Workshop on Signatures of Man-Made Isotope Production (WOSMIP)
December 4-8, 2023 Santiago, Chile

wosmip

Top-down estimates of daily global emissions of anthropogenic radioxenon during 2013-2022

LAWRENCE LIVERMORE NATIONAL LABORATORY

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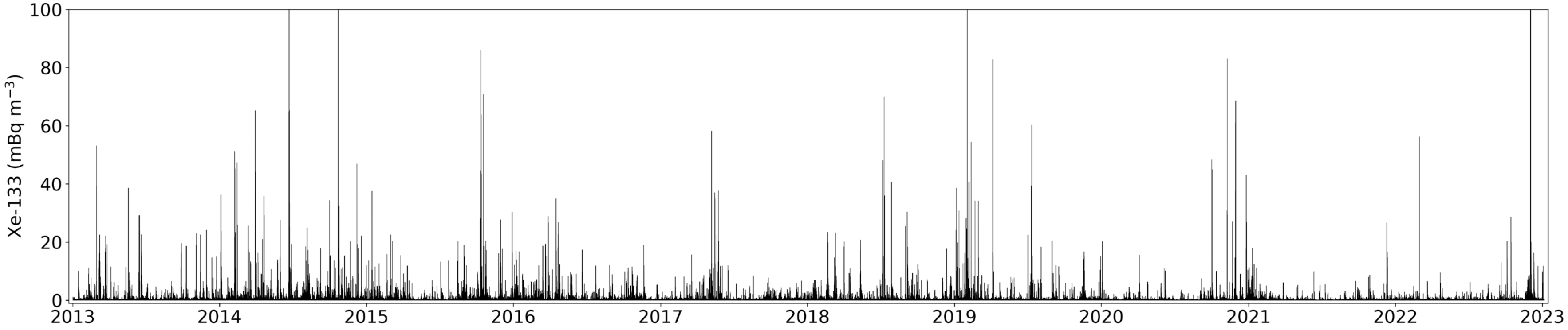
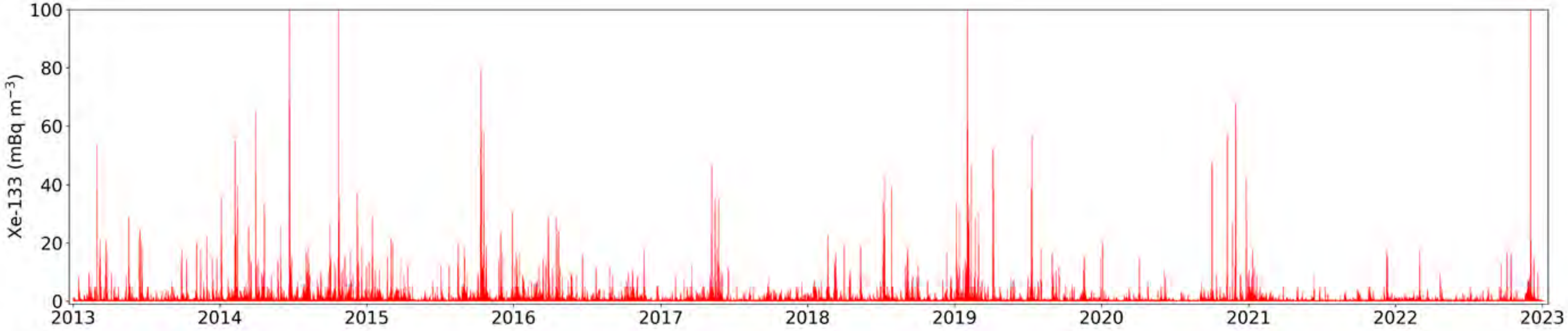
New Method for Estimating Xe-133 Emissions and Reconstructing Xe-133 concentrations

Outline

- Bottom-up versus top-down approaches.
- New 3-D global radioxenon transport model.
- Machine learning to infer daily Xe-133 emissions.
- Results
    - Concentrations
    - Emissions

Time series of Xe-133 concentrations at all IMS stations over 2013-2022





One of these time series is from our new method and other from measurements.

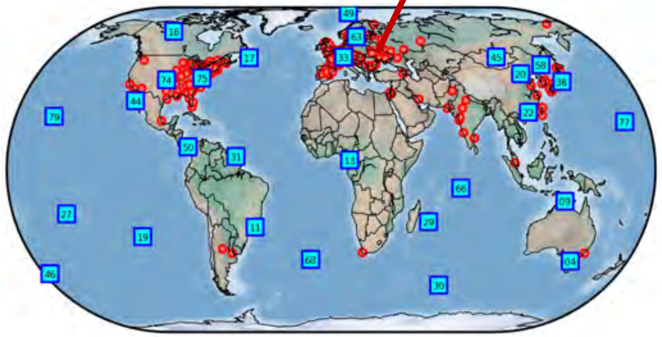
Bottom-Up Versus Top-Down

Top-Down Estimates

- ATM simulations with prior or unit emissions
- Infer emissions using inverse methods
- Simulations are optimized to match measurements

Top-down methods are used to monitor emissions of greenhouse gases and Montreal Protocol gases

Radioxenon Emissions



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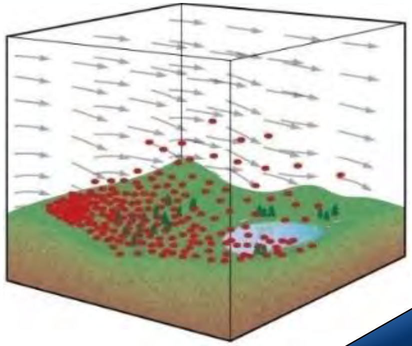
Atmospheric Transport Model



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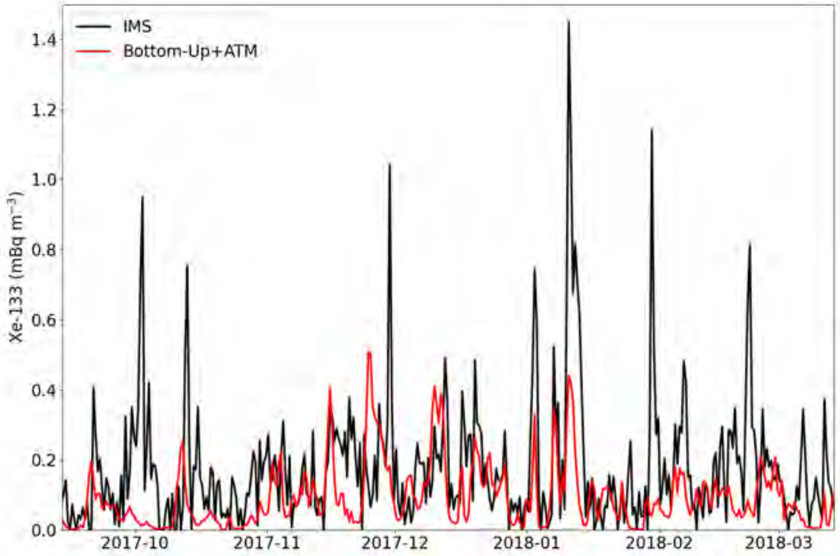
Top-Down

Bottom-Up

Simulations versus Measurements



Bottom-Up Estimates

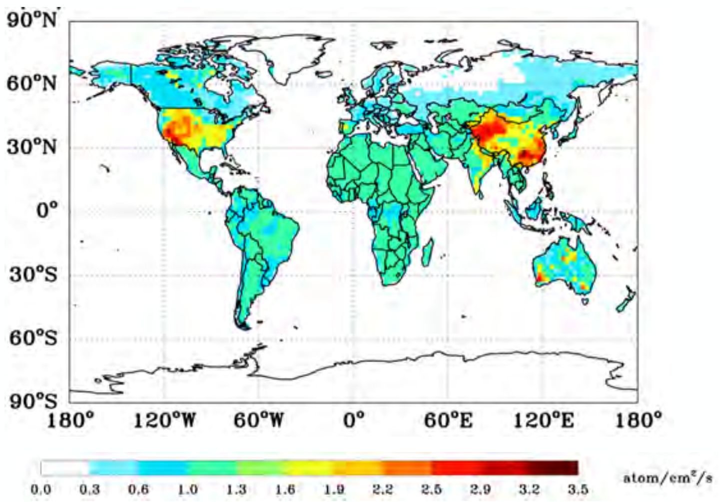
- Define emissions from inventories, stack monitors, or expert information
- ATM simulations with defined emissions
- Compare simulations and measurements

