A Novel Method for Estimating NATA 2002 Background Concentrations

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Motivation

- The National Air Toxics Assessment (NATA) is EPA's comprehensive evaluation of hazardous air pollutants (HAPs, air toxics) in the U.S.
- The ASPEN (Assessment System for Population Exposure Nationwide) model and Human Exposure Model (HEM) were used to predict concentrations for every census tract in the United States using the National Emissions Inventory (NEI)
- Our goal was to provide accurate estimates of "background" concentrations for NATA 2002

Why Background Concentrations are needed for NATA

- ASPEN/HEM estimates long-term outdoor concentrations of air toxics attributable to 2002 anthropogenic emissions within 50 km of each census tract.
- For more accuracy, we need to account for background concentrations that are not represented by atmospheric modeling of nearby anthropogenic emissions.
- For many HAPs, outdoor concentrations should include "background" components attributable to
    - long-range transport,
    - persistent concentrations in the atmosphere,
    - and/or natural or unidentified emission sources.

Previous Background Concentration Estimates

- In NATA 1996, background concentrations were provided for 13 pollutants
  - Background concentrations were not spatially variable.
- In NATA 1999, spatially varying background concentrations were provided for 13 pollutants, and an additional 14 pollutants used a homogeneous background concentration.
  - Importantly, 5 pollutants identified with background contributing >85% of predicted concentrations.
  - Spatial variability in the 1999 assessment calculated using ambient measurements from 1995 – 2001; used to produce a regression between pollutant concentrations and population.
  - Background concentrations in unmonitored counties were inferred using county population as a surrogate.

http://www.epa.gov/ttn/atw/nata/backcon.html
http://www.epa.gov/ttn/atw/nata1999/background.html

Previous NATA Background Pollutants

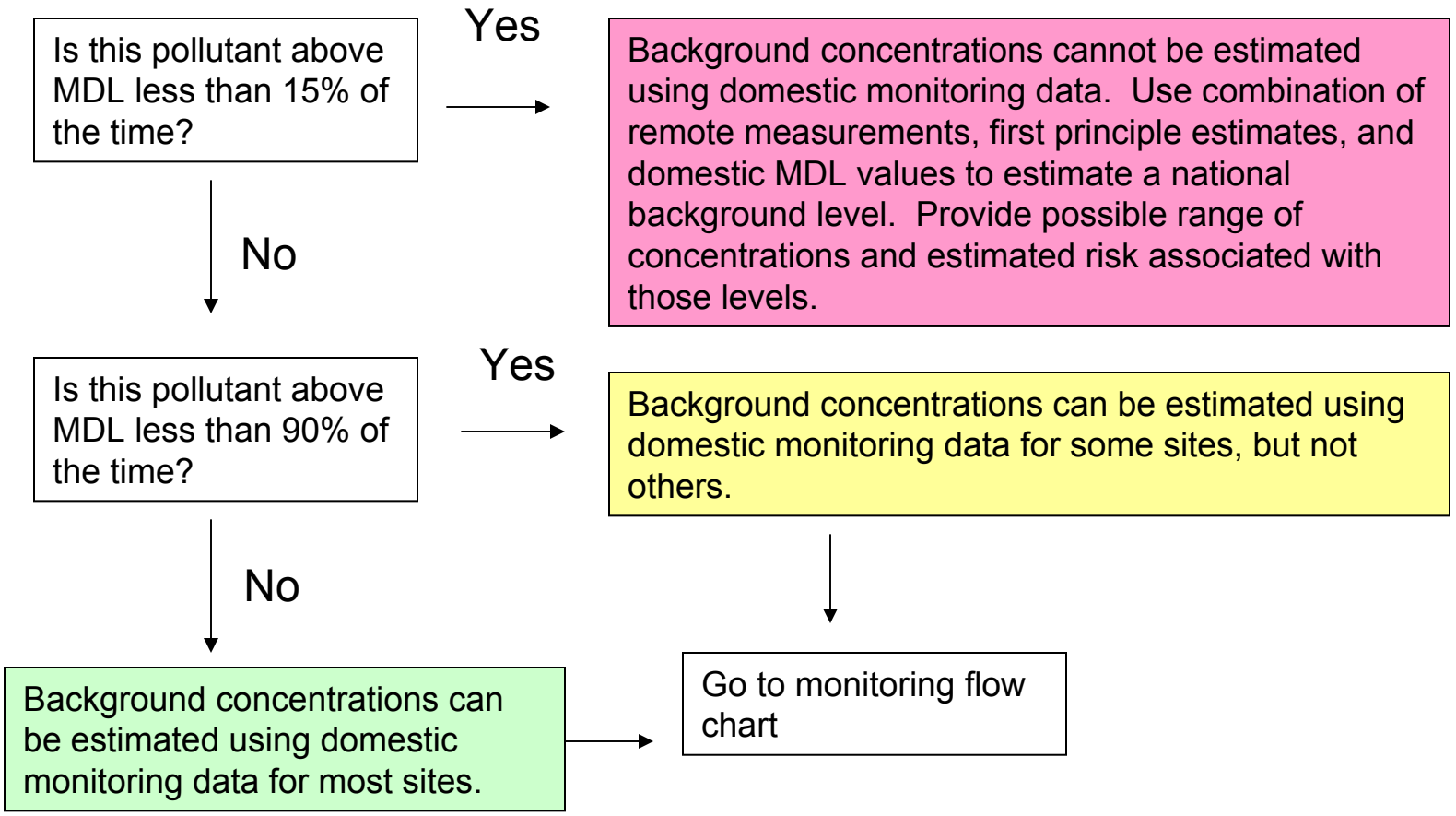
NATA 1996	NATA 1999	
Benzene	Benzene	1,3-Butadiene
Carbon tetrachloride	Carbon tetrachloride	*Bis(2-ethylhexyl)phthalate*
Chloroform	Chloroform	*Bromoform*
Dichloromethane	Dichloromethane	*Carbon disulfide*
Ethylene dibromide	Ethylene dibromide	*Chlordane*
Ethylene dichloride	Ethylene dichloride	*Hexachlorobutadiene*
Formaldehyde	Formaldehyde	*Hexachloroethane*
Mercury	*Mercury*	*Lindane*
Polychlorinated biphenyls	*Polychlorinated biphenyls*	*Methyl bromide*
Tetrachloroethylene	Tetrachloroethylene	*Methyl chloride*
Trichloroethylene	Trichloroethylene	*Methyl chloroform*
Hexachlorobenzene	Acetaldehyde	*Phosgene*
	1,1,2,2-Tetrachloroethane	Vinyl chloride
	1,2-Dichloropropane	*Xylenes*

