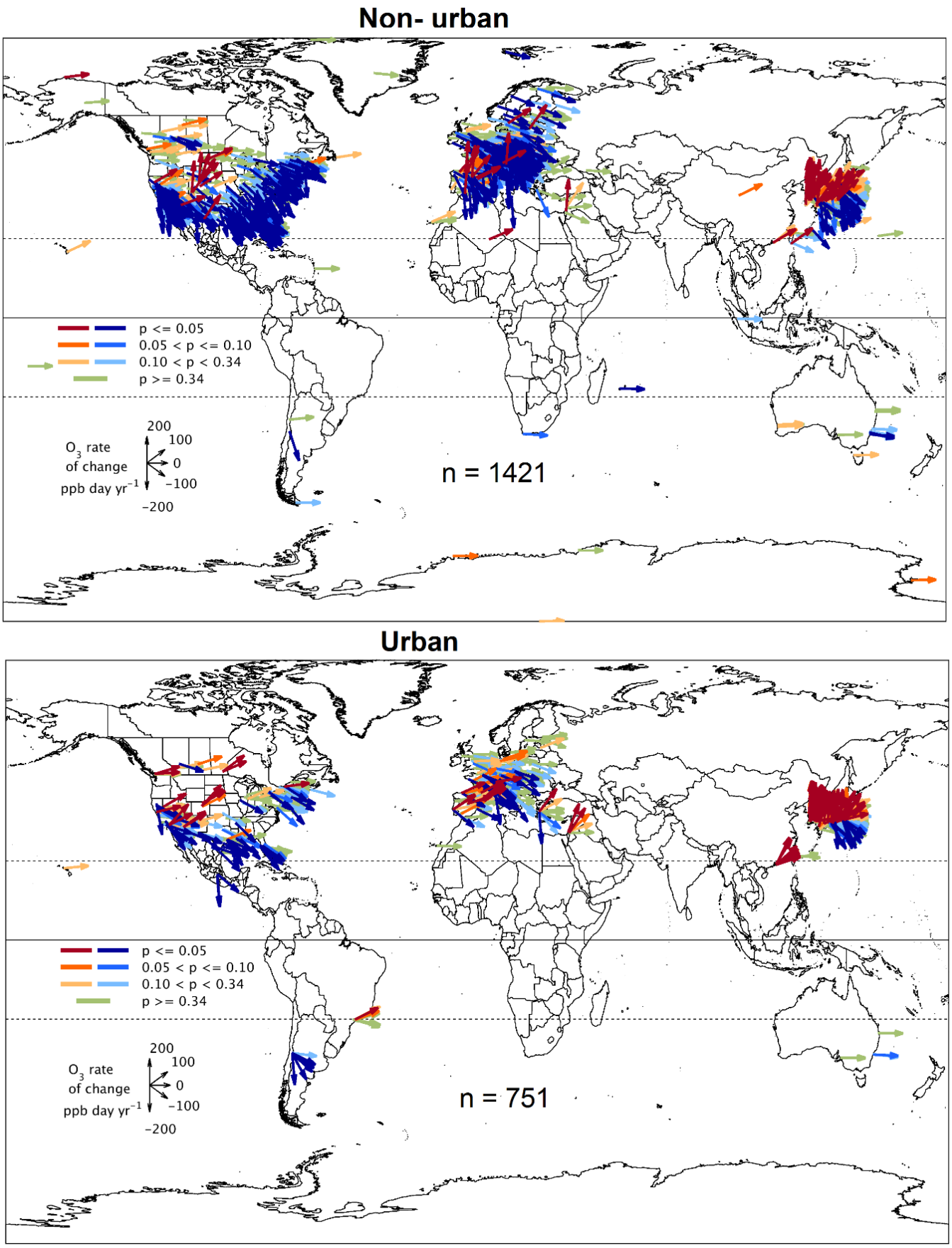


Figure S8. Regional Box and whisker plots for 4MDA8 for four trend periods.
4MDA8 (ppb/yr) trends for North America, Europe and East Asia across non-urban and urban sites for the following time periods: a) 10 yr (2005-2014), b) 15 yr (2000-2014), c) 20 yr (1995-2014) and d) 25+ yr (1970-2014) periods.