New 3-D Global Atmospheric Transport Model of Radioxenon

GEOS-Chem NASA GMAO MERRA-2

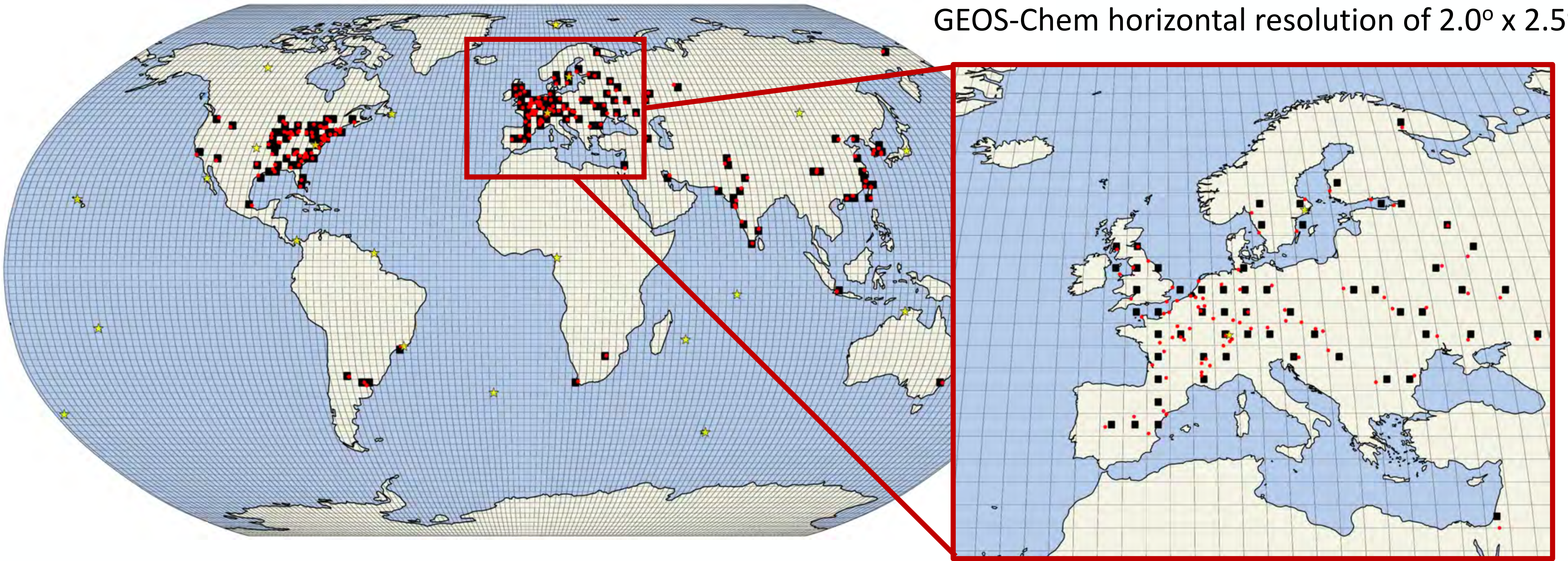
- Developed, used, and validated for 25+ years to simulate atmospheric composition on local to global scales.
- Off-line Eulerian 3-D chemical transport model driven by assimilated meteorological observations from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling Assimilation Office (GMAO).
- Contains comprehensive set of chemical and physical processes (advection, convection, boundary layer mixing, photolysis, wet and dry deposition, chemical production and loss, et cetera).
- NASA's MERRA-2 reanalysis meteorological fields (0.25°, 0.5°, or 2.0° horizontal resolution, 72 vertical levels, 3-hourly).
- Recent updates to a radionuclide tracer benchmark suite for Rn-222, Pb-210 and Be-10.

Simulation of radon-222 with the GEOS-Chem global model: emissions, seasonality, and convective transport, Zhang et al., Atmos. Chem. Phys. (2021)



Used in NASA's GEOS Composition Forecasting system

Daily Xe-133 Emissions from 144 Locations are Tagged and Tracked in GEOS-Chem over 2013-2022



GEOS-Chem horizontal resolution of 2.0° x 2.5°

● = nuclear power plant, research reactor, or medical isotope production facility (218 sources)

■ = location tracked in GEOS-Chem (144 locations)

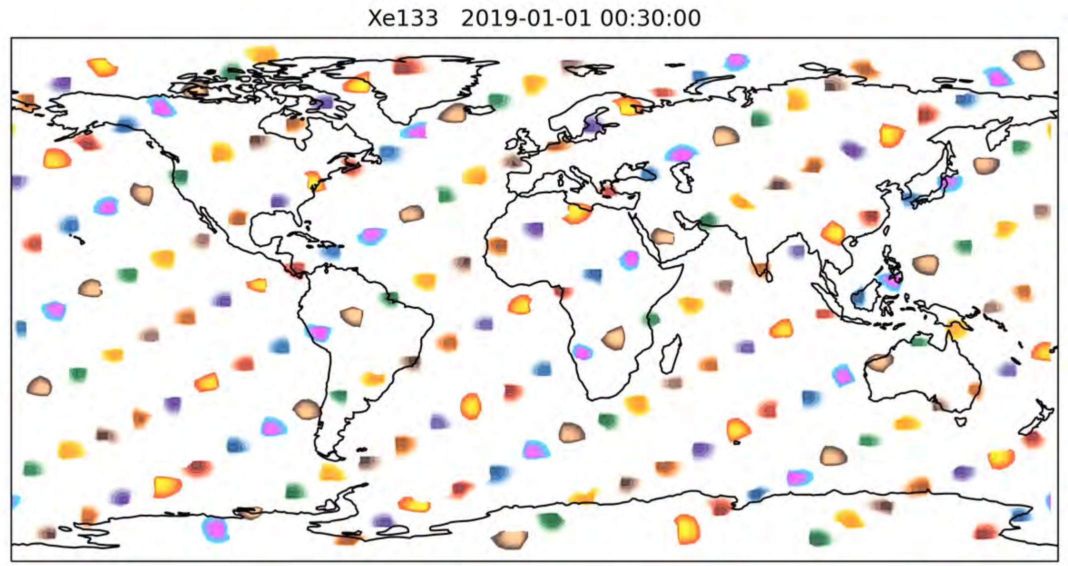
☆ = IMS stations used in top-down estimates (23 stations)

GEOS-Chem resolution consistent with IMS collection times

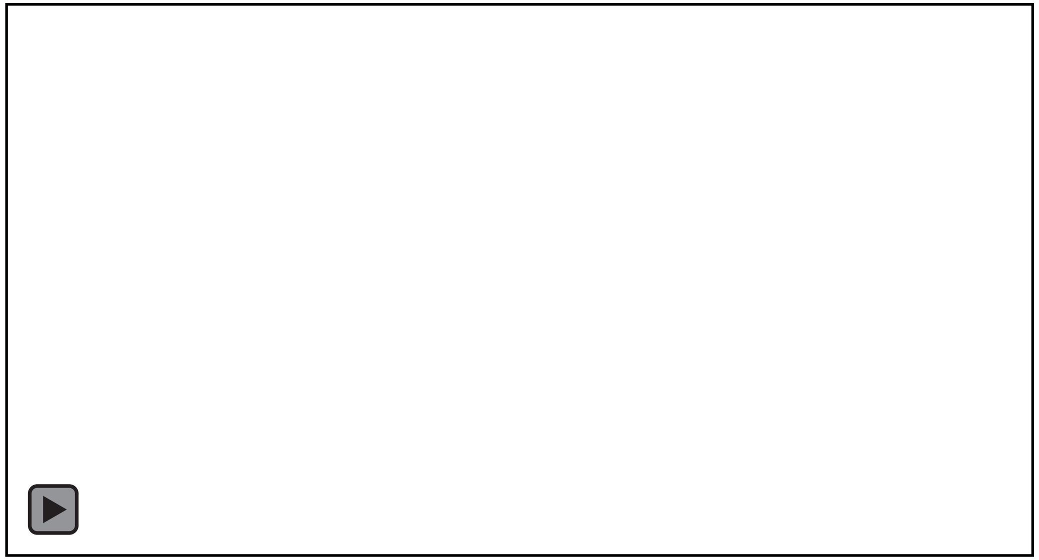
- 2° is about 100 km
- Air traveling at 5 m/s for 12 hours = 216 km

Daily Xe-133 Emissions from 144 Locations are Tagged and Tracked in GEOS-Chem over 2013-2022

- Total of 527,760 Xe-133 daily emission species are tagged in GEOS-Chem
    - 144 emission locations x 3,665 days (10 years)
- Each tagged species is simulated for 14 days.
    - Timesteps: 5-min transport, 10-min emissions and decay
- Concentrations stored hourly and processed for the IMS collections over 2013-2022.



Example of 200 tagged Xe-133 species emitted on 01 Jan 2019 and tracked for 68 hours.



Example of Xe-133 emitted from the 144 locations on 06 Jan 2014 and tracked for 336 hours.

We Start With Flat Bottom-Up Emissions Informed From Published Inventories

- Our bottom-up estimates are informed from Kalinowski et al. inventories.
- Time invariant bottom-up emissions are assumed (i.e., “flat”).
  - Validate top-down method vs known temporal changes.
  - Provides a test for *sparseness*.

