Soutenance de Thèse de doctorat de l'Institut Polytechnique de Paris

Rémy Lapere

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École Polytechnique
30 septembre 2021

Observation and modeling of pollution events in Chile



Observation and modeling of pollution events in Chile

Atmospheric


Picture @JulioGoffardMolina/Flickr


Picture @BBC

Observation and modeling of pollution events in Chile

Atmospheric

Particulate matter – PM

- Aerosols (solid or liquid particles)
- Diameter <2.5µm ($PM_{2.5}$) or <10µm (PM_{10})
- Decreased visibility
- Cardiovascular and lungs diseases, asthma
- Direct and indirect radiative effects

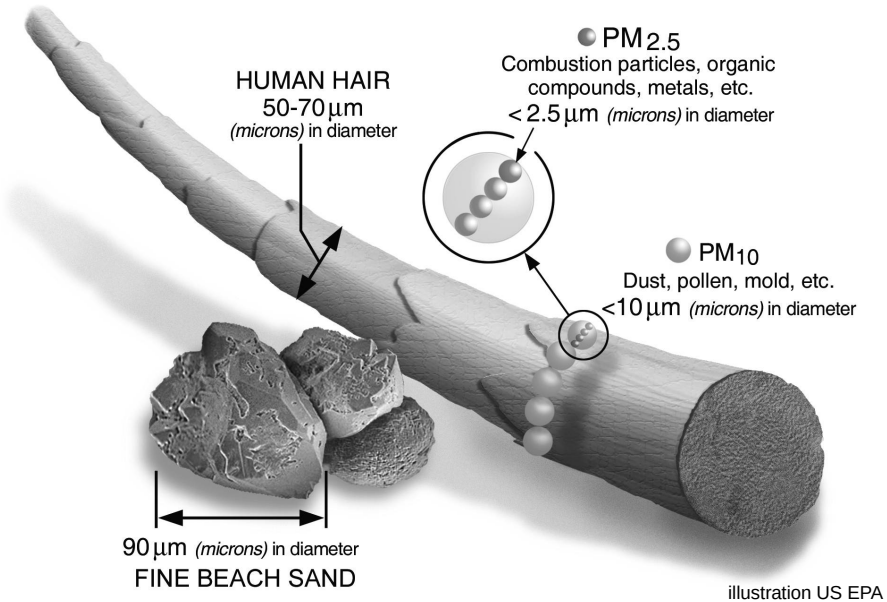

illustration US EPA

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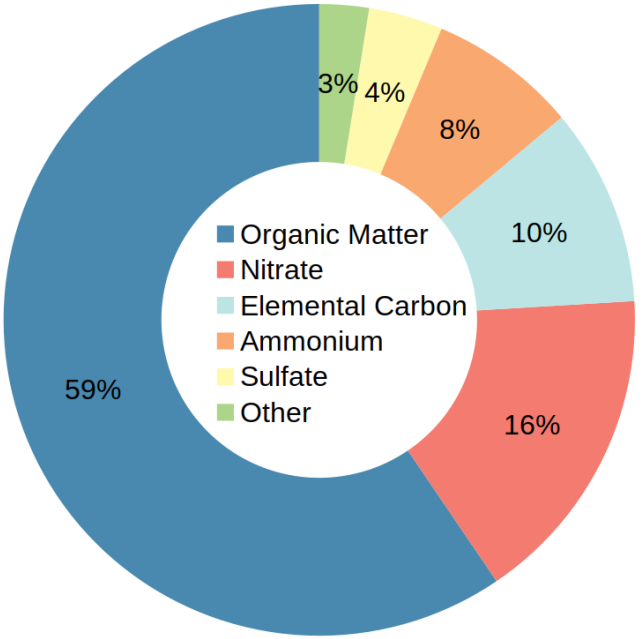

Bulk composition of PM2.5 in Santiago - June 2013.

Adapted from *Villalobos et al., 2015*

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- Secondary pollutant & greenhouse gas
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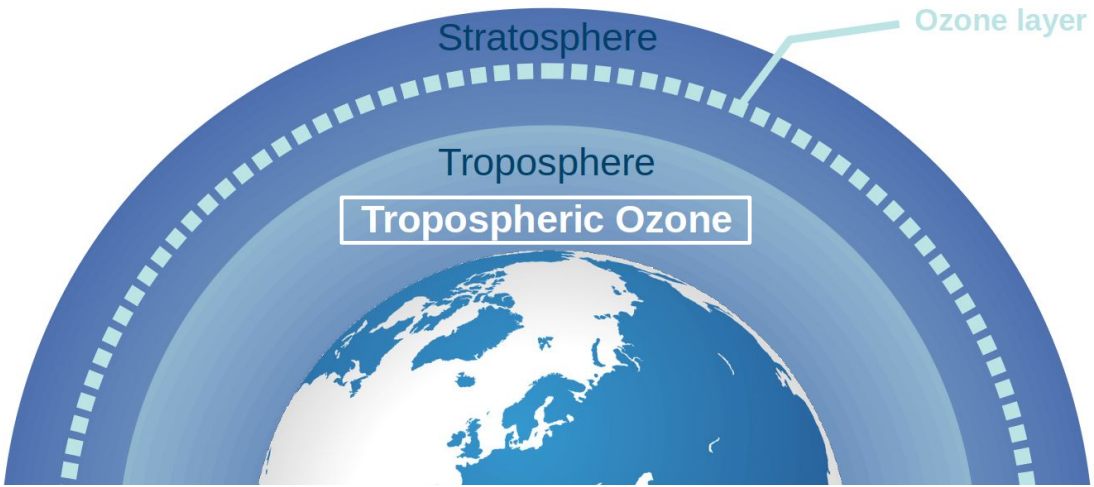

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Nitrogen oxides – NO_X

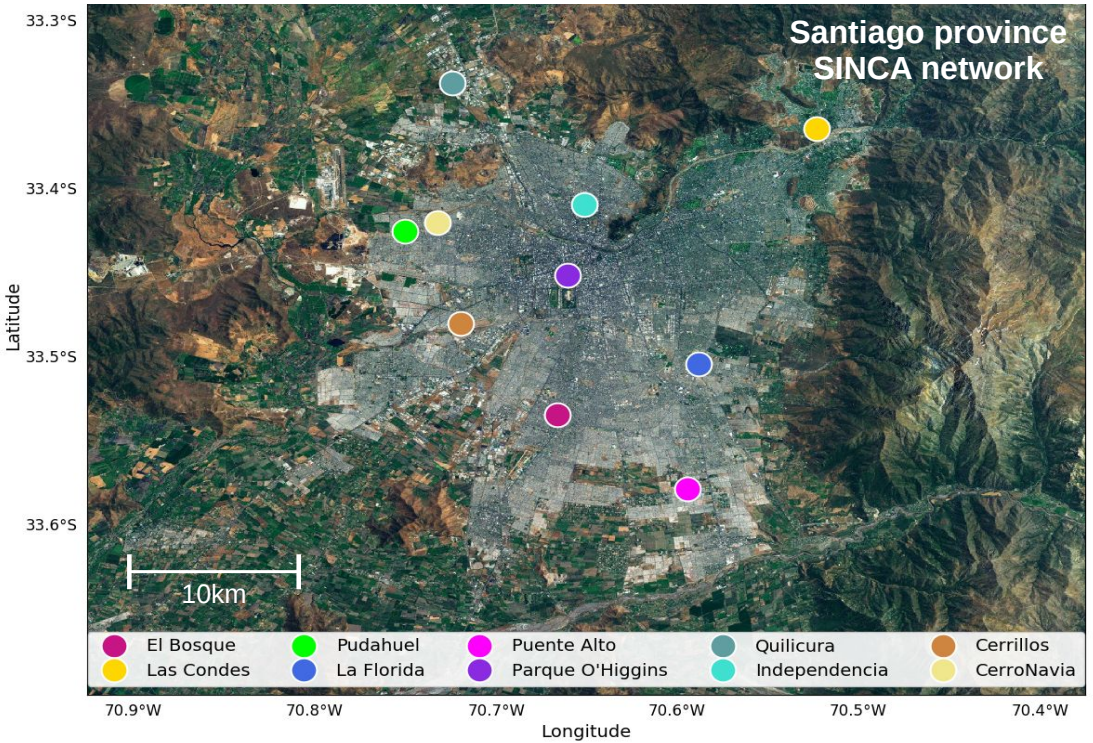
Sulfur dioxide – SO_2

Carbon monoxide – CO

Acid rains, buildings deterioration, eutrophication...

Observation and modeling of pollution events in Chile

- Local automated networks
- Satellite products (MODIS, IASI, CALIOP)
- Specific measurement campaigns



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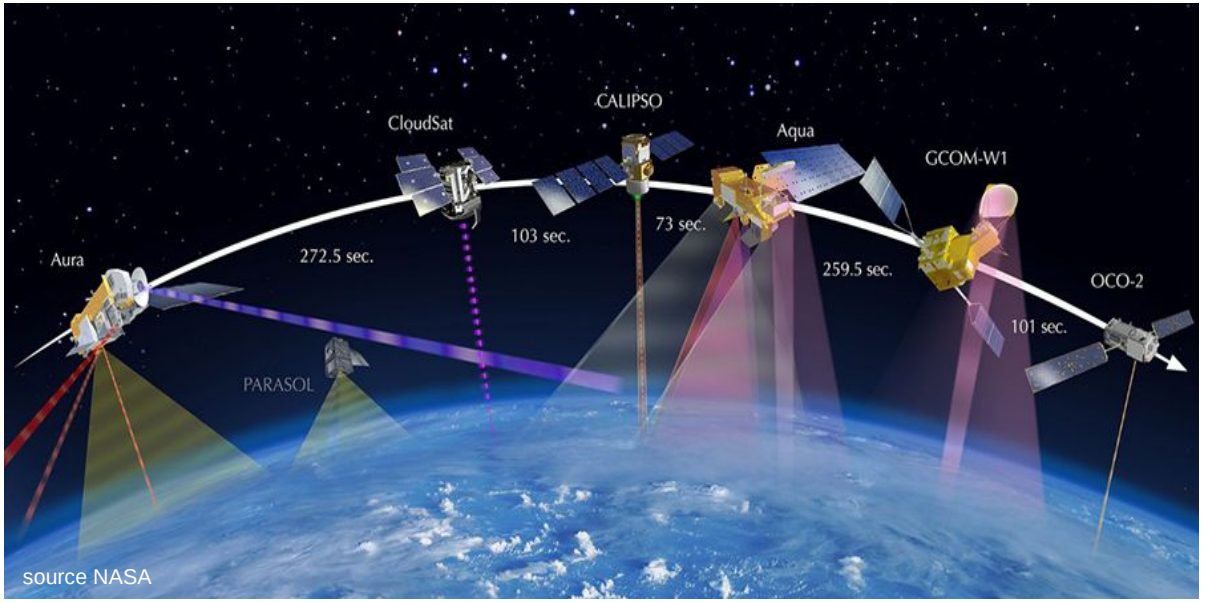