Approach for NATA 2002

- Determine whether ambient measurements are sufficient to estimate background concentrations
  - Are there enough ambient measurements available for a given pollutant?
  - Are the measurements good enough to estimate background concentrations?
- For well-measured pollutants, background concentrations were estimated using ambient measurements from the domestic monitoring network in a method similar to that for NATA 1999.
- If pollutants were not well-measured in the ambient network, an emissions-based approach was used to estimate background concentrations.

This presentation focuses on the emissions-based approach.

Pollutant Decision Tree



Note: MDL = method detection limit

2002 NATA Background Pollutants

Ambient-based Method	Emissions-based Method	Assigned Concentrations
1,3-Butadiene	Hydrazine	Carbon tetrachloride
1,4-Dichlorobenzene	Chromium (VI)	Methyl Chloride
Acetaldehyde	Ethylene Dichloride	Methyl Bromide
Arsenic	Naphthalene	Methyl Chloroform
Benzene	Propylene Dichloride	
Chloroform	Ethylene Oxide	
Chromium	Acrylonitrile	
Dichloromethane	Cadmium	
Formaldehyde	Beryllium	
Lead	Ethylene Dibromide	
Manganese	Benzidine	
Nickel	Quinoline	
Tetrachloroethylene	Bis(2-Ethylhexyl)Phthalate	
Toluene	1,2-Dibromo-3-Chloropropane	
	Trichloroethylene	
	1,1,2,2-Tetrachloroethane	