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Pure and Applied Geophysics

Global Radioxenon Emission Inventory from Nuclear Power Plants for the Calendar Year 2014

Martin B. Kalinowski[1] and Halit Tatlisu[1]

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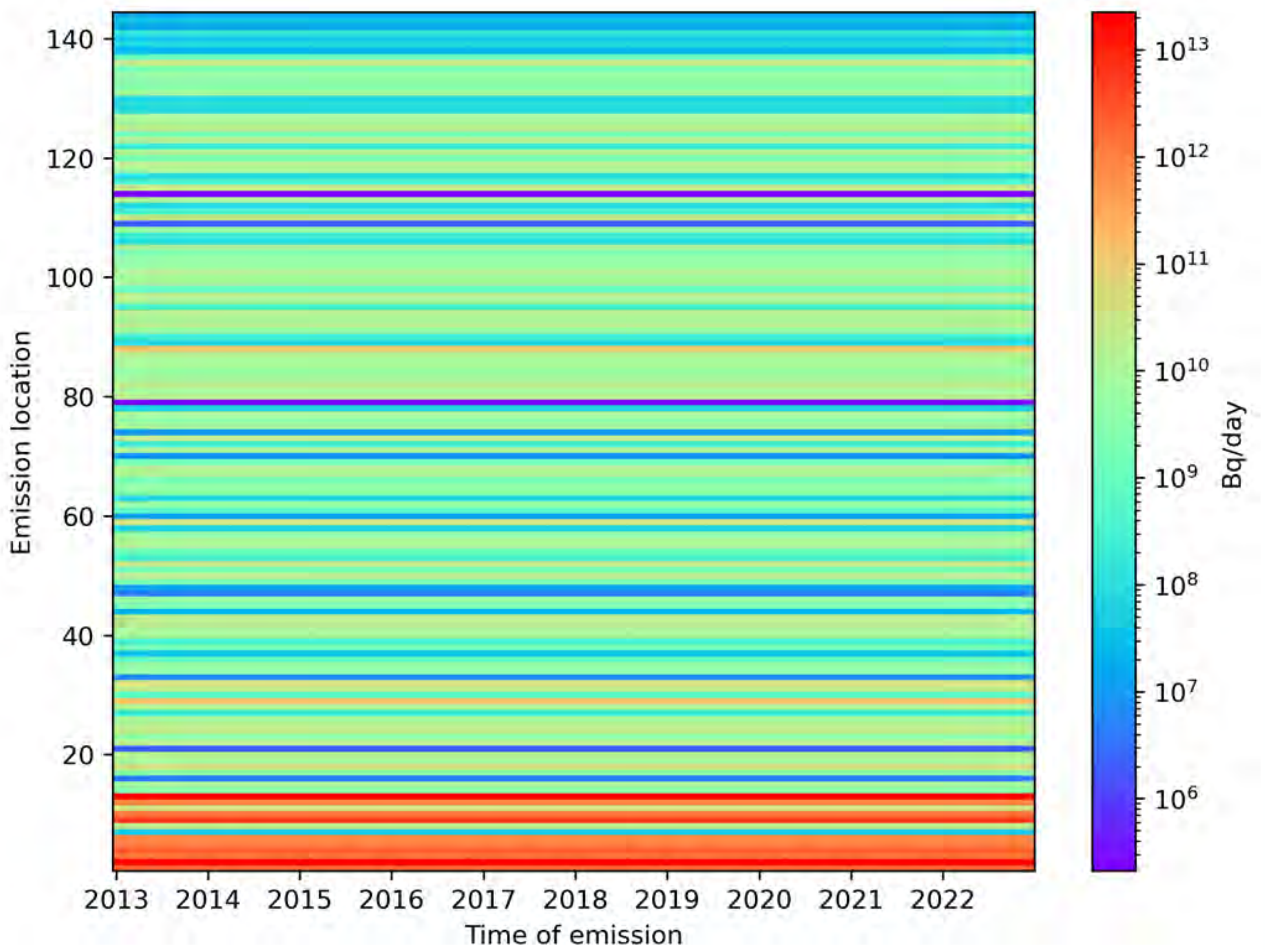
https://doi.org/10.1007/s00024-021-02719-w

Pure and Applied Geophysics

Global Radioxenon Emission Inventory from Nuclear Research Reactors

Martin B. Kalinowski,[1] Pouneh Tayyebi,[2] Michael Lechermann,[3] and Halit Tatlisu[1]

144 locations x 3665 days = 527,760 emission parameters



Large Matrix is Inverted for Top-Down 2013-2022 Emission Estimates

Xe-133 Observation Target Vector

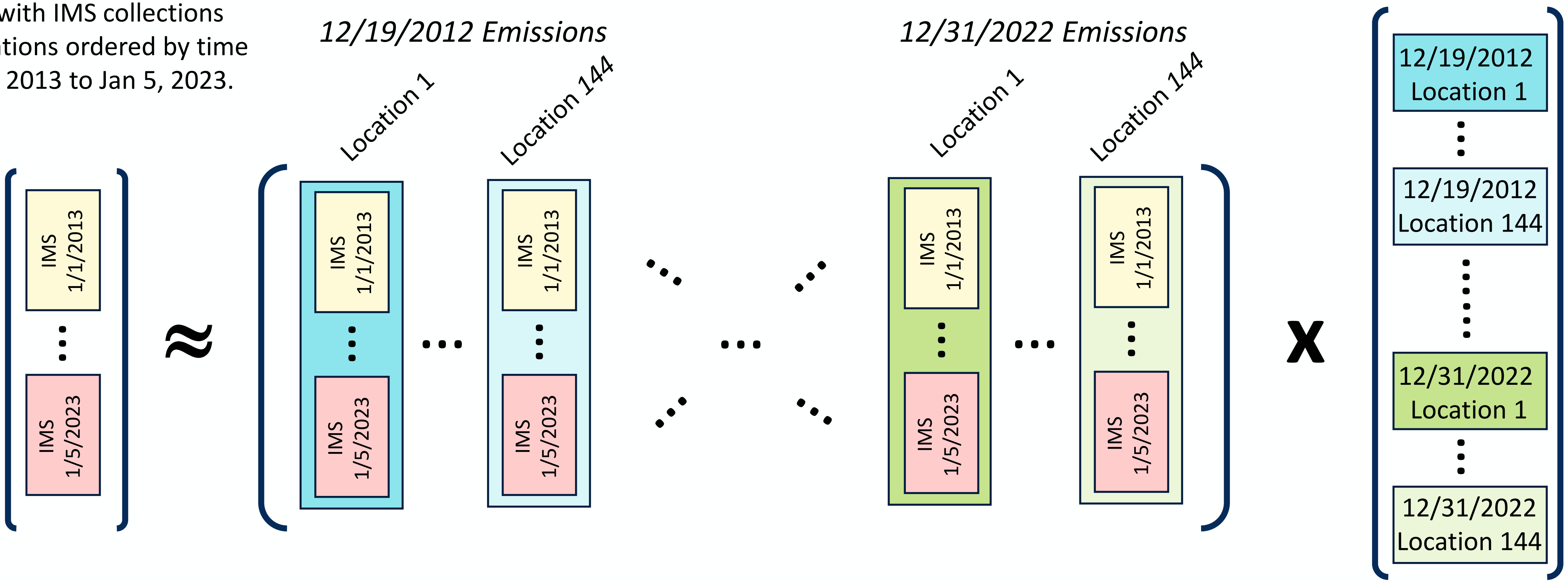
Populated with IMS collections from 23 stations ordered by time from Jan 1, 2013 to Jan 5, 2023.

GEOS-Chem Design Matrix

Populated with tagged GEOS-Chem simulations using flat, bottom-up emissions.