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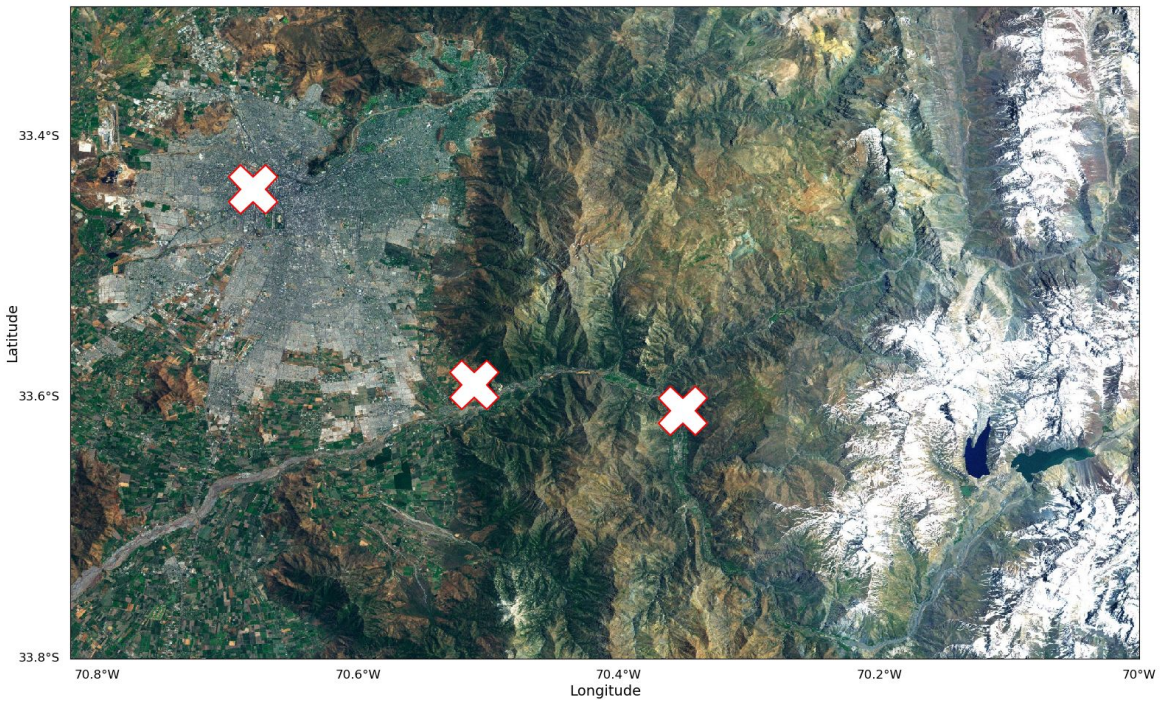


Observation and modeling of pollution events in Chile



image Google Earth

Observation and modeling of pollution events in Chile

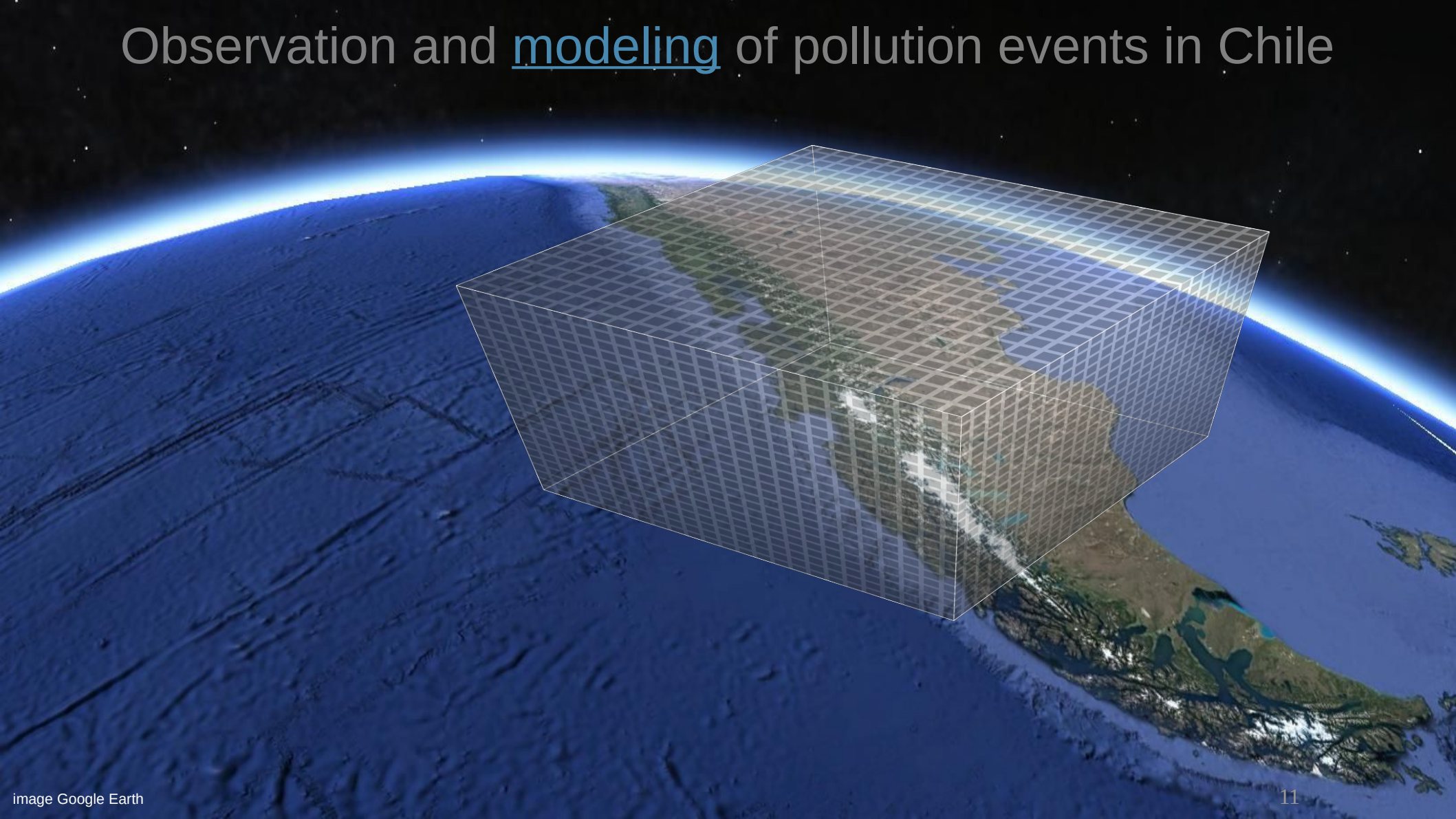
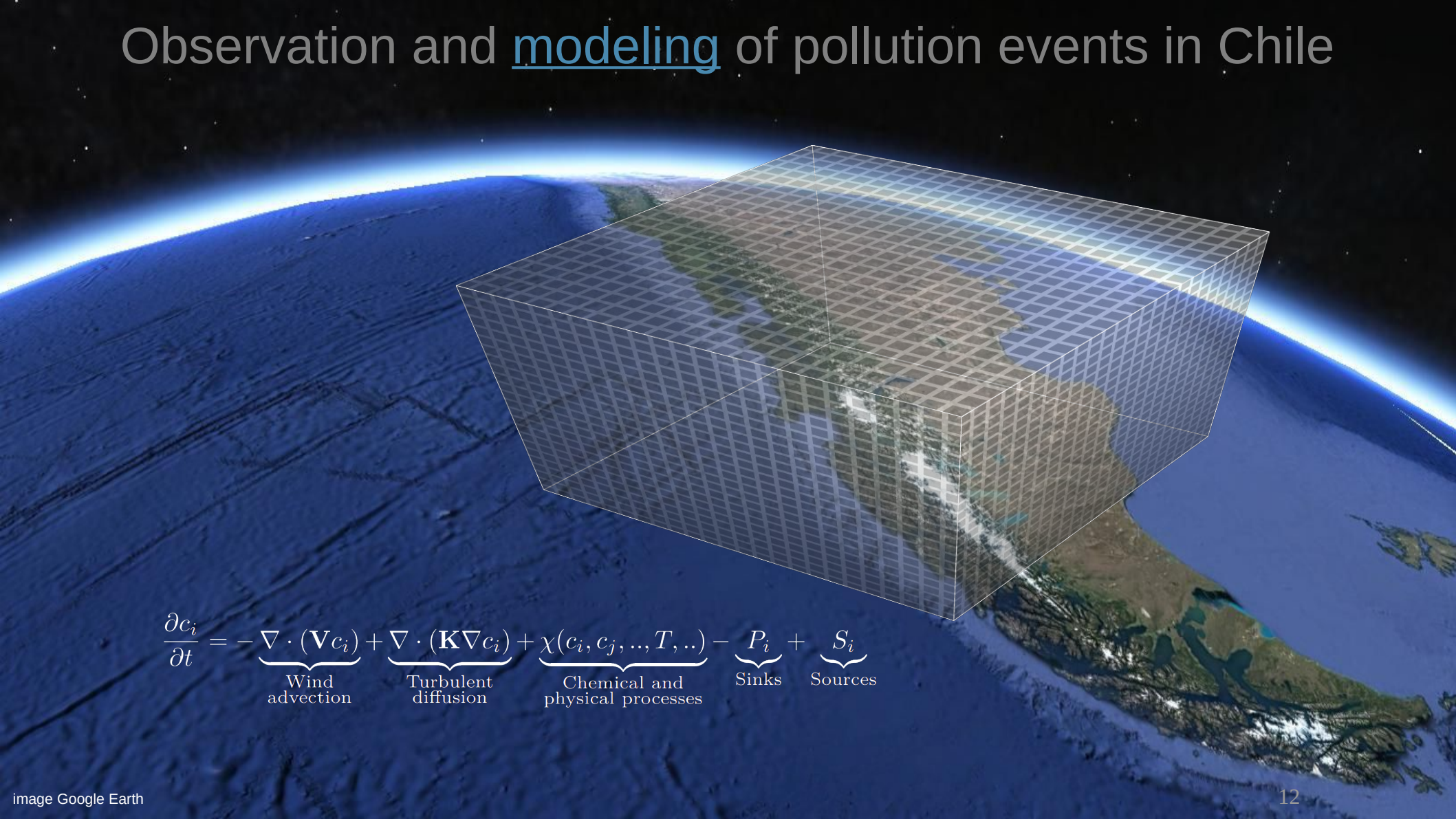


image Google Earth

Observation and modeling of pollution events in Chile



$$\frac{\partial c_i}{\partial t} = -\underbrace{\nabla \cdot (\mathbf{V} c_i)}_{\text{Wind advection}} + \underbrace{\nabla \cdot (\mathbf{K} \nabla c_i)}_{\text{Turbulent diffusion}} + \underbrace{\chi(c_i, c_j, .., T, ..)}_{\text{Chemical and physical processes}} - \underbrace{P_i}_{\text{Sinks}} + \underbrace{S_i}_{\text{Sources}}$$