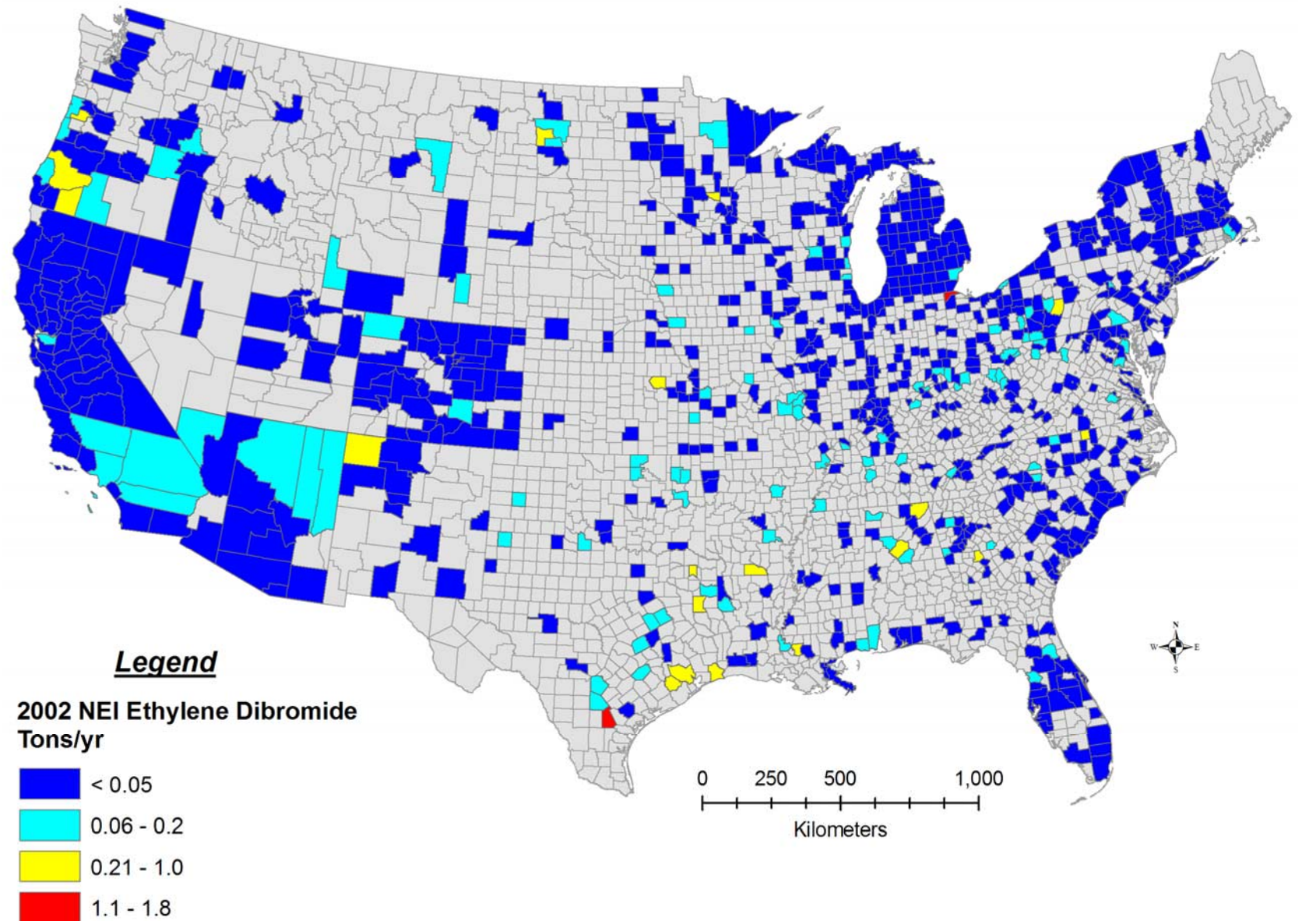
Emissions-based method

- Since ambient measurements alone are insufficient to determine background concentrations an alternate approach was needed
- We combined the following information to provide spatially varying background estimates
  - NEI 2002 emissions data
  - HAPs residence times
  - NATA 1999 model predicted concentrations in the highest U.S. county
  - Remote background concentration estimates

Emissions-based Approach

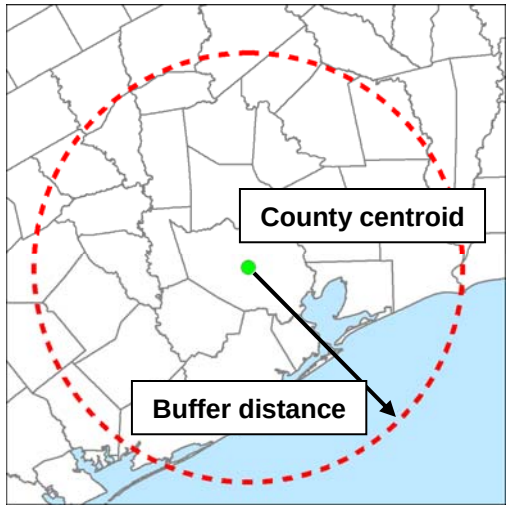
1. Import NEI data into a Geographic Information System (GIS) and create emission density maps
2. Apply a spatial weighting scheme to derive emissions gradients
3. Normalize the emissions gradients
4. Convert emissions gradients to background concentration values

1. Import NEI data into GIS



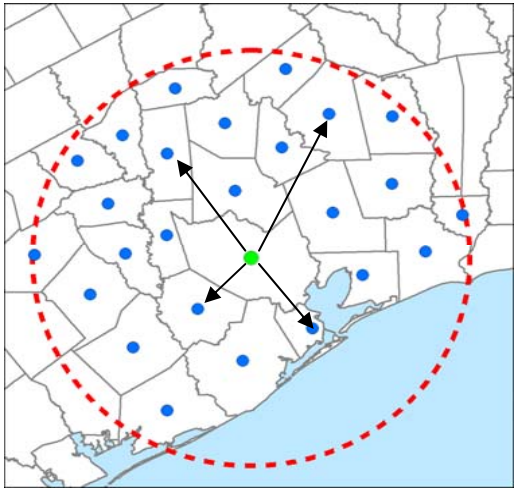
2a.Apply spatial weighting scheme

(1)



Calculate buffer distances for each pollutant and create buffers centered on county centroids.

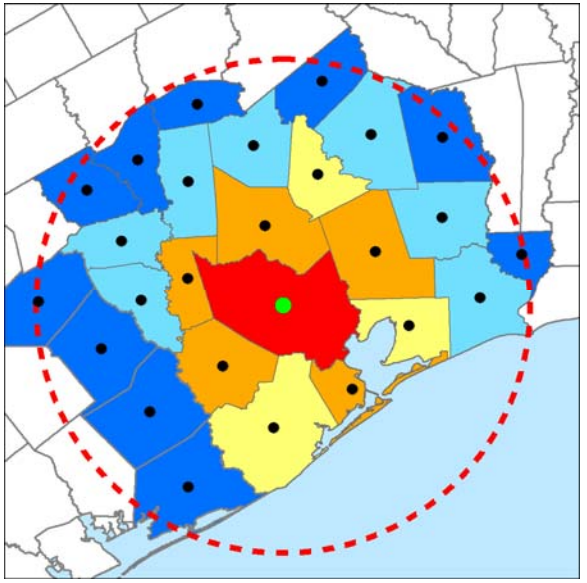
(2)



Calculate the distance(s) between all county centroids within the buffer. Each county centroid has an associated emissions value representing total county emissions for a specific pollutant.

(3)

Spatially distributed emissions values based on the spatial weighting scheme.