Emission Weight Vector

To be inferred



118,012 rows x 1 column

118,012 rows x 527,760 columns

527,760 rows x 1 column

Large Matrix is Inverted for Top-Down 2013-2022 Emission Estimates

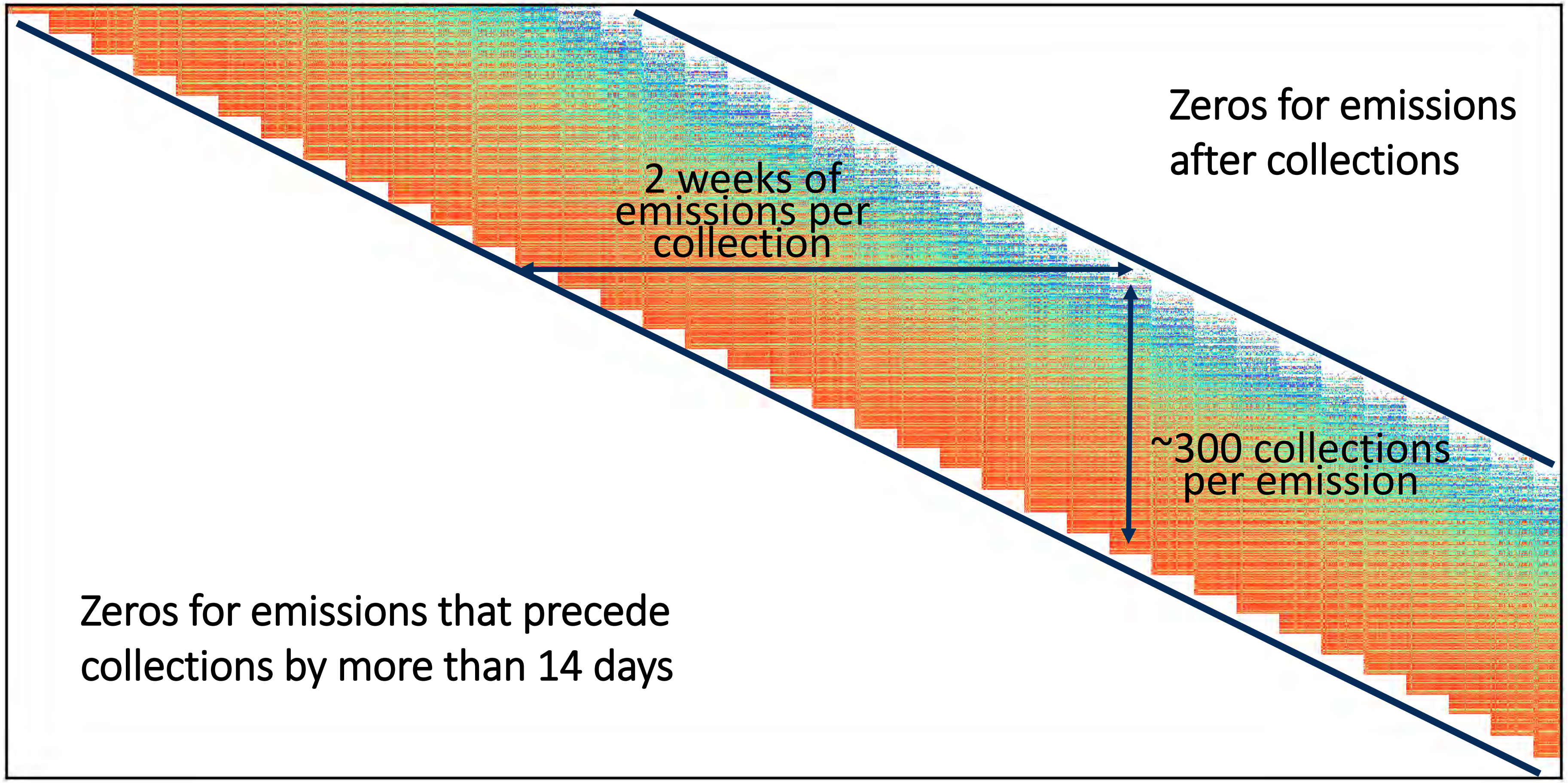
Portion of actual design matrix

- 850 rows of IMS collections (~1 month)
- 5277 columns of Xe-133 from daily emissions

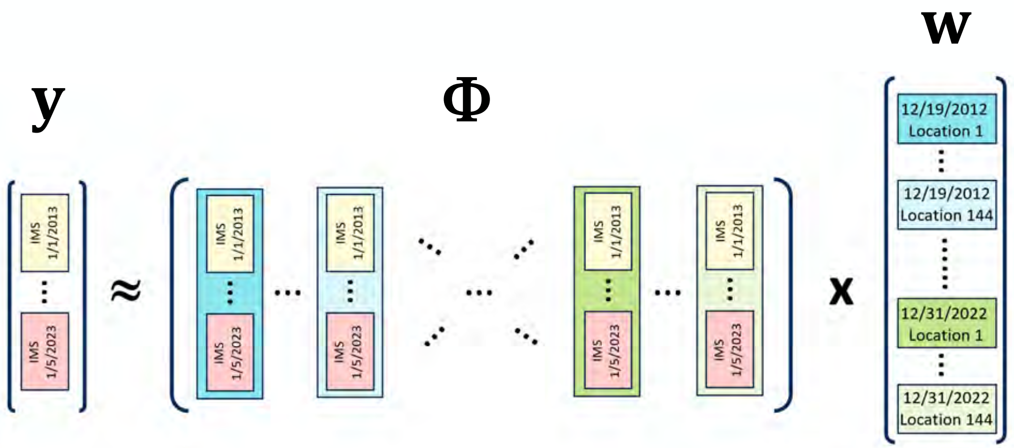
If dense, the full design matrix would require ~250 GB to store.

By exploiting the sparse structure, the design matrix requires only 1.7 GB using sparse matrix packages!

GEOS-Chem Design Matrix is Highly Sparse



We Solve the Xe-133 Emissions Inverse Problem Using Compressive Sensing

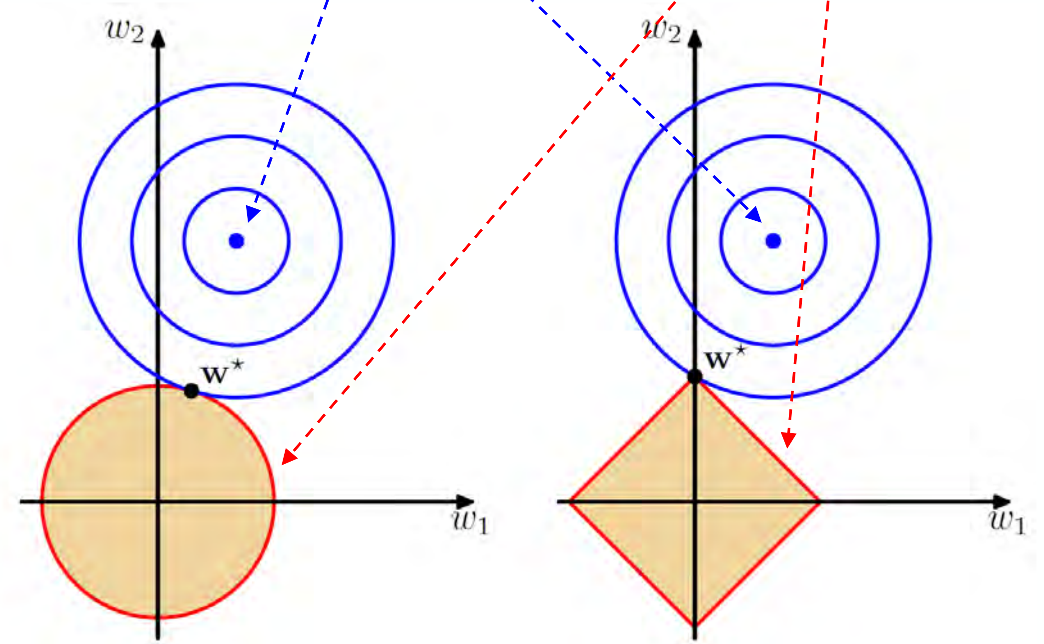


- Compressive sensing reconstructs signals in underdetermined systems. (aka lasso)
- Mathematically proven in famous 2006 paper [1].
- Two key assumptions
  - *Sparsity* [few non-zero weights in **w**]
  - *Incoherence* [weak correlation between **w** and any single row in **Φ**]

[1] Candès et al, "Robust Uncertainty Principles: Exact Signal Reconstruction From Highly Incomplete Frequency Information," *IEEE Trans Inf Theory* (2006)

Regularized Least Squares

$$||\mathbf{y} - \mathbf{\Phi}\,\mathbf{w}||_{L_2}^2 + \alpha||\mathbf{w}||_{L_x}$$

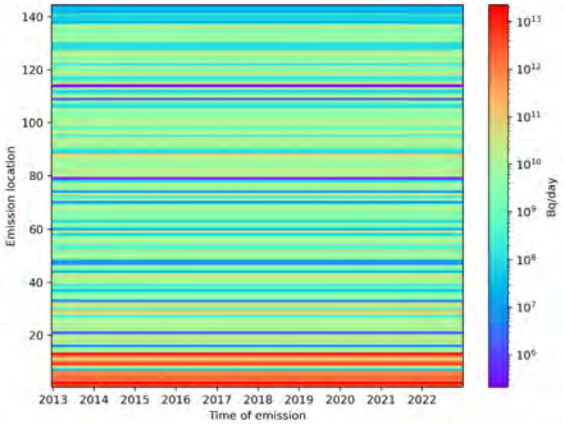


L_2 constraint provides **dense** solutions

L_1 constraint provides **sparse** solutions

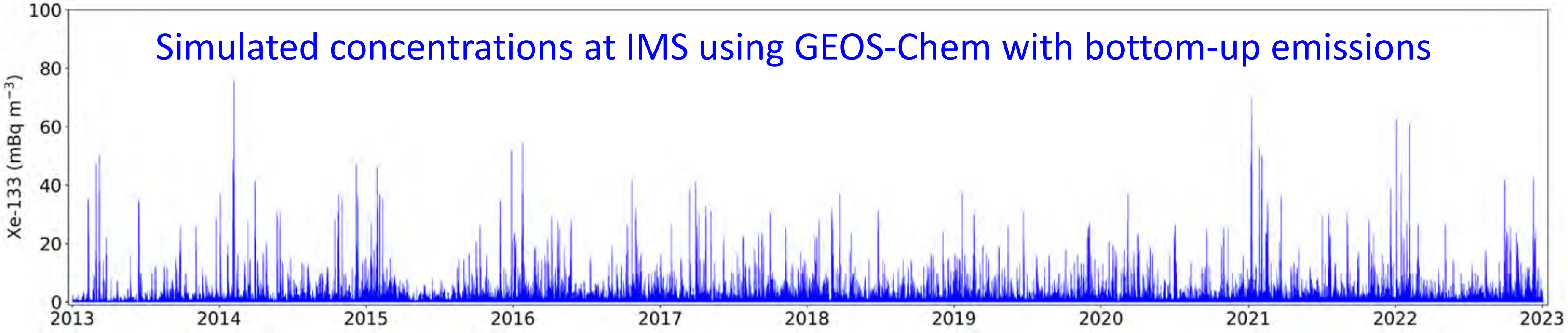
Evidence for Sparsity of Daily Xe-133 Emissions