image Google Earth

Observation and modeling of pollution events in Chile

WRF

+

Chimere

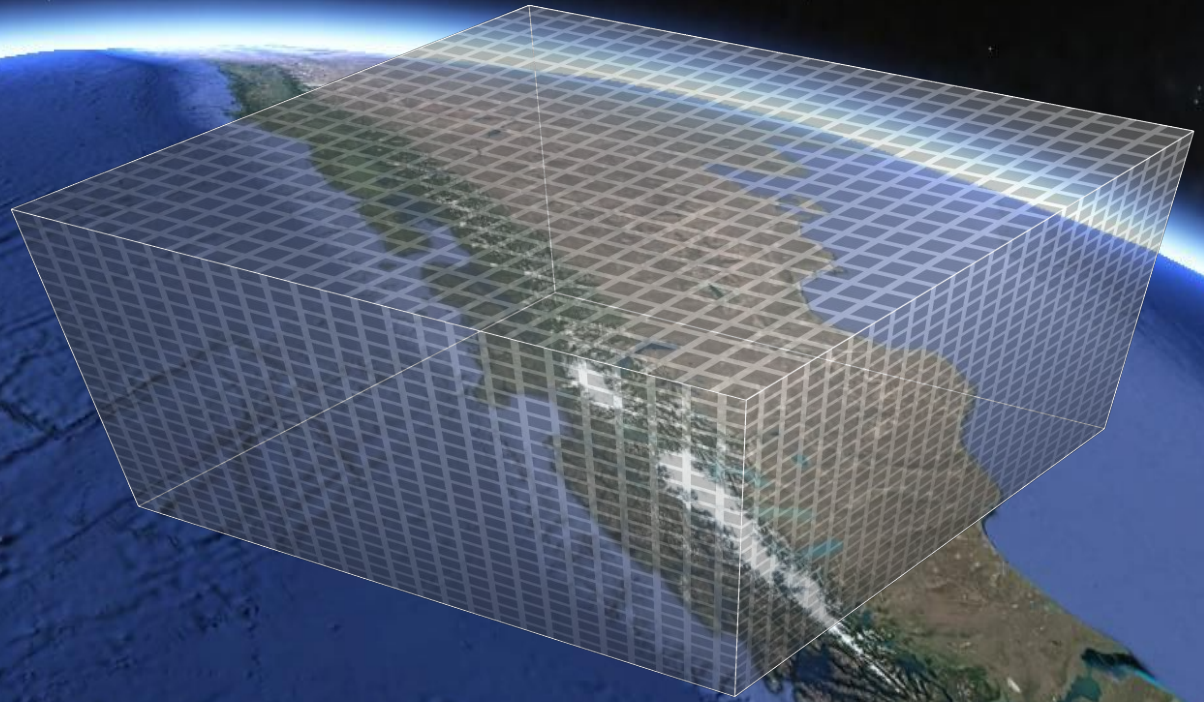


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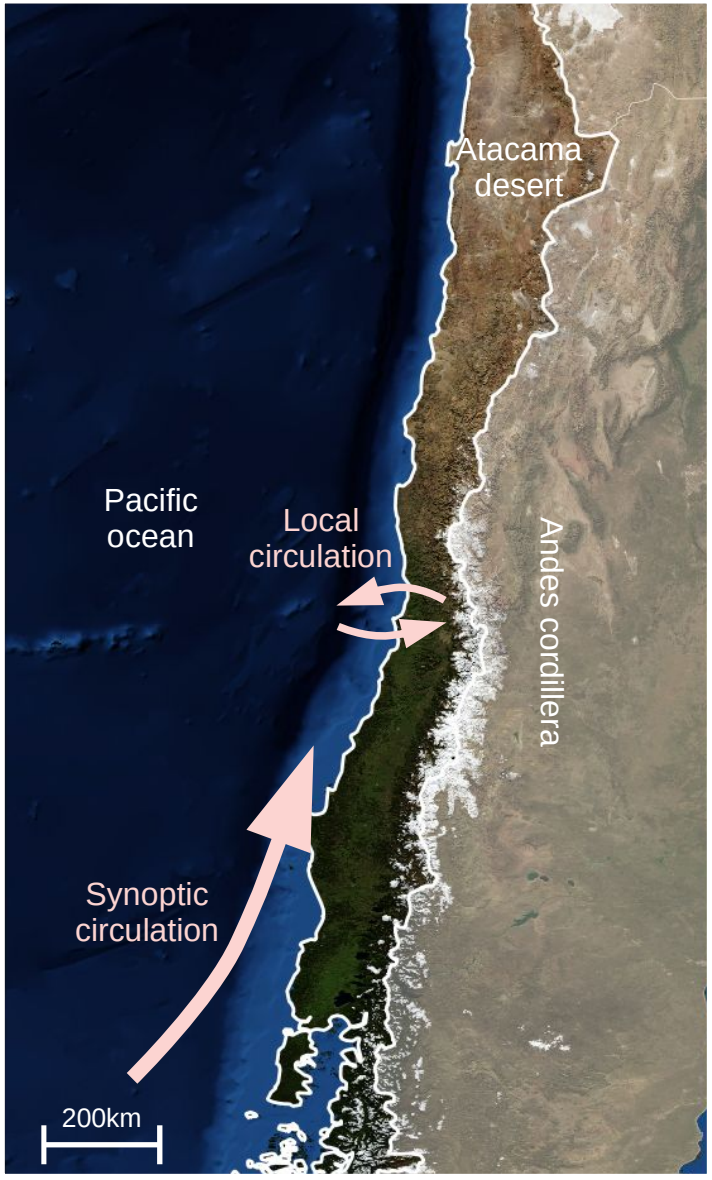
image Google Earth