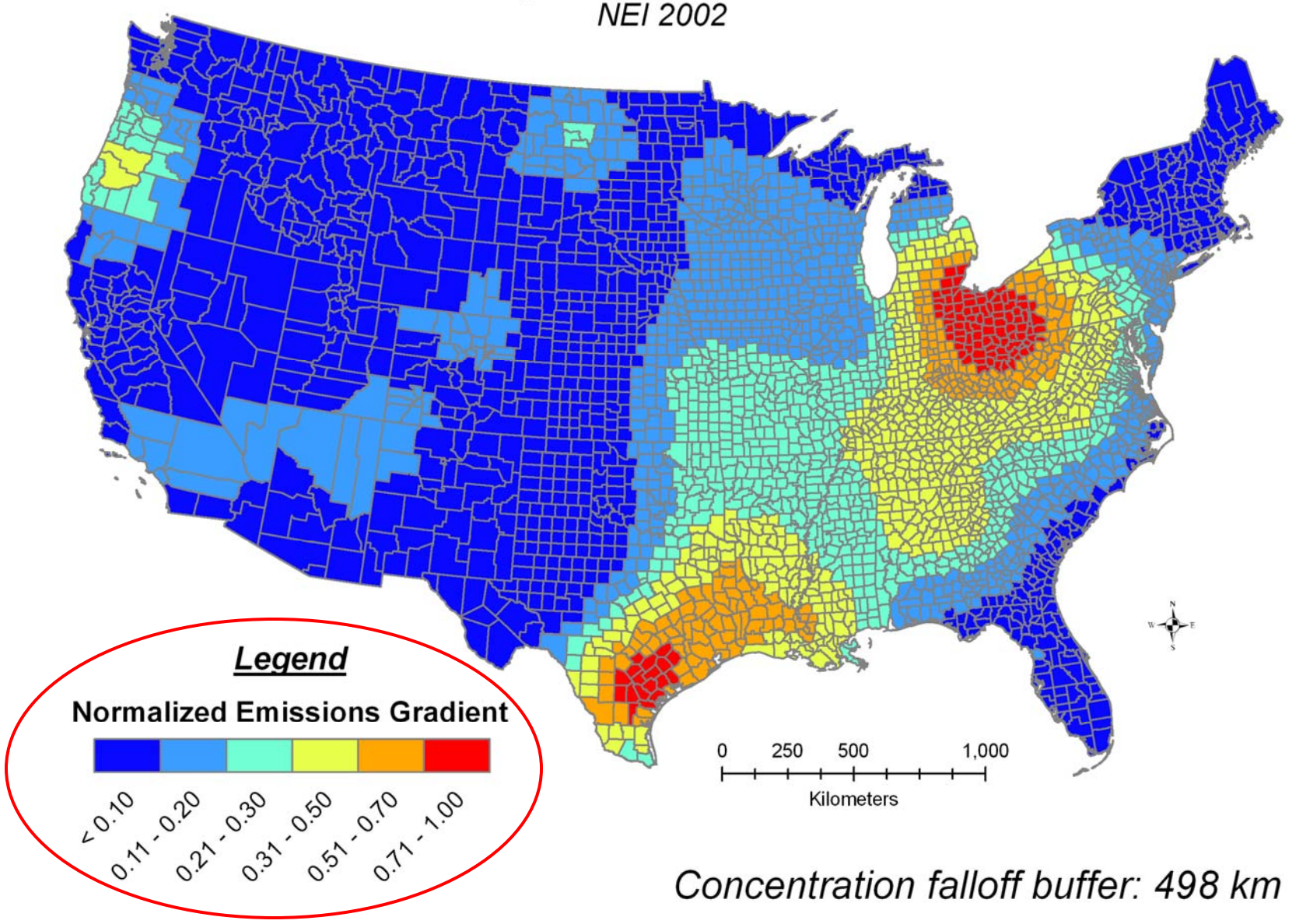


2b. Apply weighting scheme

3. Normalize emissions gradient

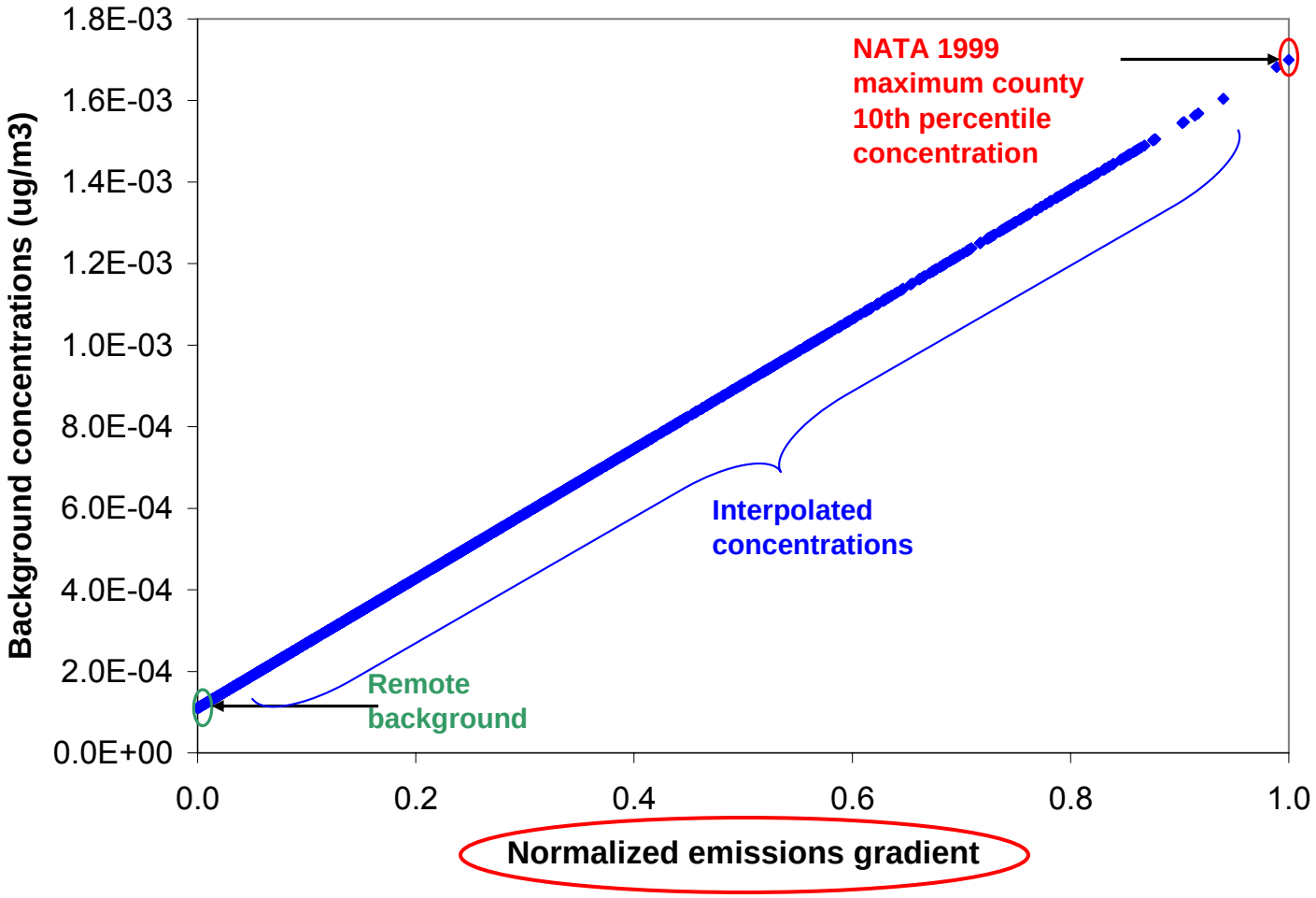
Ethylene Dibromide

NEI 2002



Concentration falloff buffer: 498 km

4a. Convert emissions gradient to background concentration



From the previous slide

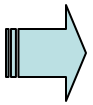
All we need are two “anchor” points to convert the normalized emissions gradient (x-axis) to background concentrations (y-axis).

4b. Estimating Remote Background Concentrations

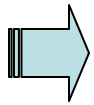
- For air toxics with little-to-no information from the monitoring network, we can estimate remote background concentrations based on first principles.
- Multiply National Emissions Inventory (NEI) emissions for the U.S. by the residence time of the pollutant in the atmosphere.
- Compare resulting weighted emissions to other pollutants that are measured. The ratio of these numbers can be used to provide an estimate of remote background concentrations. For example,

$$\left(\frac{\text{Trichloroethylene}}{\text{tetrachloroethylene}}\right) = 0.28$$

Weighted Emissions



Trichloroethylene Background ~ 28% Tetrachloroethylene



Trichloroethylene Background ≈ 0.0062 µg/m^3

(~0.022 µg/m^{3}x0.28)