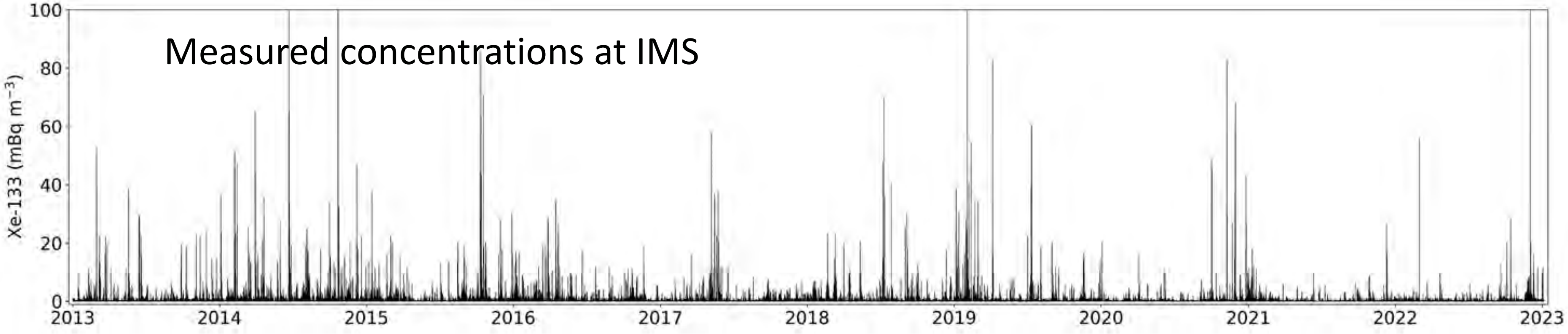
Flat bottom-up emissions



144 x 3665 = 527,760

Not every location is actively emitting on every day!

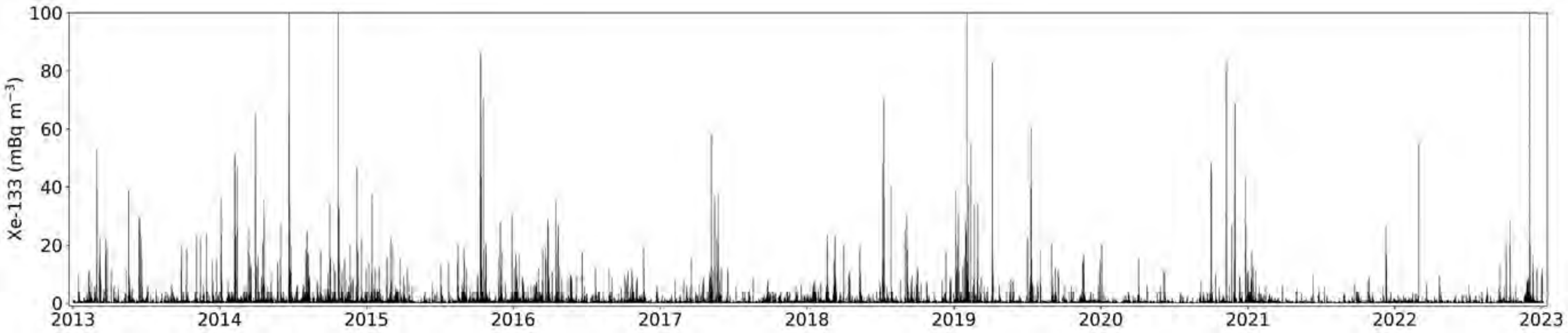




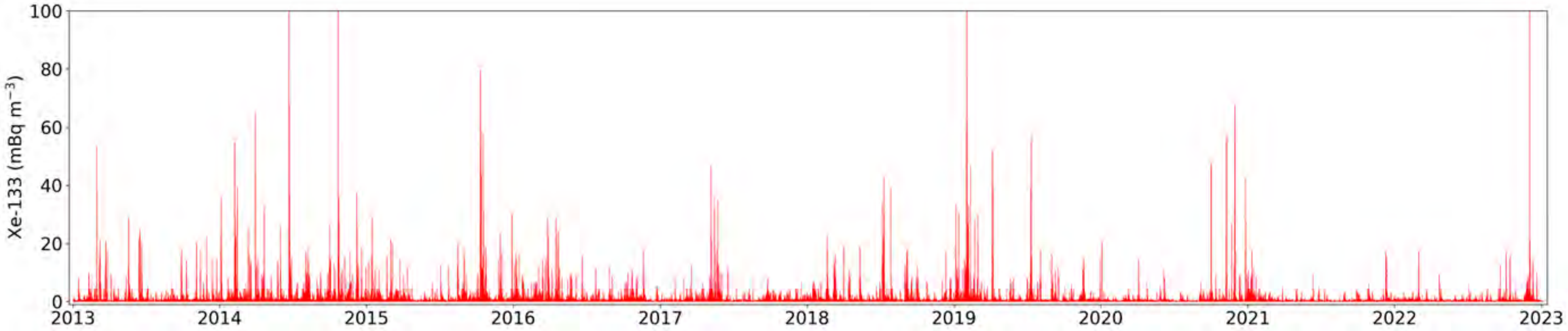
- Bottom-up concentrations contain more peaks and structure than IMS.
- We can use compressive sensing to determine daily emissions.

Top-Down Emissions Improve Simulations of IMS Measurements

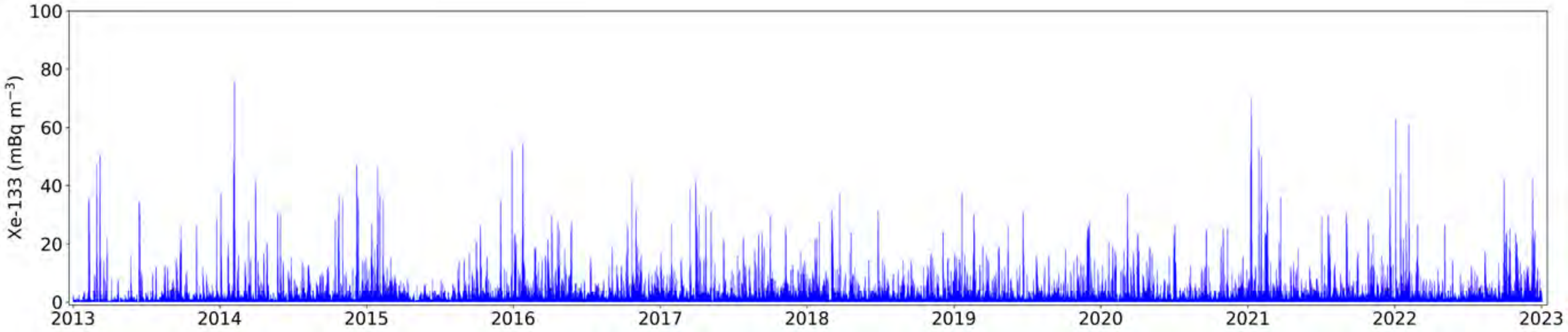
IMS measurements of Xe-133 at all stations during 2013-2022