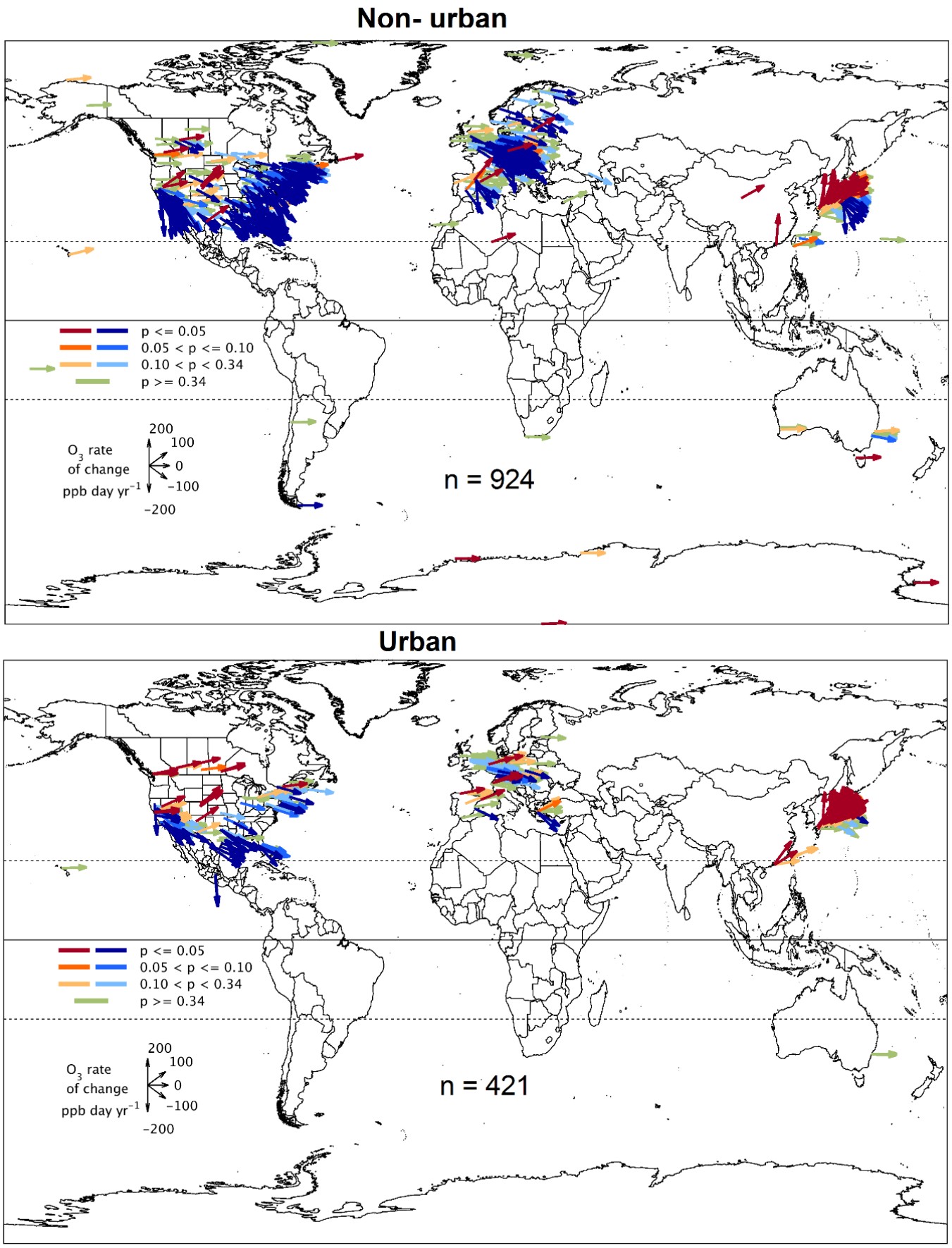
a) 10 yr (2005-2014)

Figure S9. SOMO35 trends over four different time periods for non-urban and urban sites.