Weighted Emissions:

10808 tpy / 35577 tpy * 6 days / 6.5days = 0.28

4c. Convert emissions gradients to background concentration (Proof of Concept)

Name	Res. time (days)	2002 Emissions (tons per year)	Emissions x residence time (tons)	Fraction of tetrachloro-ethylene	Measured remote conc. ($\mu g/m^3$)	Estimated remote conc. ($\mu g/m^3$)
Tetrachloro-ethylene	6.5	35577	633.1	1.00	0.022	0.022
Toluene	0.5	891520	1220.4	1.93	0.041	0.042
Chloroform	80	6782	1485.5	2.35	0.059	0.052
Benzene	3.0	410892	3375	5.33	0.14	0.12

Remote background Anchor Point: Calculate an estimated concentration based on NEI total emissions (E), residence time (t), and the relationship to tetrachloroethylene concentration [$C_{tetrachloroethylene}$].

$$[C_i] = \frac{E_i * t_i * [C_{tetrachloroethylene}]}{E_{tetrachloroethylene} * t_{tetrachloroethylene_i}}$$

4d. Convert emissions gradients to background concentration

Name	Res. time (days)	2002 Emissions (tons per year)	Emissions x residence time (tons)	Fraction of tetrachloro-ethylene	Measured remote conc. ($\mu g/m^3$)	Estimated remote conc. ($\mu g/m^3$)
Ethylene Dibromide	50	24	3.2	0.005		1.1E-04
Vinyl Chloride	2	1306	7.1	0.011		2.5E-04
Ethylene Oxide	7	695	13.3	0.02		0.0005
Tetrachloro-ethylene	6.5	35577	633.1	1.00	0.022	0.022