GEOS-Chem with top-down emissions



GEOS-Chem with flat bottom-up emissions

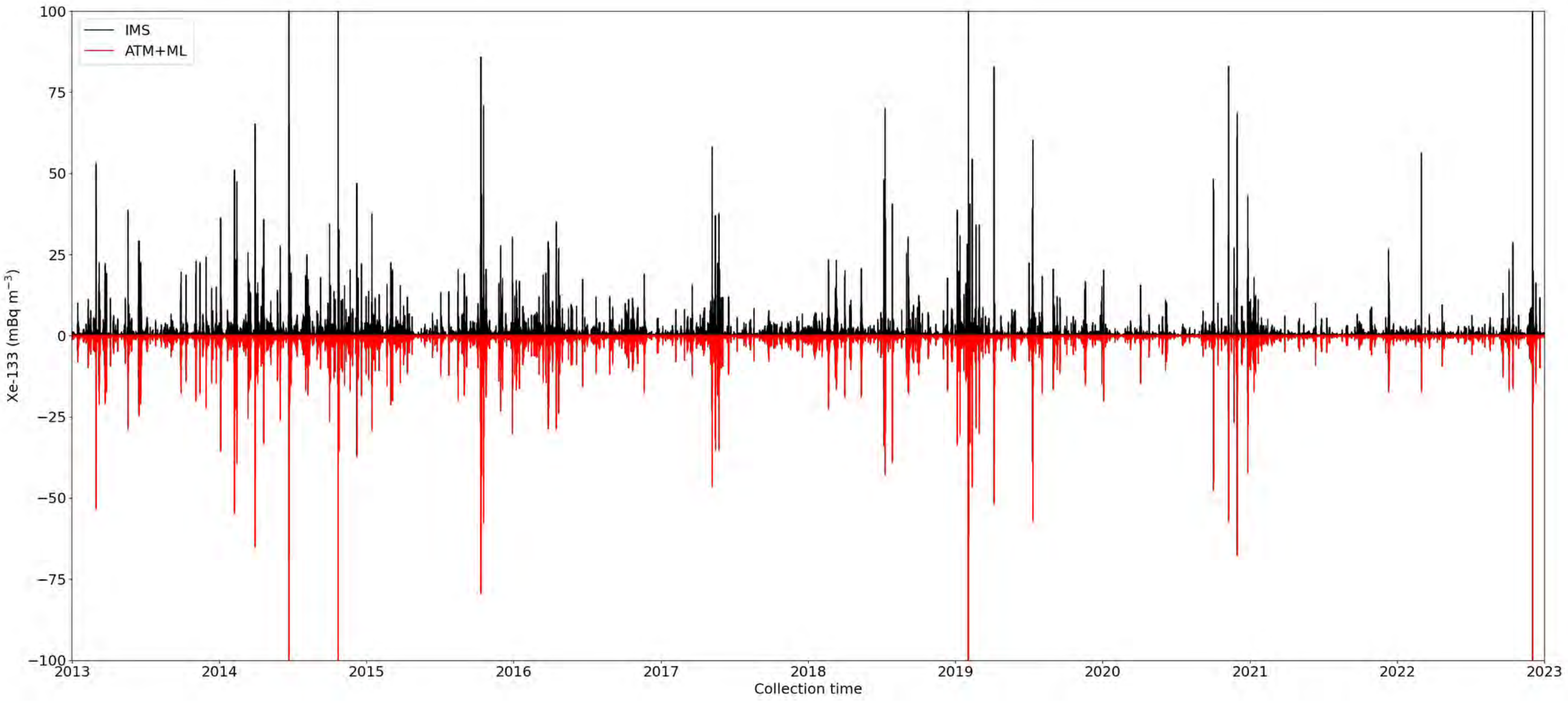


Top-Down Emissions Improve Simulations of IMS Measurements (at many scales)

Simulations also improved for small magnitude concentrations

IMS measurements
(all stations)

GEOS-Chem with
Top-Down Emissions



View 1
10 years

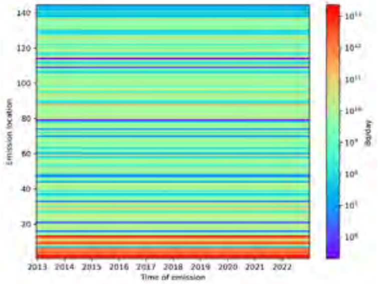
View 2
1 year

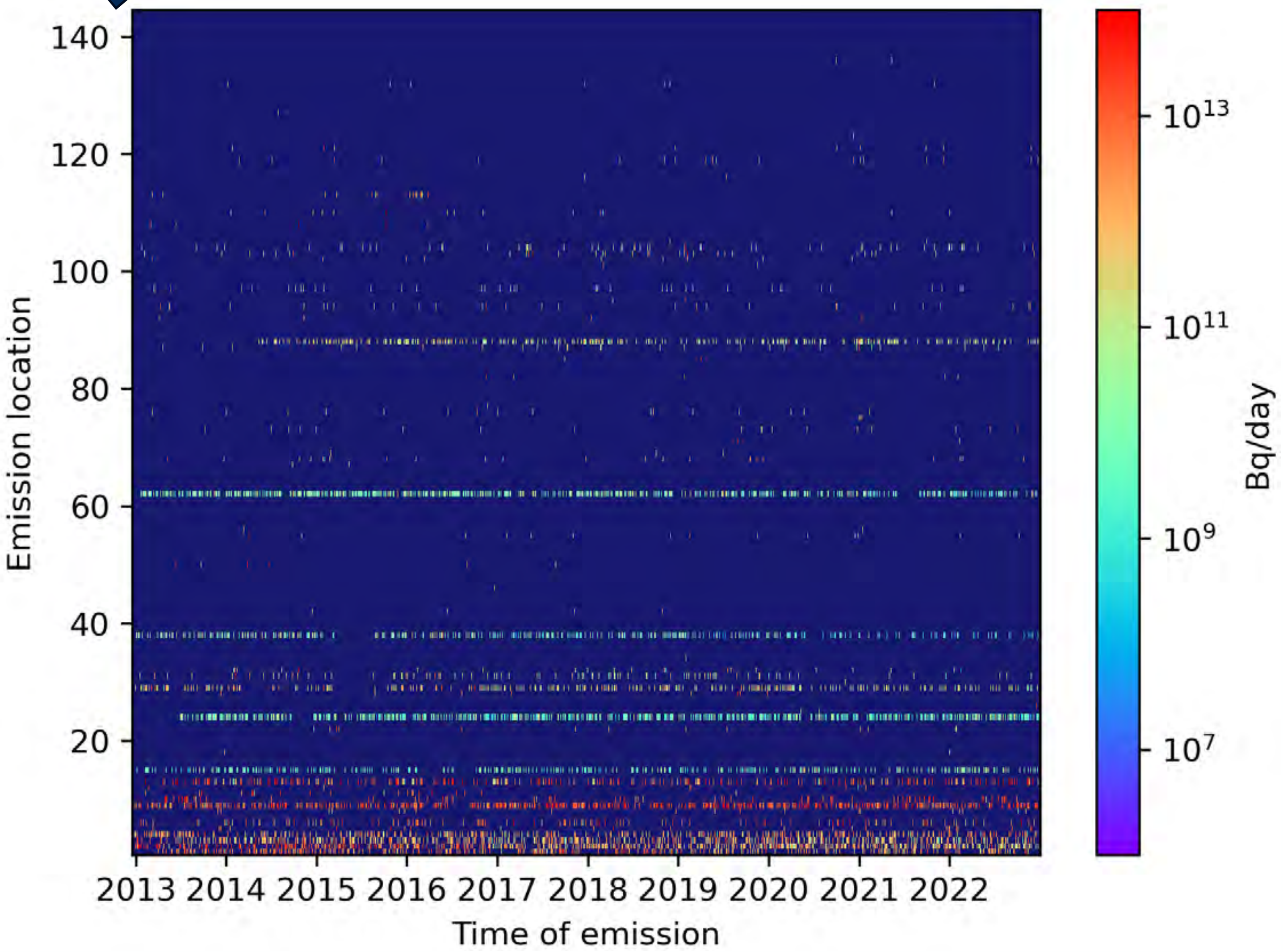
View 3
2 months

Top-Down Daily Xe-133 Emission Estimates

- Relatively few locations exhibit regular releases over 2013-2022.
  - e.g., large isotope production facilities
- Many locations exhibit intermittent or sporadic releases.
- Majority of location-day emissions are reduced to zero.
  - Solution on the right has 15,219 non-zero emissions (out of 527,760).
- Top-down estimates compare favorably with known trends and stack values.
  - e.g., Chalk River after 2017
- Uncertainty is being estimated to assess uniqueness.