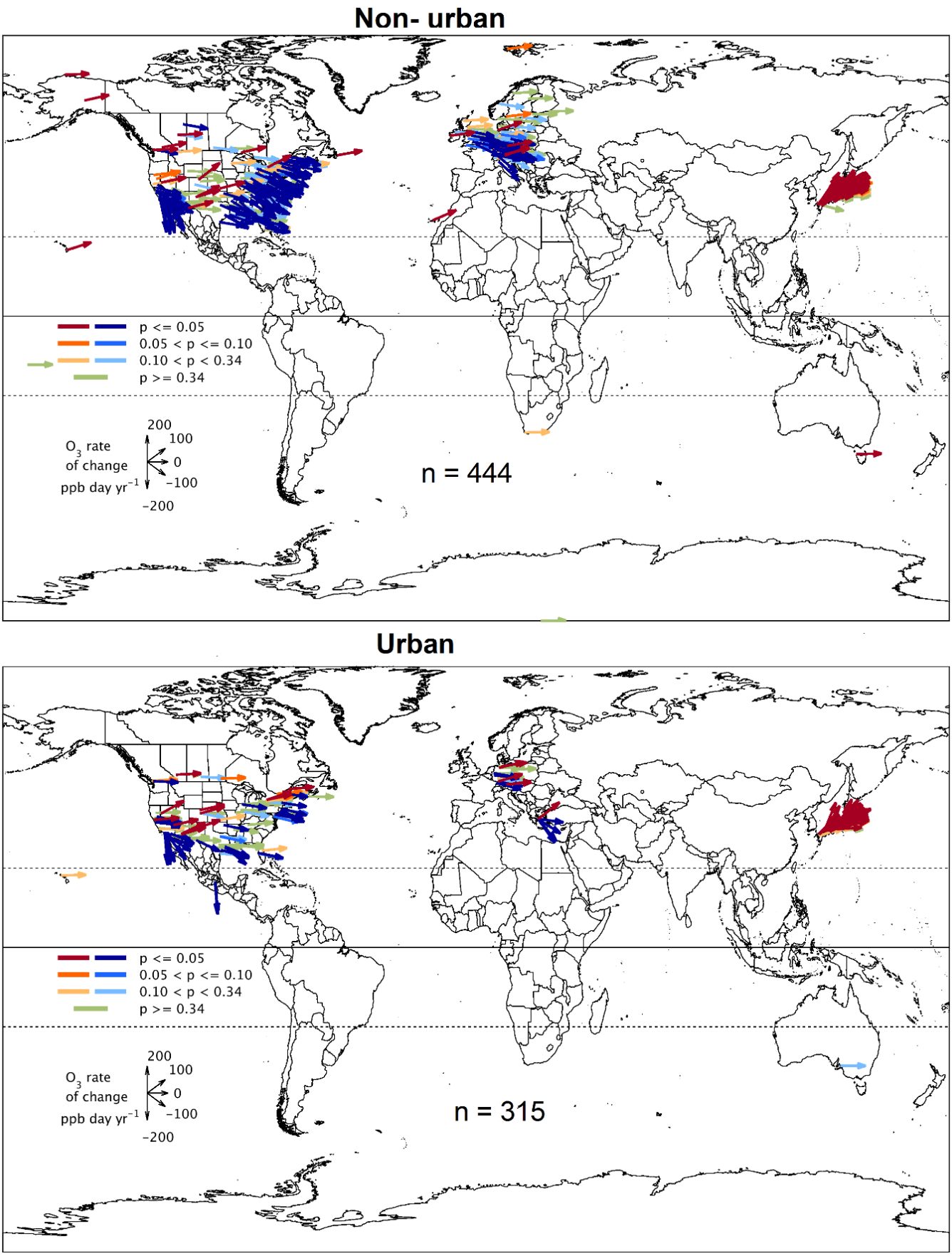
b) 15-yr (2000-2014)

Figure S9. SOMO35 trends over four different time periods for non-urban and urban sites.



c) 20 yr (1995-2014)

Figure S9. SOMO35 trends over four different time periods for non-urban and urban sites.



d) 25+ yr (1970-2014)

Figure S9. SOMO35 trends over four different time periods for non-urban and urban sites.