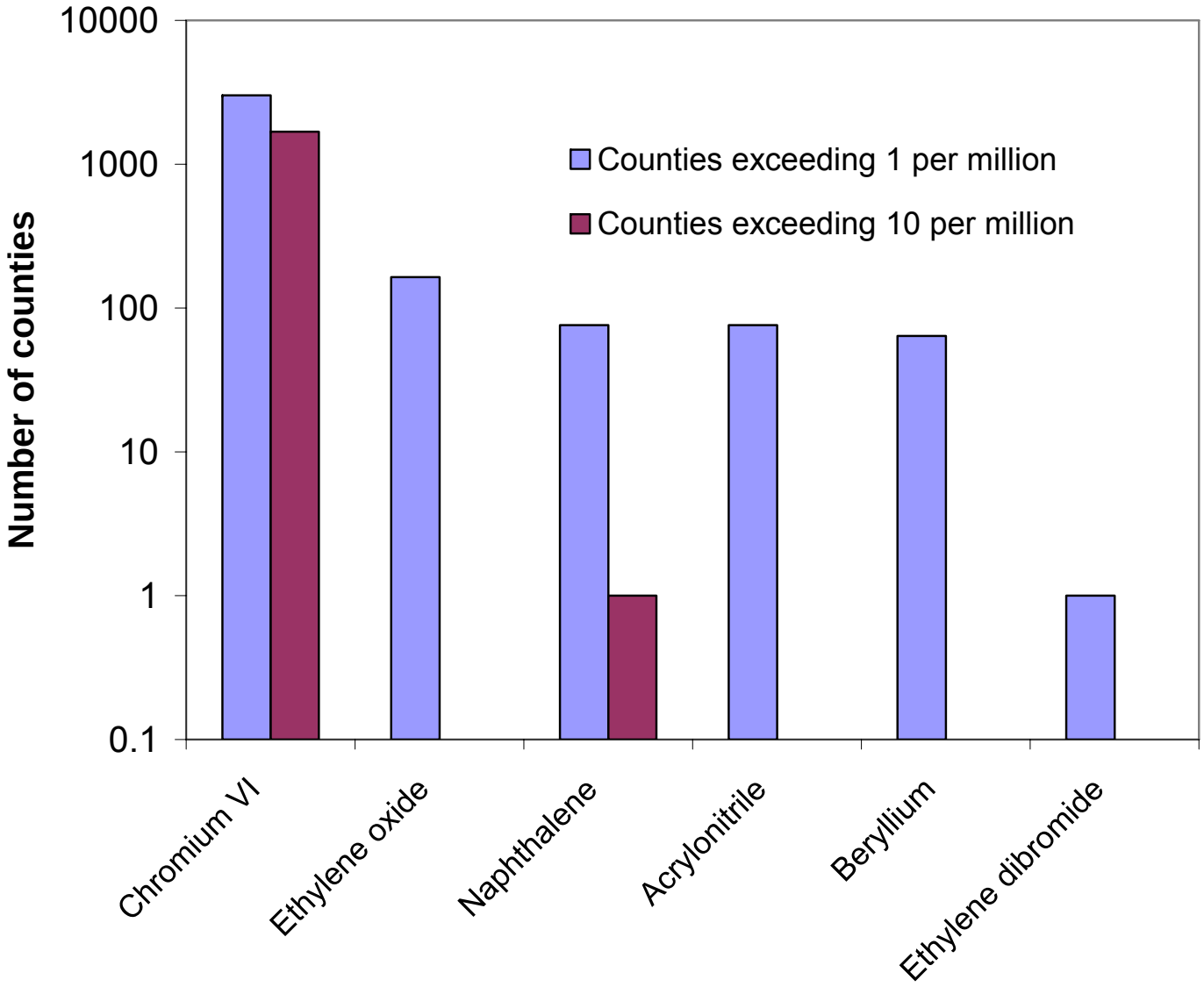
- These estimates
  - are a reasonable approximation; however, they are a function of the certainty in residence time and NEI approximation of U.S. and global emissions.
  - are not useful for pollutants with secondary production or non-anthropogenic emissions.

4e. Convert emissions gradients to background concentration

Upper background anchor point: Use the highest predicted county concentrations from NATA 1999. Take the 10th percentile concentration from that county.

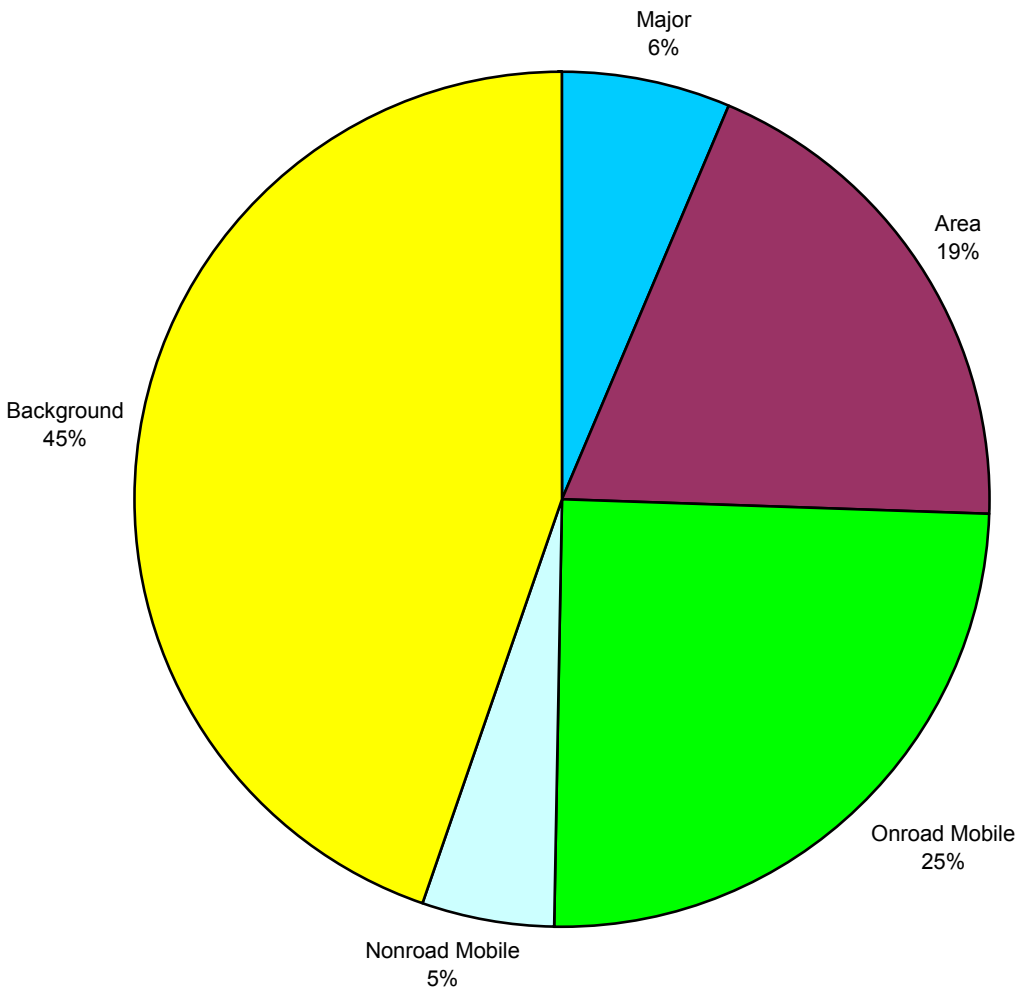
Pollutant	Remote estimate	1999 NATA max county 10th percentile	Cancer benchmark	RfC	Cancer Risk remote	Cancer Risk max	RFC max
Acrylonitrile	0.00055	0.0302	1.47E-02	2	0.0	2.1	0.02
Beryllium	1.80E-05	1.05E-03	4.10E-04	0.020	0.0	2.6	0.05
Benzidine	9.90E-09	6.78E-06	1.49E-05	10	0.0	0.5	0.00
Bis(2-ethylhexyl)phthalate	0.0052	7.77E-03	4.17E-01	10	0.0	0.0	0.00
Cadmium	3.70E-05	1.29E-04	5.50E-04	0.020	0.1	0.2	0.01
Chromium VI	2.20E-05	4.27E-03	8.33E-05	0.100	0.3	51.2	0.04