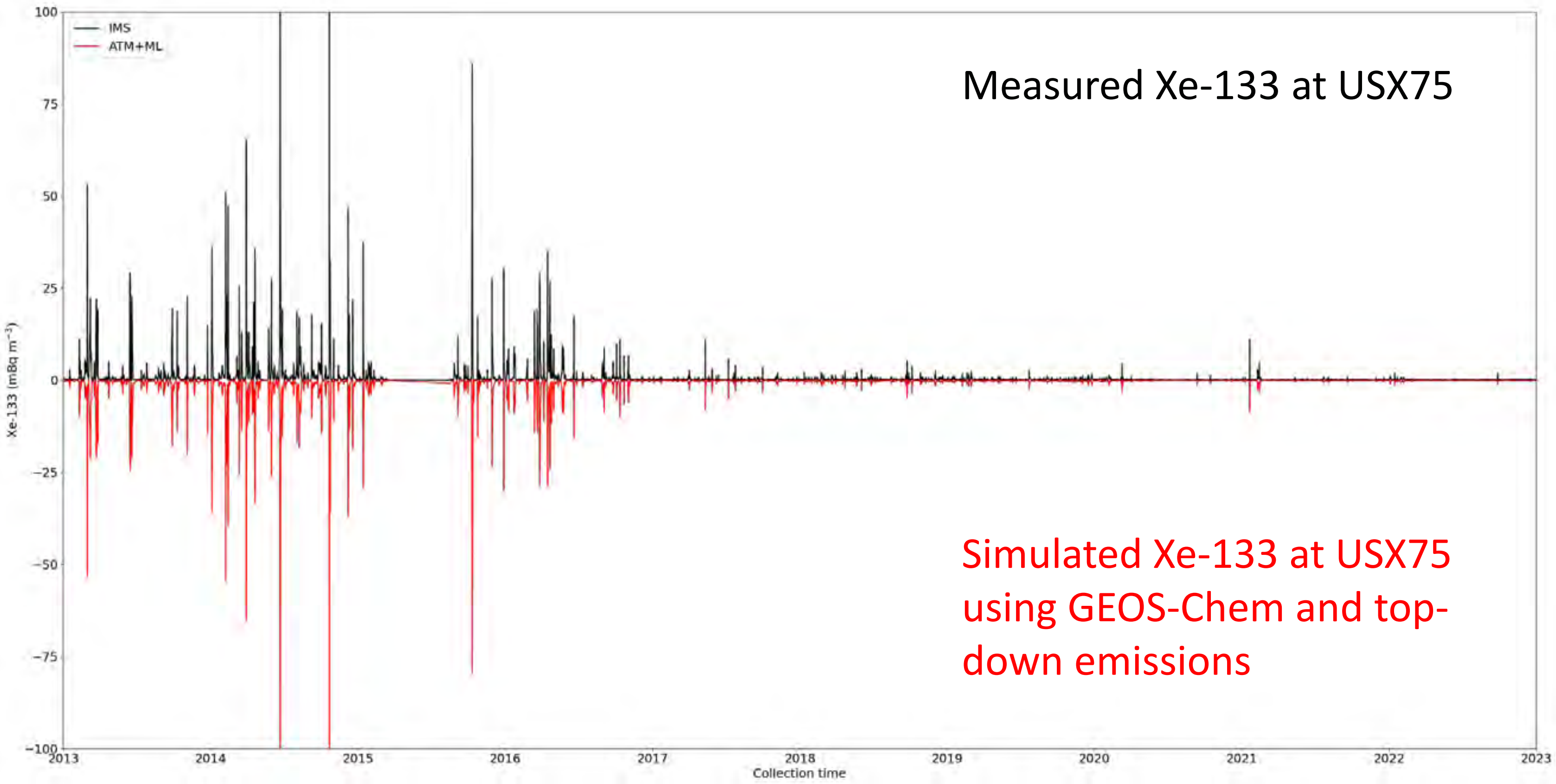
Flat bottom-up emissions

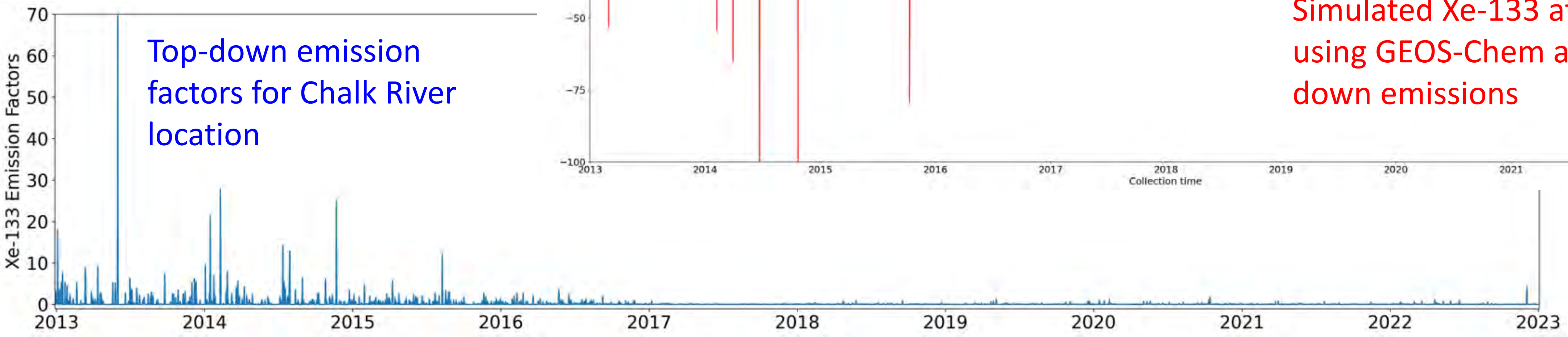




Top-Down Estimates Available for Individual Emission Locations and IMS Stations

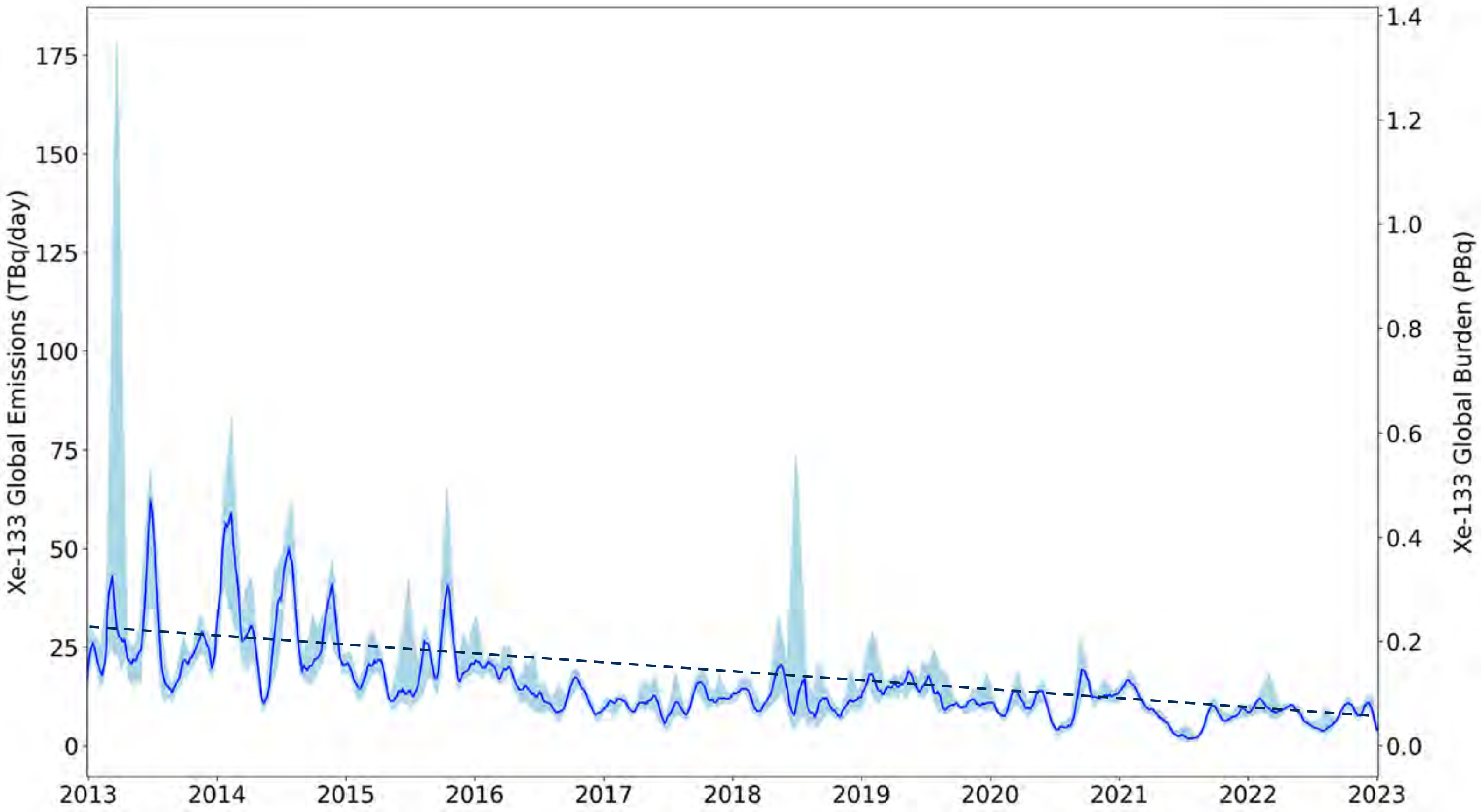
- Bottom-up estimates assume constant emissions from Chalk River over 2013-2022.
- Top-down method infers that emissions were reduced after 2017.





We Detect a Downward Trend in the Global Emissions and Burden of Xe-133 over 2013-2022

- Using our top-down estimates, we calculate the global emission rates and atmospheric burden of Xe-133 over the past decade.
- Despite increasing demand for radioisotopes and a slight increase in the number and capacity of nuclear power plants over this period, we find decreasing Xe-133 emissions over 2013-2022.
- This trend is mainly due to reduced emissions from Chalk River after 2017.