Observation and modeling of pollution events in Chile



image Google Earth

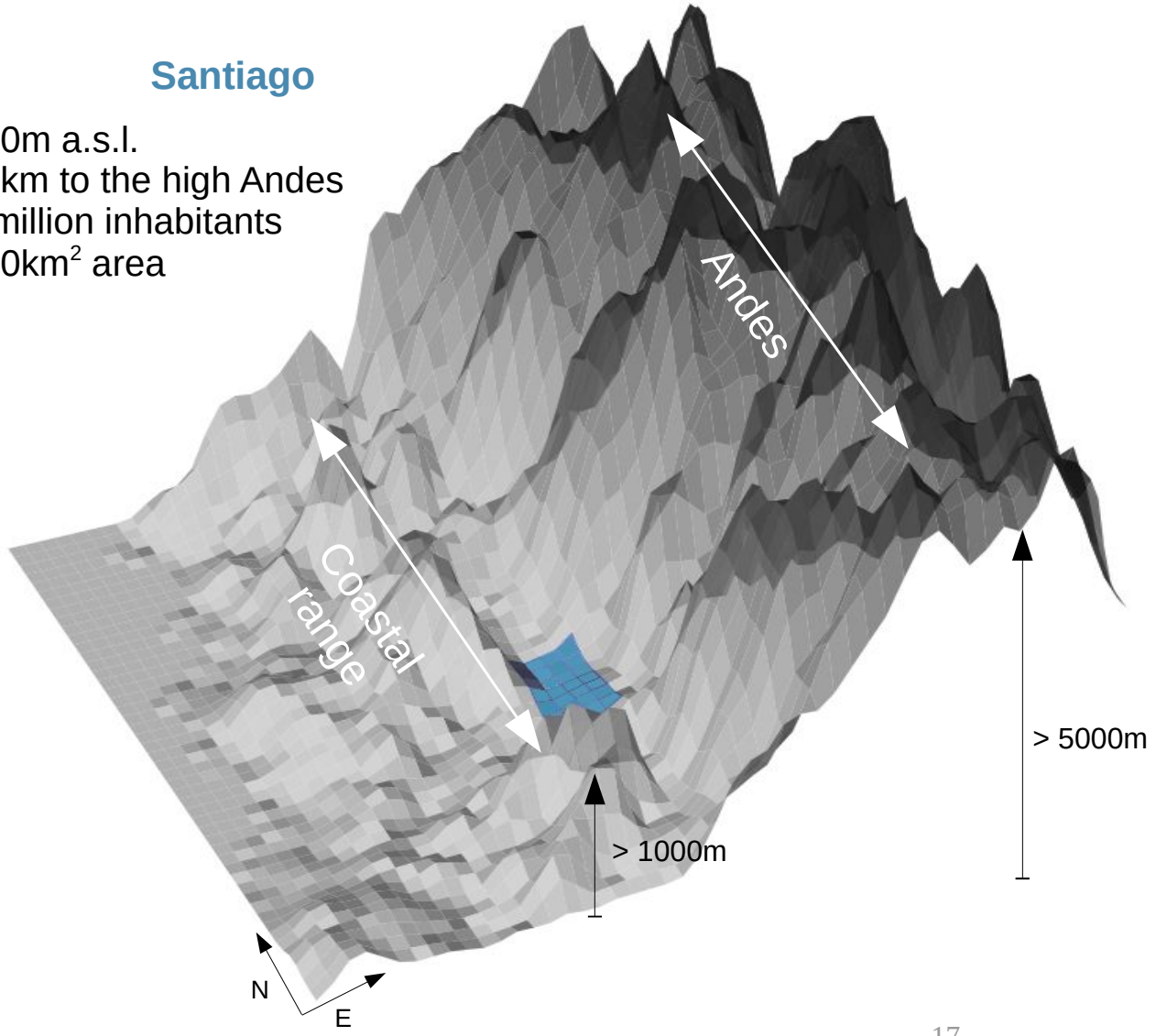

Atacama
desert
Pacific
ocean
Andes cordillera
200km

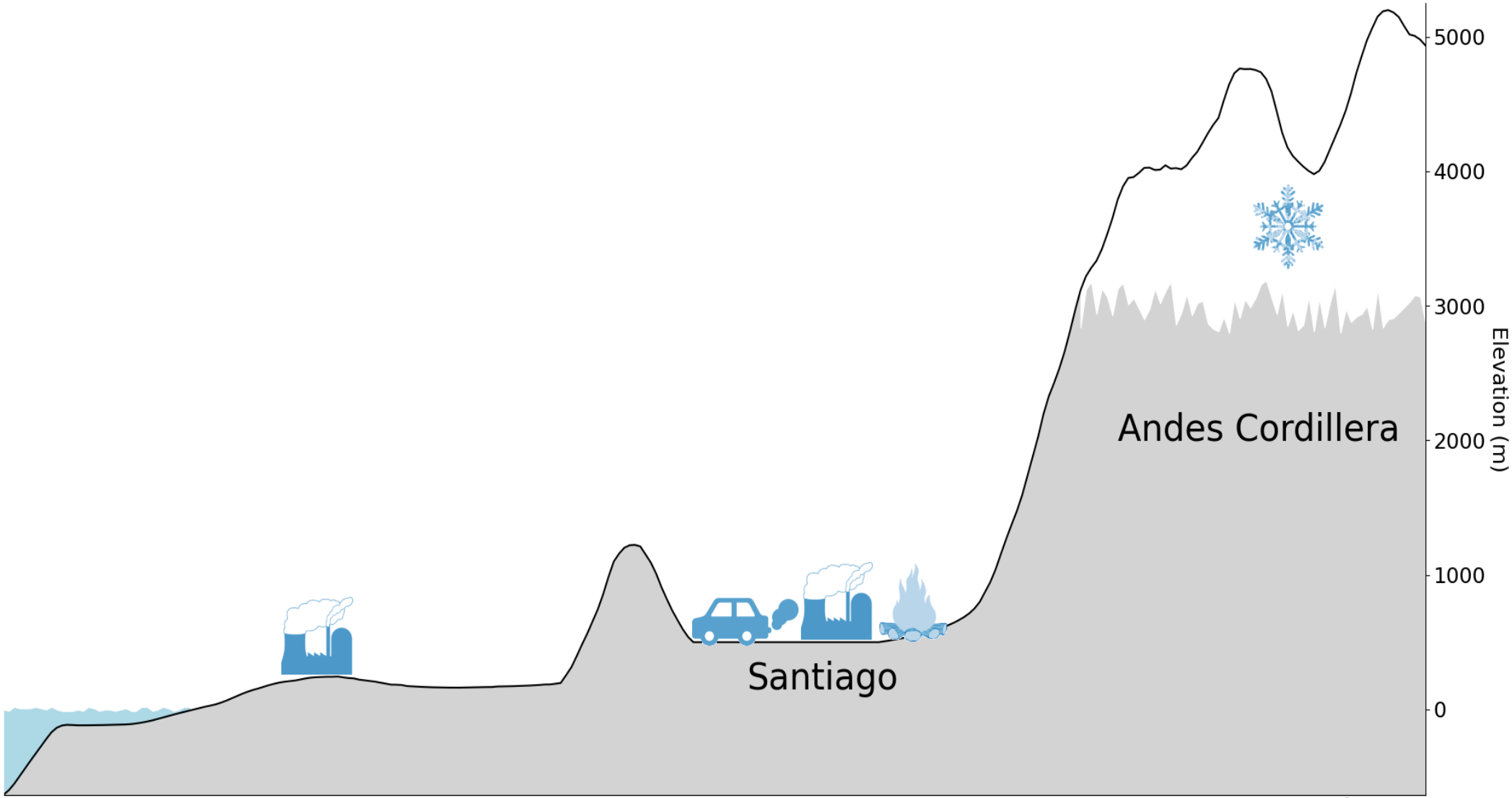

Atacama
desert
Pacific
ocean
Local
circulation
Andes cordillera
Synoptic
circulation
200km

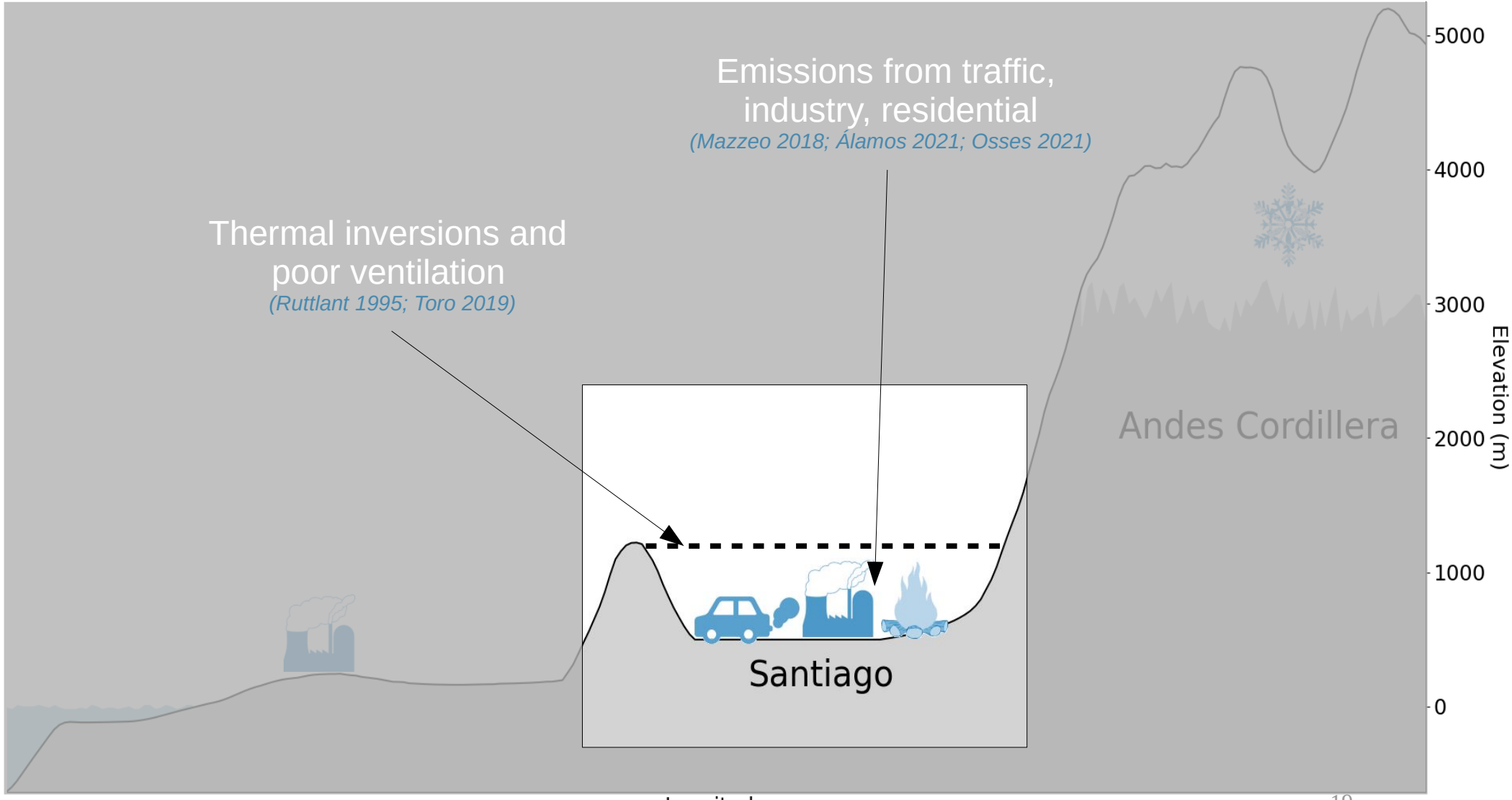


Santiago

- 500m a.s.l.
- 40km to the high Andes
- 6 million inhabitants
- 640km^2 area




5000
4000
3000
2000
1000
0
Elevation (m)
Andes Cordillera
Santiago
Longitude


Emissions from traffic, industry, residential
(Mazzeo 2018; Álamos 2021; Osses 2021)
Thermal inversions and poor ventilation
(Ruttlant 1995; Toro 2019)
Andes Cordillera
Santiago
5000
4000
3000
2000
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Elevation (m)
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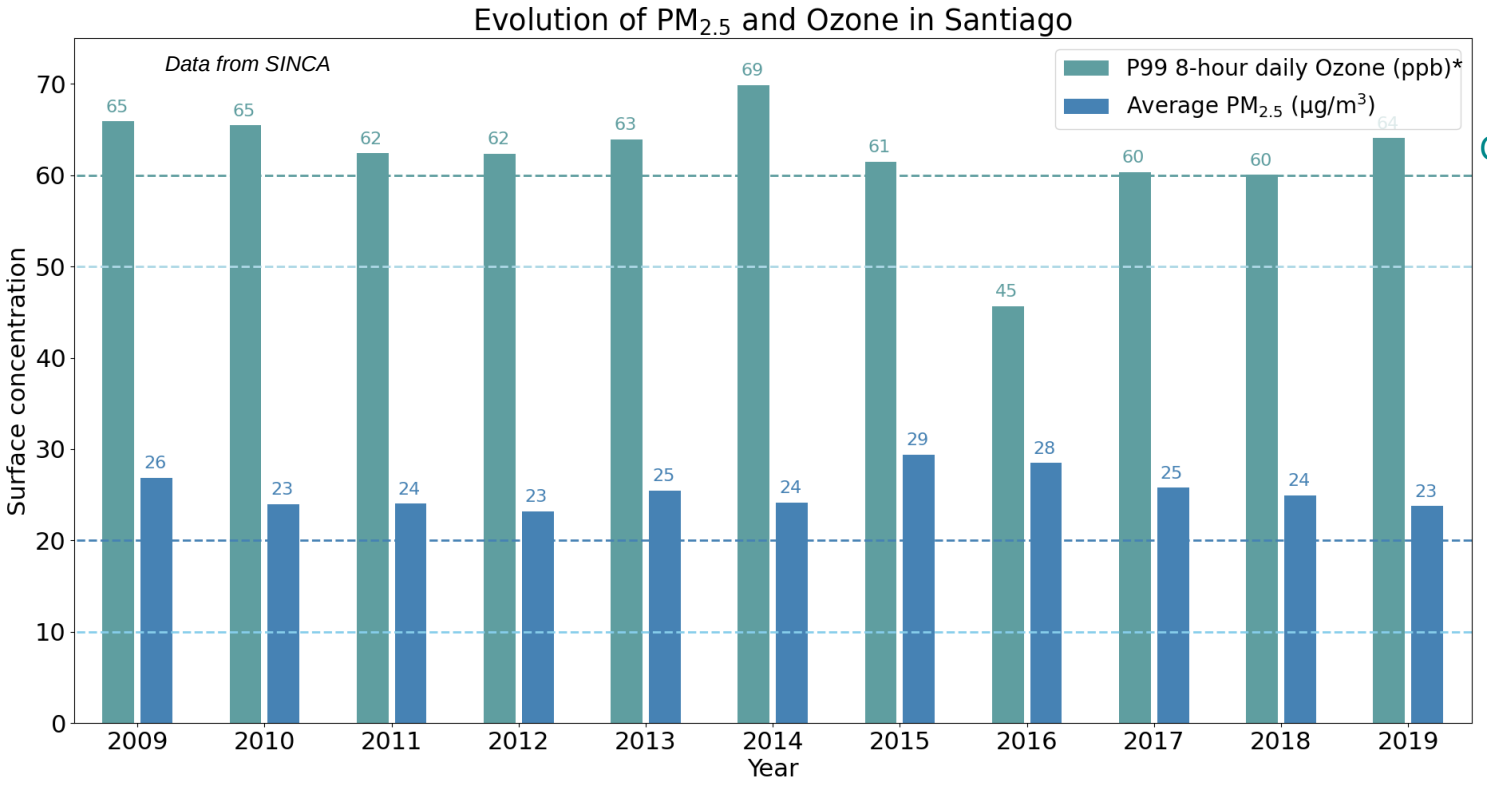
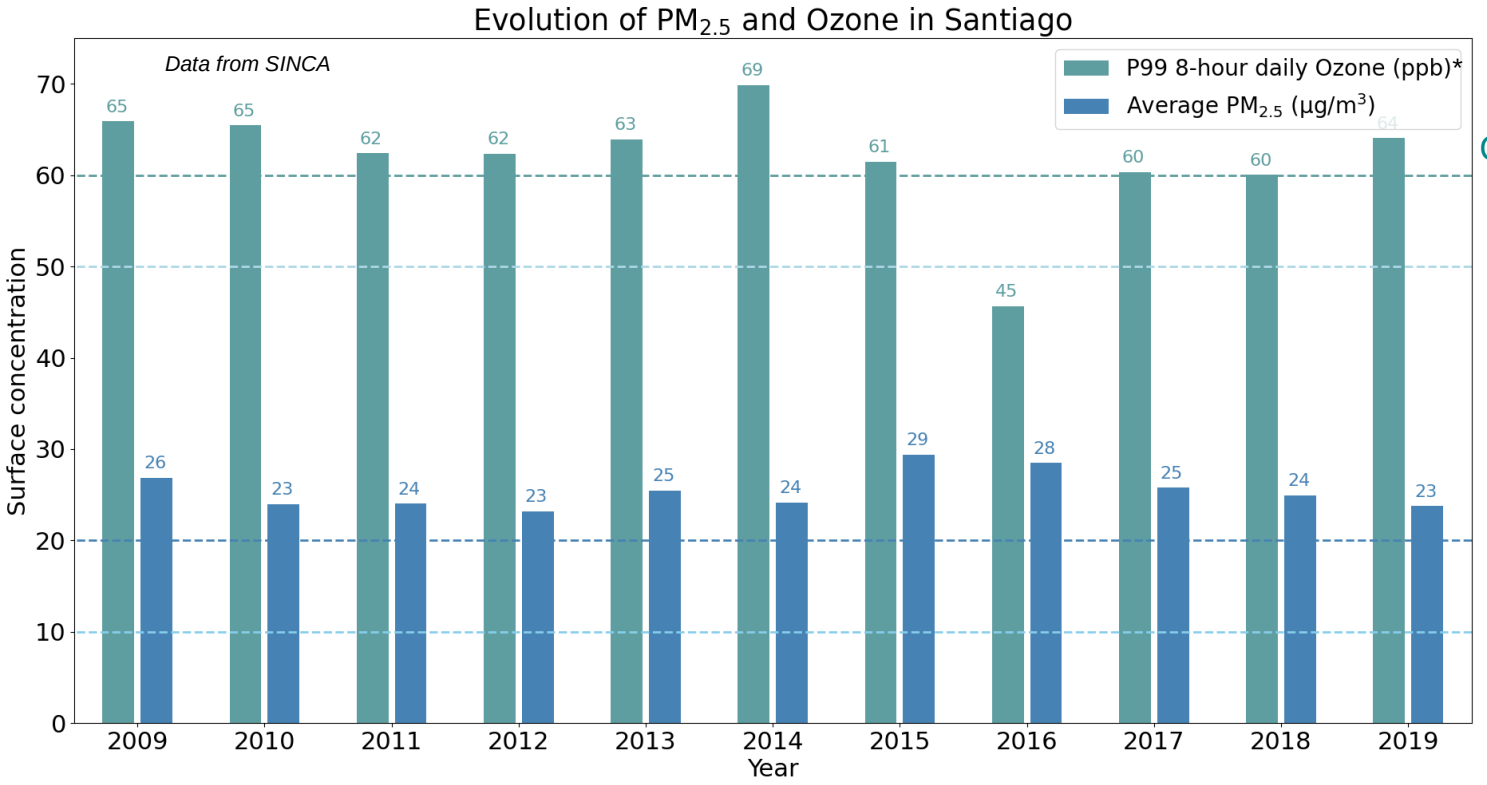