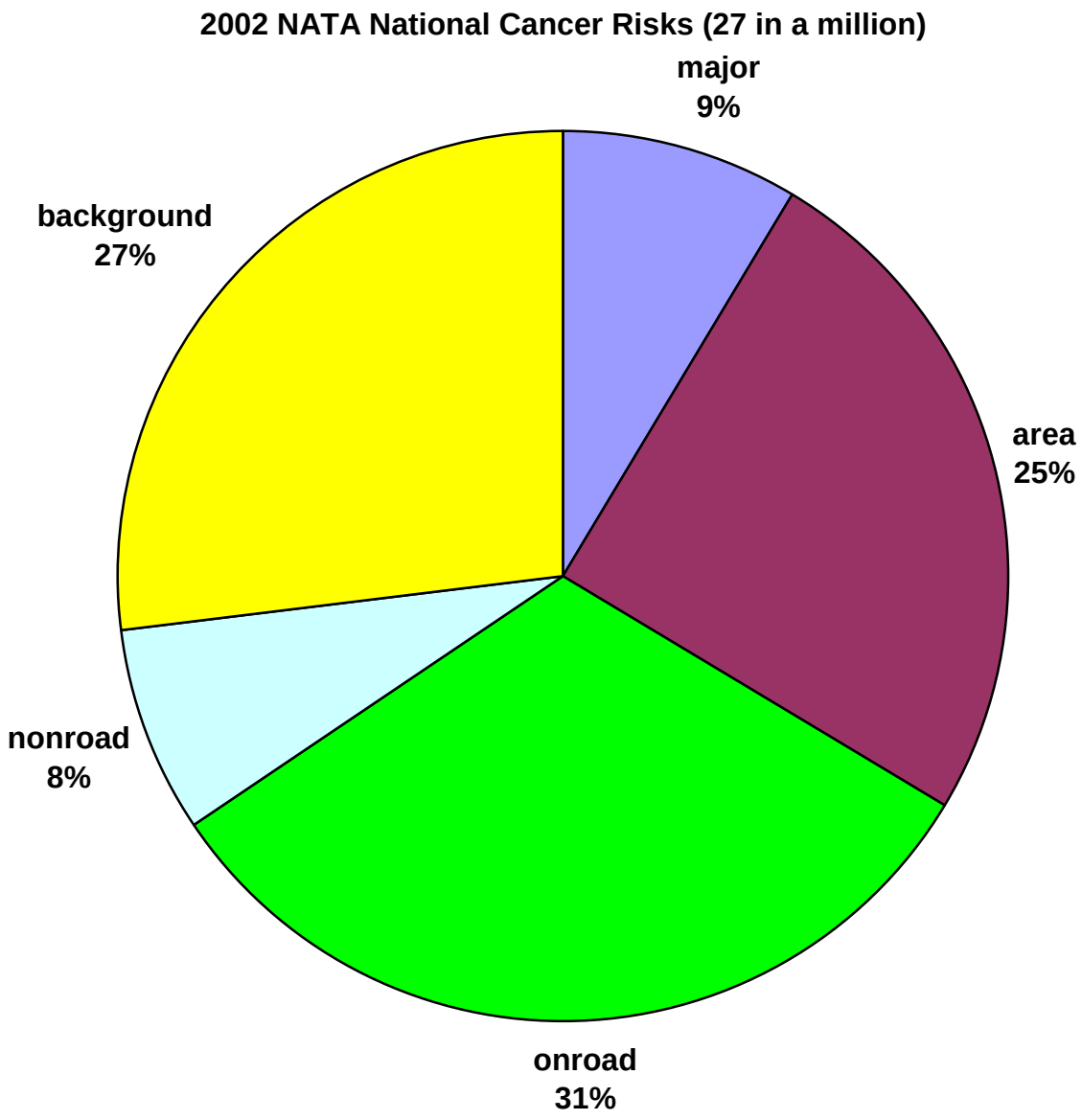
Counties with Background Estimates above Threshold Levels