2x 30-day smoothing windows were applied to the global emissions time series to remove high frequency emission components.

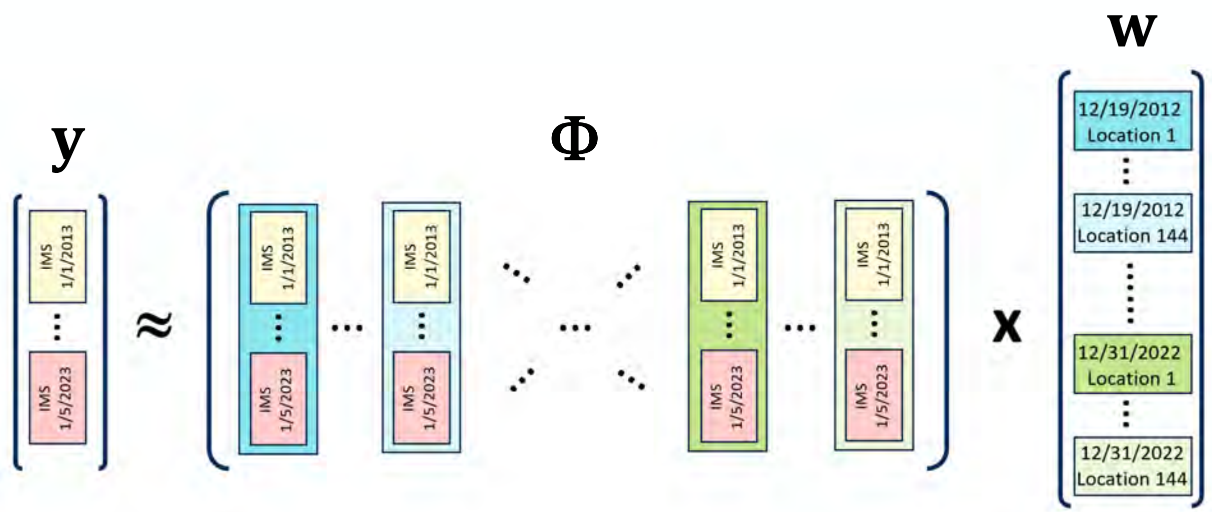
Key Conclusions and Next Steps

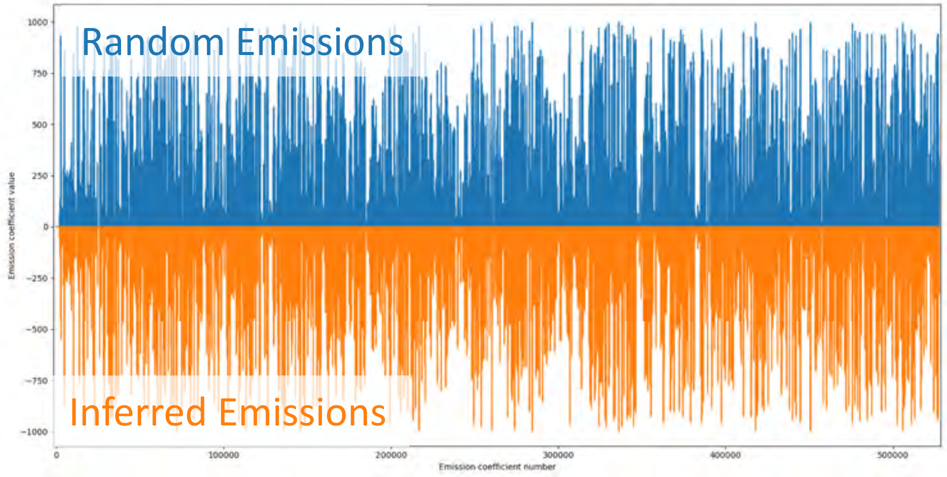
- Successful global reconstruction of Xe-133 over 2013-2022.
  - New 3-D global Eulerian xenon transport model (GEOS-Chem)
  - Sparse matrix and sparse machine learning methods (compressive sensing)
  - Considers 118,012 collections from 23 IMS stations simultaneously
- Only ~10^4 daily emission sources are needed for reconstructions over this period.
- Global emissions and burden of Xe-133 have decreased over the past decade.

- Currently adding uncertainty to assess robustness and non-uniqueness.
  (Bayesian approaches, bootstrapping, and model error)
- Continuing to evaluate our top-down estimates against independent sources.
  (stack emissions, annual or quarterly releases reports, operational status)

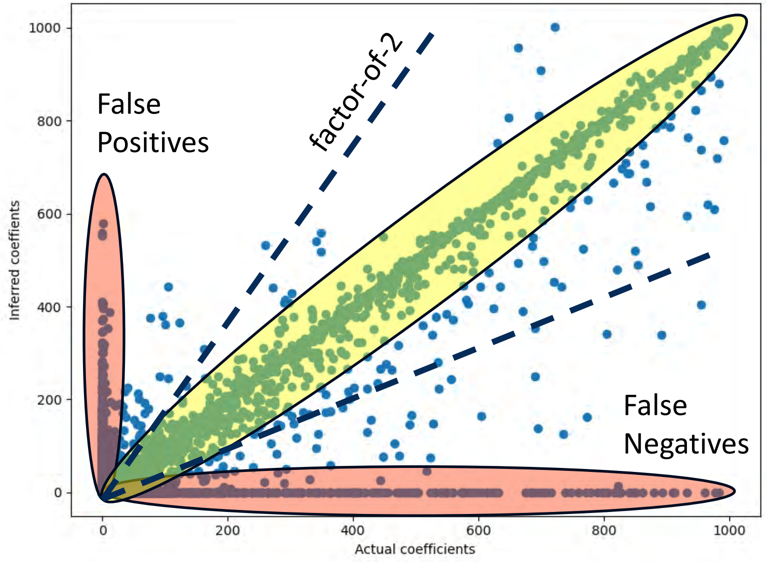
Questions?

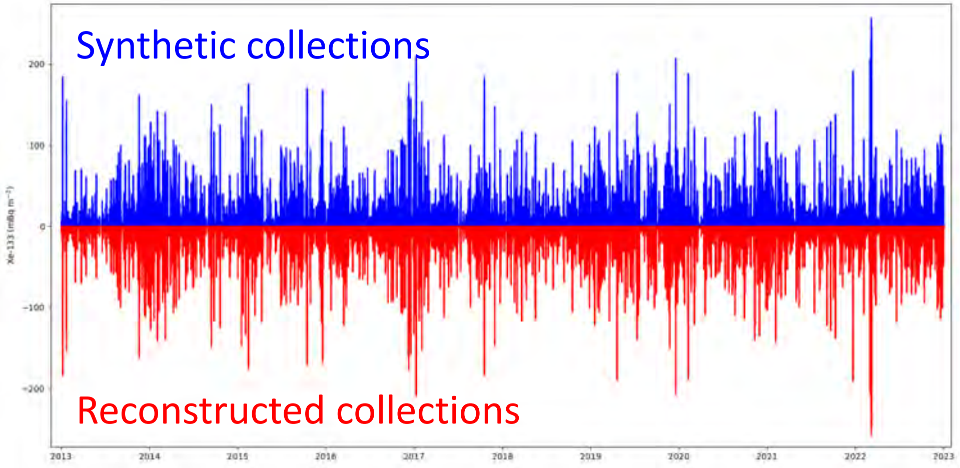
Synthetic Data Provides Confidence in Top-Down Estimates





1. Draw a small set of random weights. (17,889 emissions are used in this example)
2. Apply weights to "high sensitivity" columns in phi.
3. Compute observation vector via $\mathbf{y} = \mathbf{\Phi}\, \mathbf{w}$.
4. Use compressive sensing to infer emission weights.





Abstract

Low levels of radioxenon can be emitted to the atmosphere during routine operations and maintenance from hundreds of nuclear power plants, research reactors and medical isotope production facilities distributed worldwide. These emissions create persistent and ever-changing amounts of radioxenon in the atmosphere, the presence of which complicates efforts to detect radioxenon produced from nuclear weapons testing. Quantifying the radioxenon background requires information about facility emissions on a daily to weekly basis, but this information is not generally available. Stack monitors that directly measure facility emissions are costly and have been deployed at only a few locations. "Bottom-up" emission inventories help fill the gap by estimating emissions from statistical analysis and expert knowledge of nuclear processes and facilities (e.g., Kalinowski et al., 2009, 2021). However, bottom-up emissions are unable to adequately explain xenon measurements from the International Monitoring System (IMS) network. To reconcile these differences, we developed a new "top-down" inverse approach to estimate daily global leakage rates of xenon-133 over the period 2013-2022. This approach combines 118 thousand measurements of Xe-133 from the global IMS network, simulations of 527 thousand tagged species using a new global Eulerian xenon transport model, and machine learning techniques to determine which locations may be actively emitting on a given day. We describe our top-down method, introduce our new global xenon transport model, and show results demonstrating spatiotemporal reconstructions of the global radioxenon background.

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