*Yearly 99th percentile of daily 8-hourly maximum Ozone



Chilean population	Death/year from air pollution(1)	Estimated economic cost of non-compliance(2)
19 million	**6,000**	**>200M$ / year**

(1) Health Effects Institute, State of Global Air 2020 *(2) MMA, 2012*

*Yearly 99th percentile of daily 8-hourly maximum Ozone

Outline

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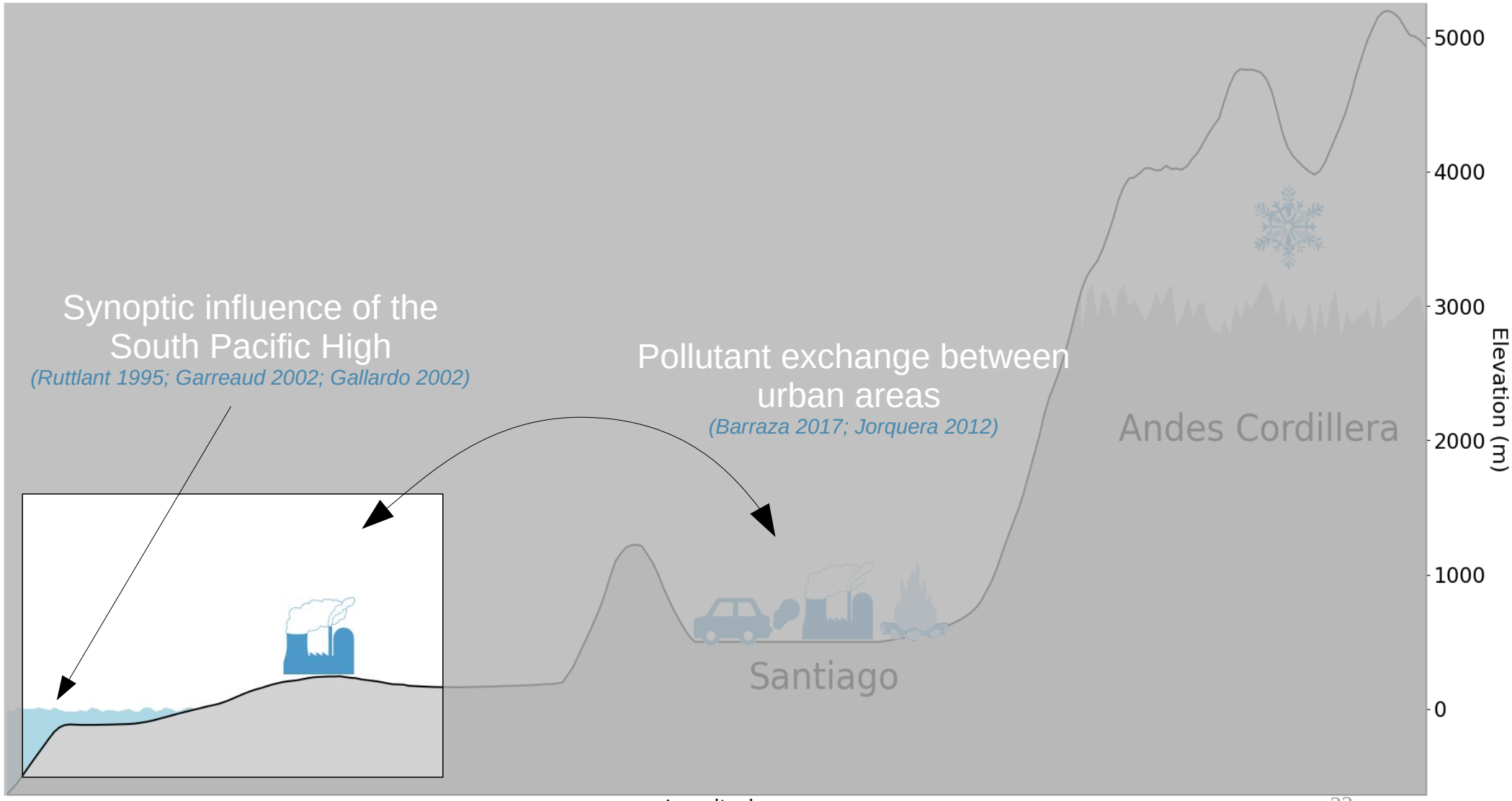
- ϕ Can we explain recent record-breaking winter $PM_{2.5}$ peak events?
- ϕ What were the atmospheric impacts of the 2017 mega-fires?


Synoptic influence of the South Pacific High
(Ruttlant 1995; Garreaud 2002; Gallardo 2002)
Pollutant exchange between urban areas
(Barraza 2017; Jorquera 2012)
Andes Cordillera
Santiago
5000
4000
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2000
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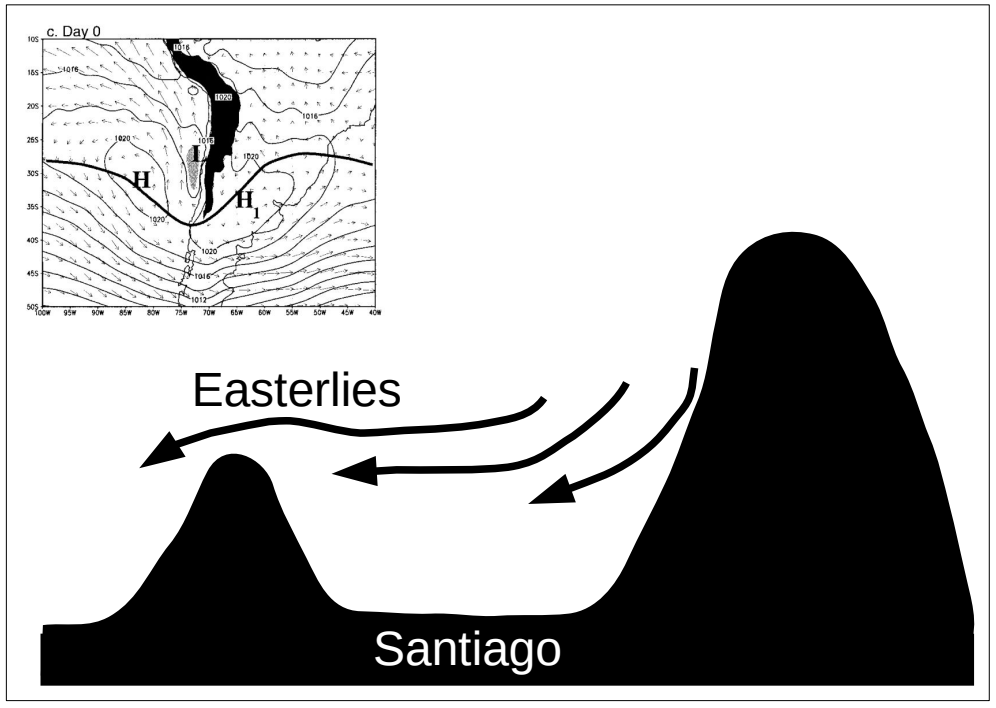
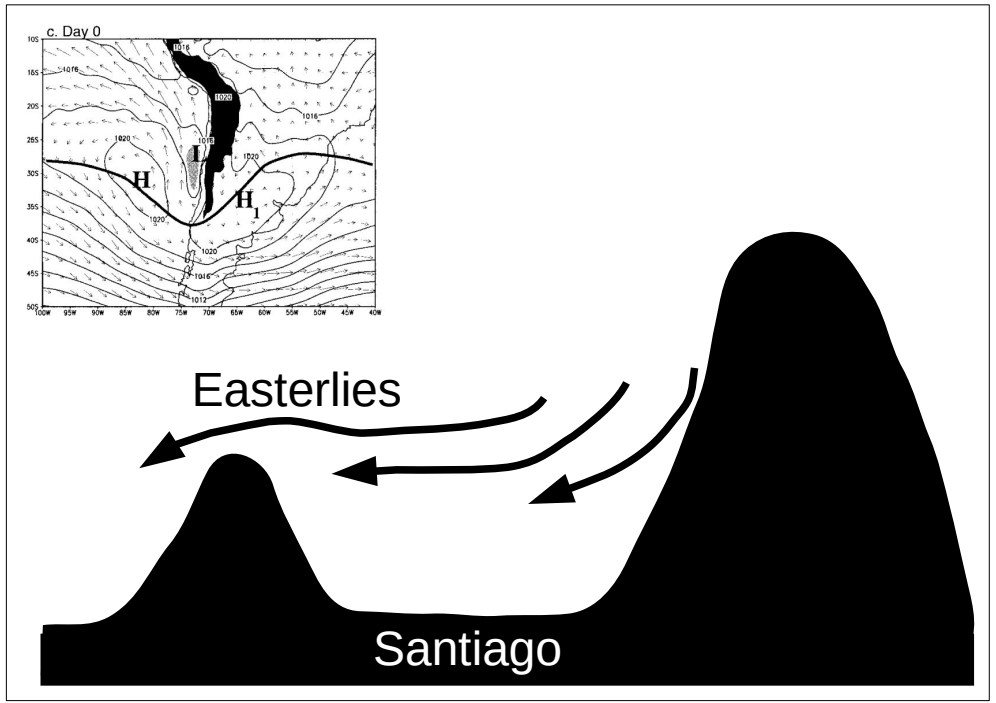
0
Elevation (m)
Longitude