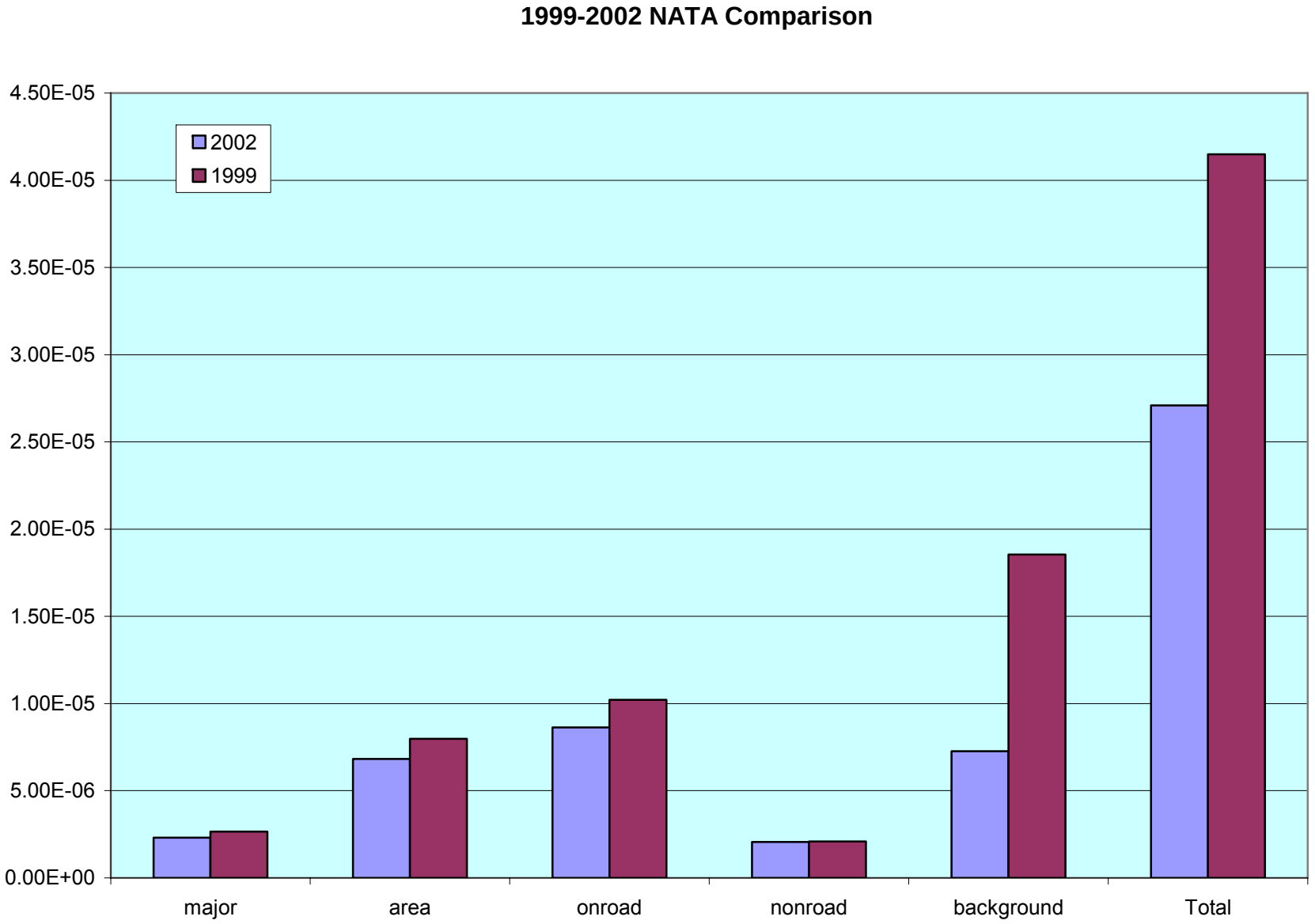


Only a few pollutants had counties with concentrations above the 10^{-6} risk-weighted concentration level.

1999 NATA National Cancer Risks (42 in a million)




2002 NATA National Cancer Risks (27 in a million)
major
9%
area
25%
onroad
31%
nonroad
8%
background
27%


1999-2002 NATA Comparison
4.50E-05
4.00E-05
3.50E-05
3.00E-05
2.50E-05
2.00E-05
1.50E-05
1.00E-05
5.00E-06
0.00E+00
2002
1999
major
area
onroad
nonroad
background
Total

Conclusions: Benefits and Limitations

- Benefits
  - This approach provides better spatial estimates of background concentrations than using ambient measurements alone
  - Background concentrations better match the NATA definition of background than a flat (i.e., non-spatially varying) background
  - This approach was relatively easily implemented
- Limitations
  - This approach may double-count emissions in counties with emissions
  - No wind or terrain adjustments for the dispersion of concentrations
  - Upper-end background concentrations are somewhat arbitrary
  - Problems with the emissions inventory may be multiplied using this approach

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