Coastal lows strengthen
inversion above Santiago



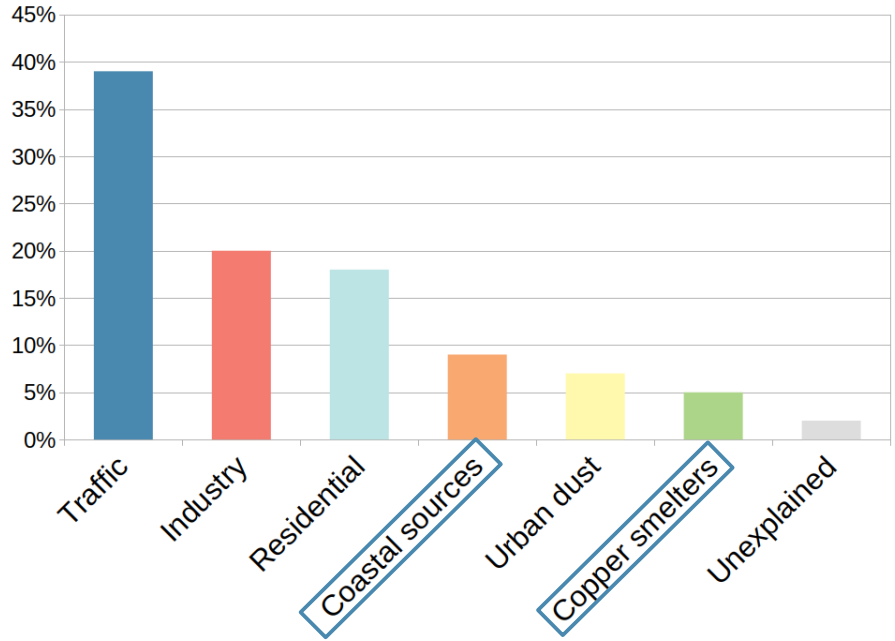
Adapted from *Garreaud et al., 2002* and *Gallardo et al., 2002*

Coastal lows strengthen inversion above Santiago



Adapted from *Garreaud et al., 2002* and *Gallardo et al., 2002*

Contribution of emission sectors to $PM_{2.5}$ in Santiago, Chile. 2011–2012.



Adapted from *Barraza et al., 2017*

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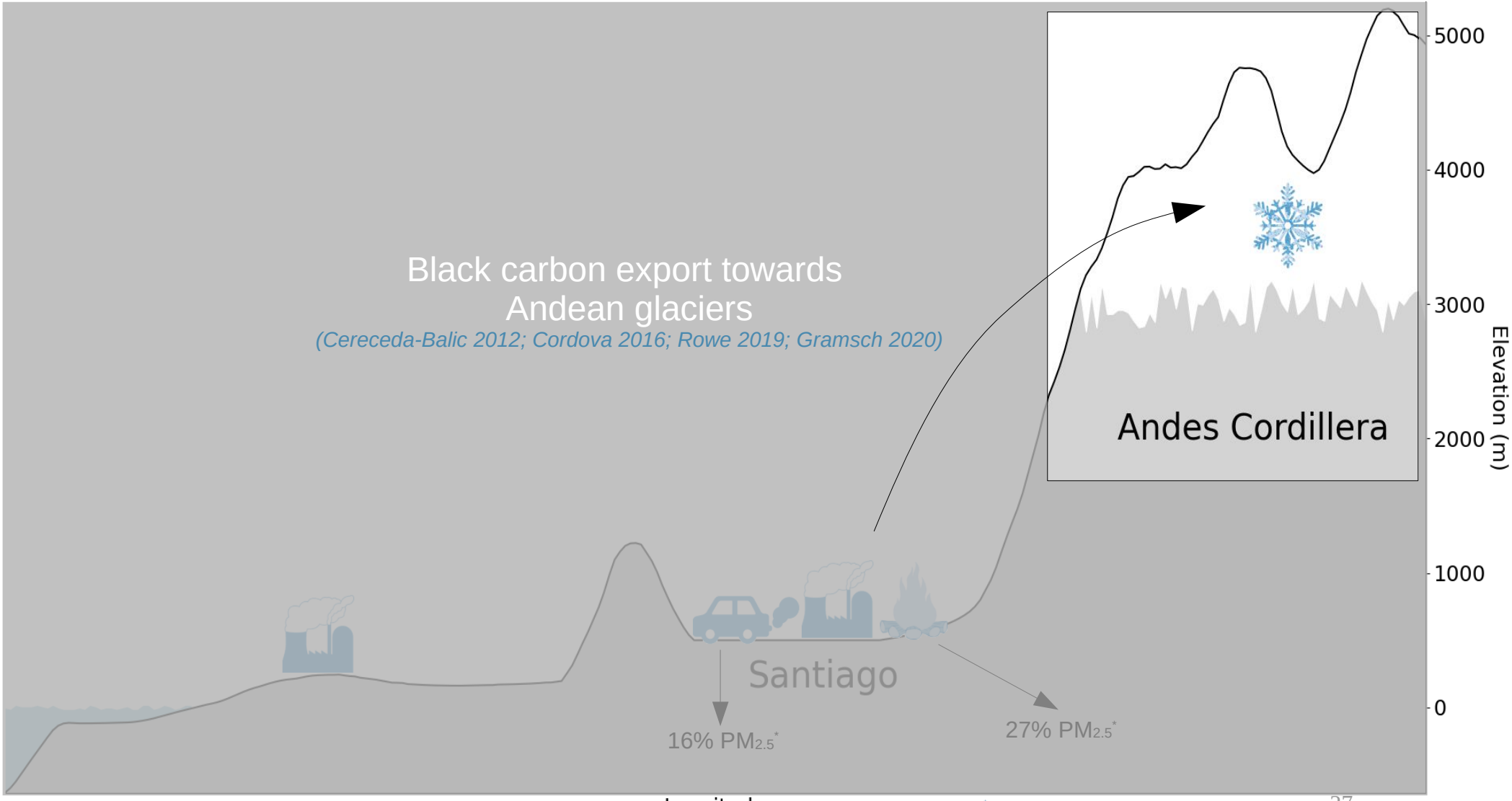
- ϕ What are the characteristics of seasonal air pollutant transport?
- ϕ How far do emissions from Santiago transport?



*Villalobos et al., 2015



source pexels.com



source pexels.com



- faster melting
- stress on water supply in summer
- positive climate feedback

source pexels.com

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1.a Winter 2016 *(Lapere et al., 2020)*
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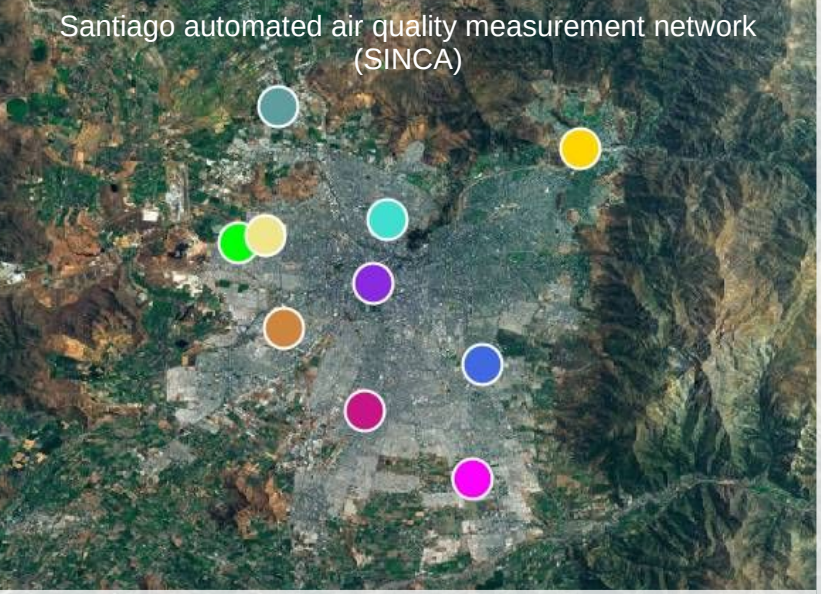
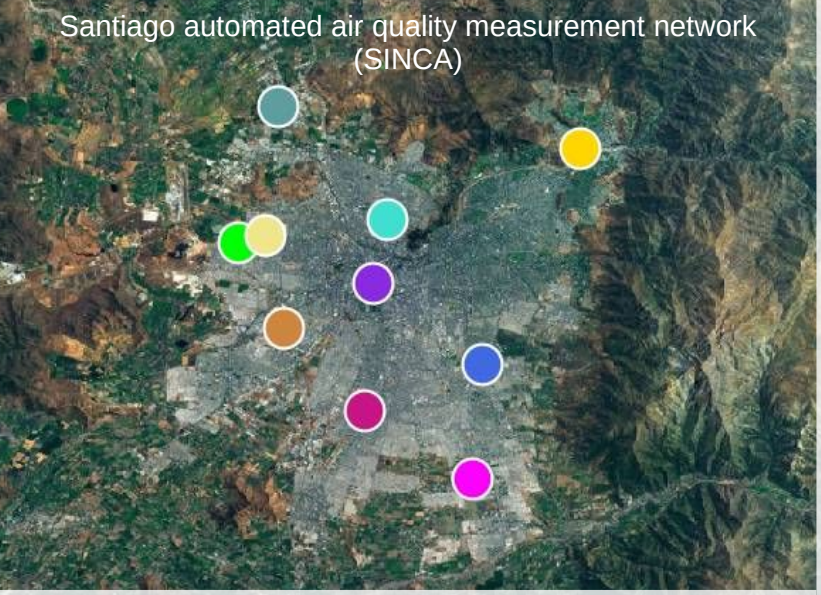

Santiago automated air quality measurement network
(SINCA)



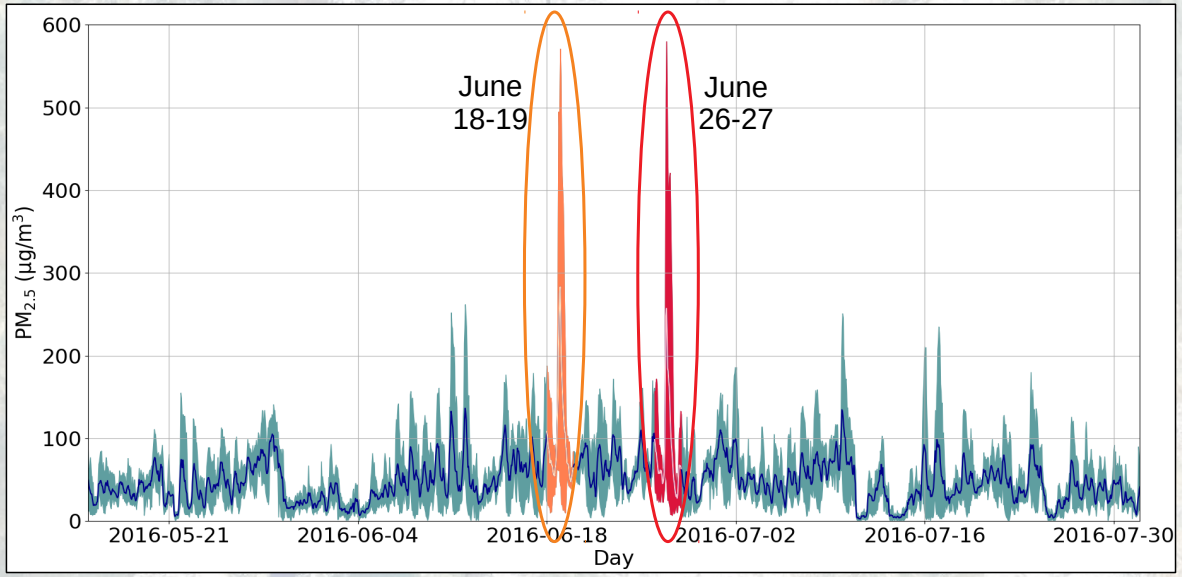
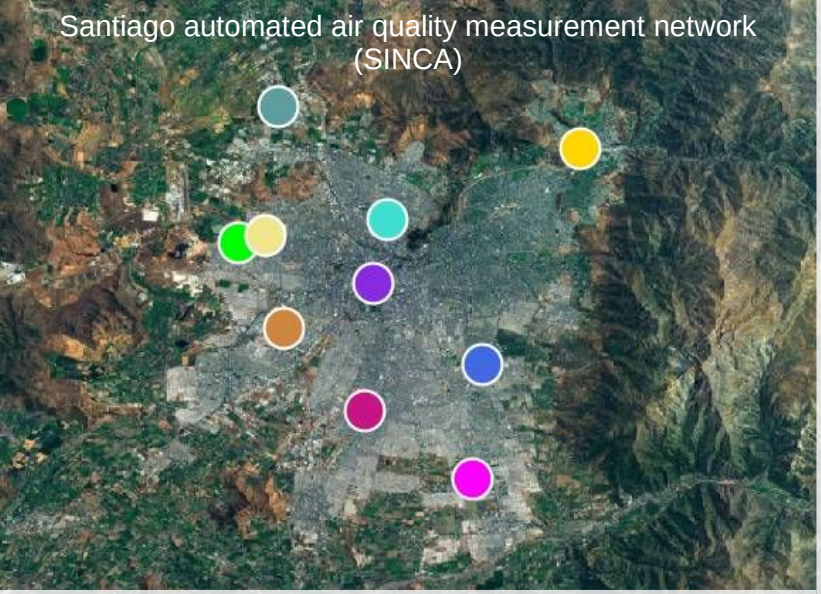
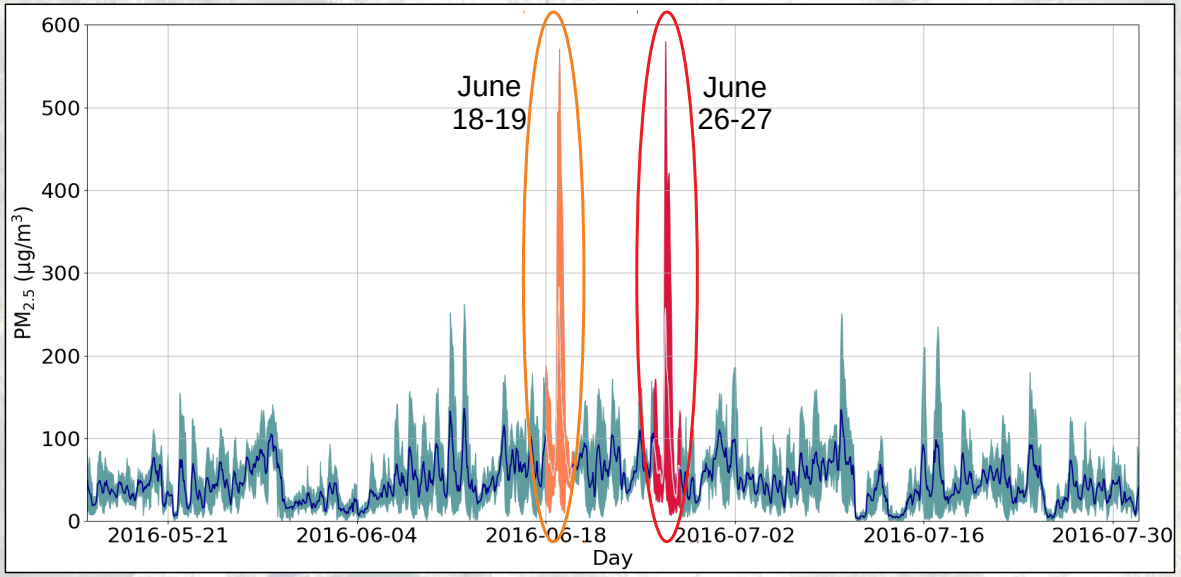


Figure Time series of hourly $PM_{2.5}$ concentrations in Santiago. Average/min/max over the automated network. May-July 2016. Data from SINCA.





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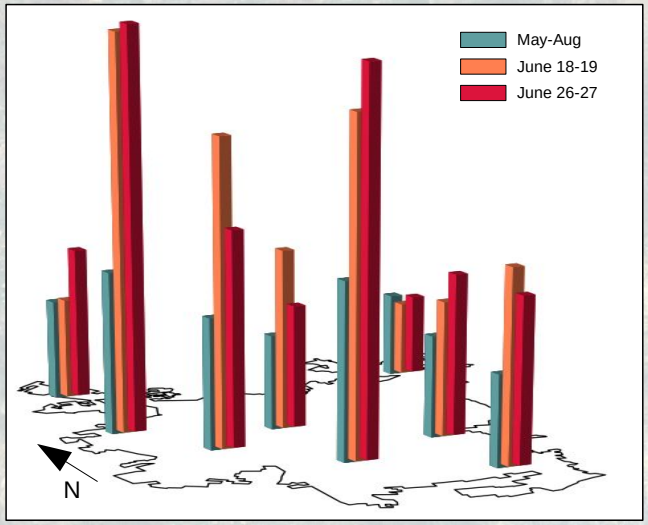
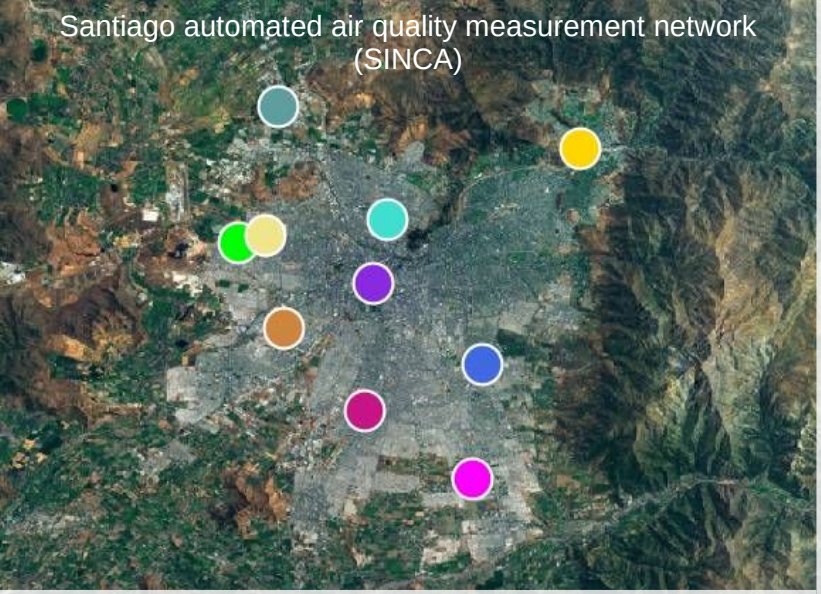
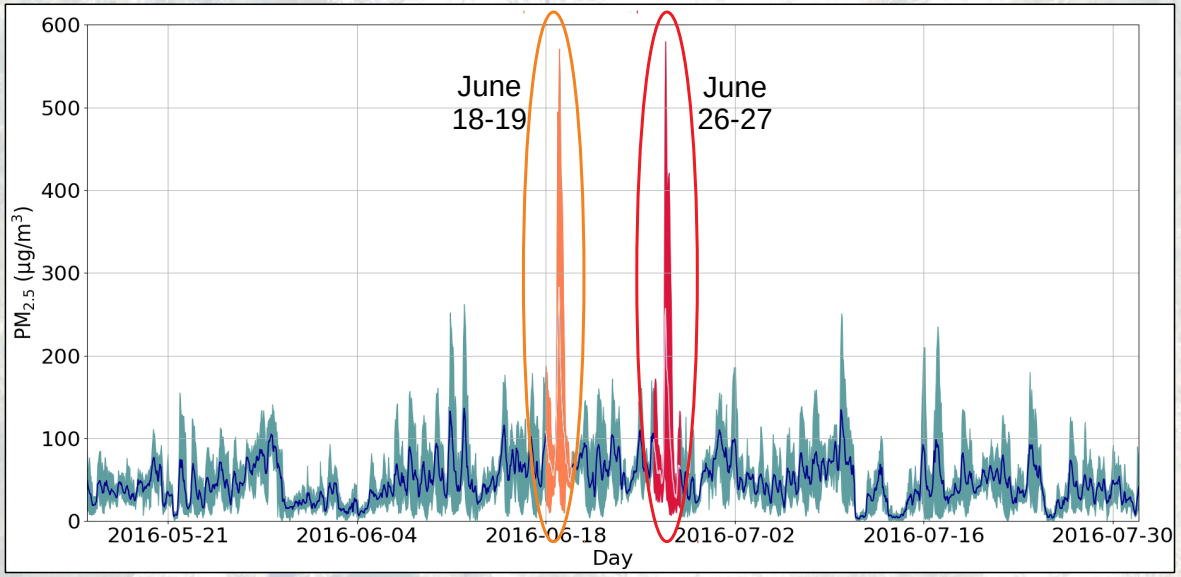


Figure Maximum hourly $PM_{2.5}$ in Santiago during winter 2016





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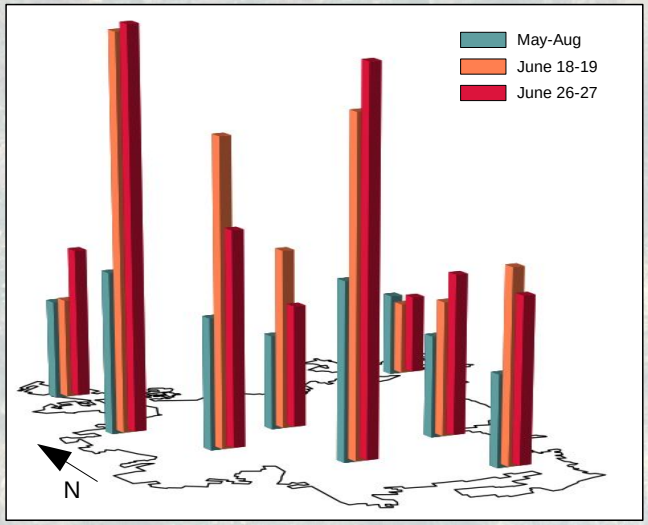

Figure Maximum hourly $PM_{2.5}$ in Santiago during winter 2016

Why did these peaks occur?

Can we model these events?

Candidate origins

Possible explanations include:

- Extreme meteorological conditions
- Plume advection from remote source
- Exceptional local emissions

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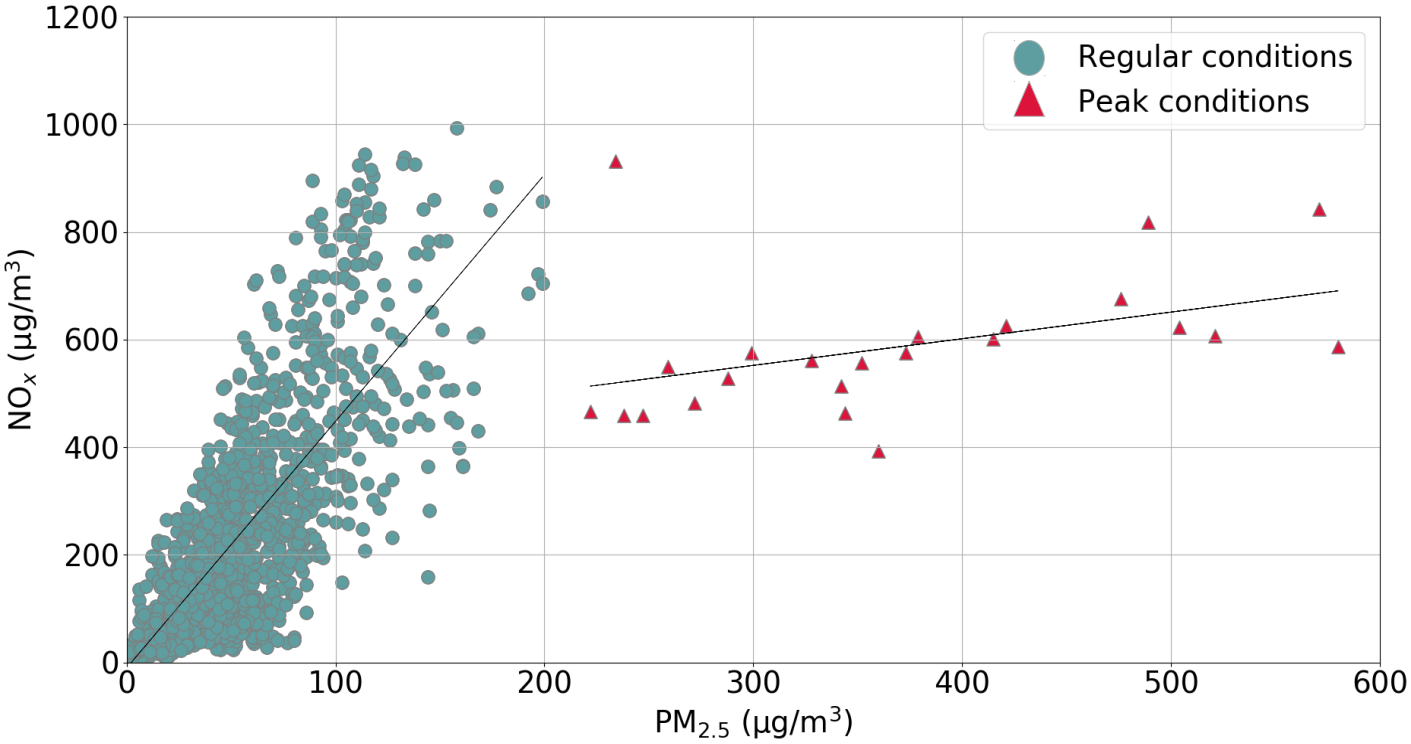
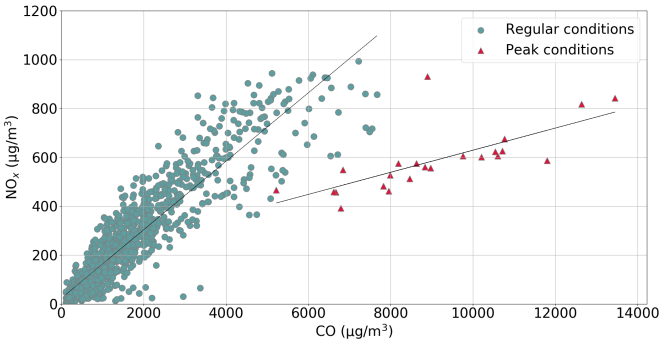
A specific chemical footprint

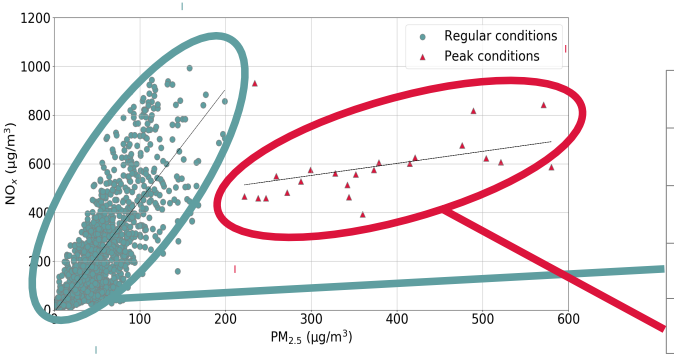




Concentration **ratios of pollutants** greatly differ between usual and peak conditions

Figure Observed NO_X and $PM_{2.5}$ under regular conditions (blue dots) and high $PM_{2.5}$ load (red triangles). Stations *Pudahuel*, *Las Condes*, *La Florida*, *Parque O'Higgins*, *Puente Alto* and *Cerrillos*. June-July 2016. Data from SINCA.

A specific chemical footprint



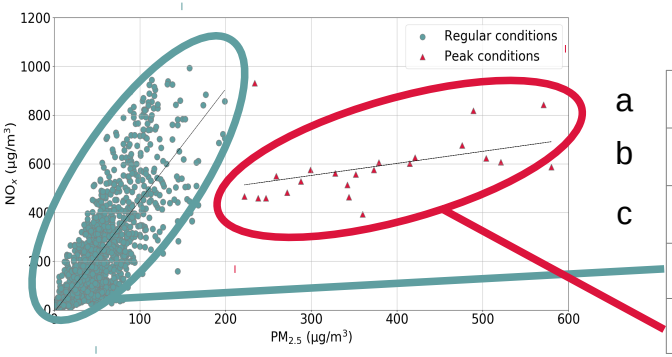
	$NO_x/PM_{2.5}$	NO_x/CO
Traffic	1750%	7.5%
Residential	200%	19%
Industry	186%	20%
Observed	**502%**	**14%**
Observed	**73%**	**5%**

Moderate PM load

High PM load (>200µg/m^3)

Table Emission ratios by source type for Santiago. Data from HTAP and SINCA.

A specific chemical footprint



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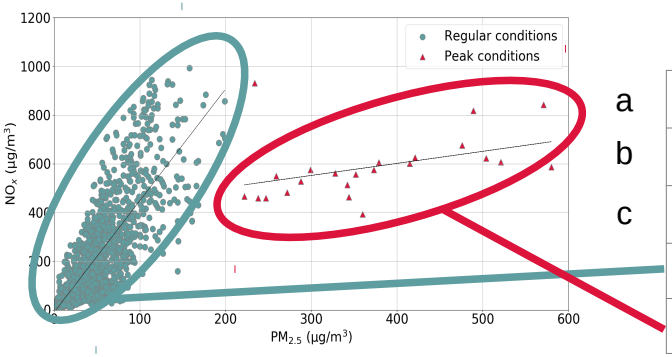

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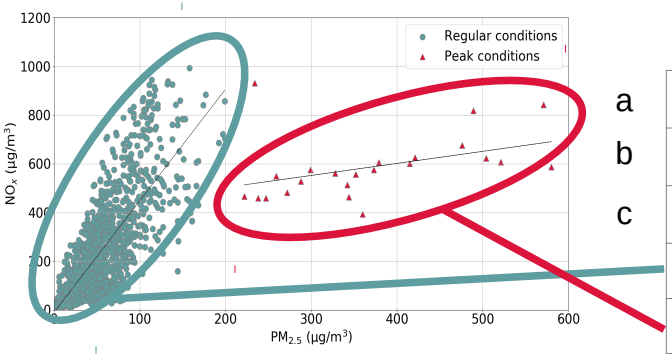
(a, b, c) exist
(a, b, c) do not

A specific chemical footprint



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$$\begin{cases} a \times 1750 + b \times 200 + c \times 186 = Observed_NO_X/PM_{2.5} \\ a \times 7.5 + b \times 19 + c \times 20 = Observed_NO_X/CO \end{cases}$$

(a, b, c) exist
(a, b, c) do not

The **usual mix** of sources **cannot account** for the observed chemical footprint during peak events

A correlation with soccer games of *La Roja*

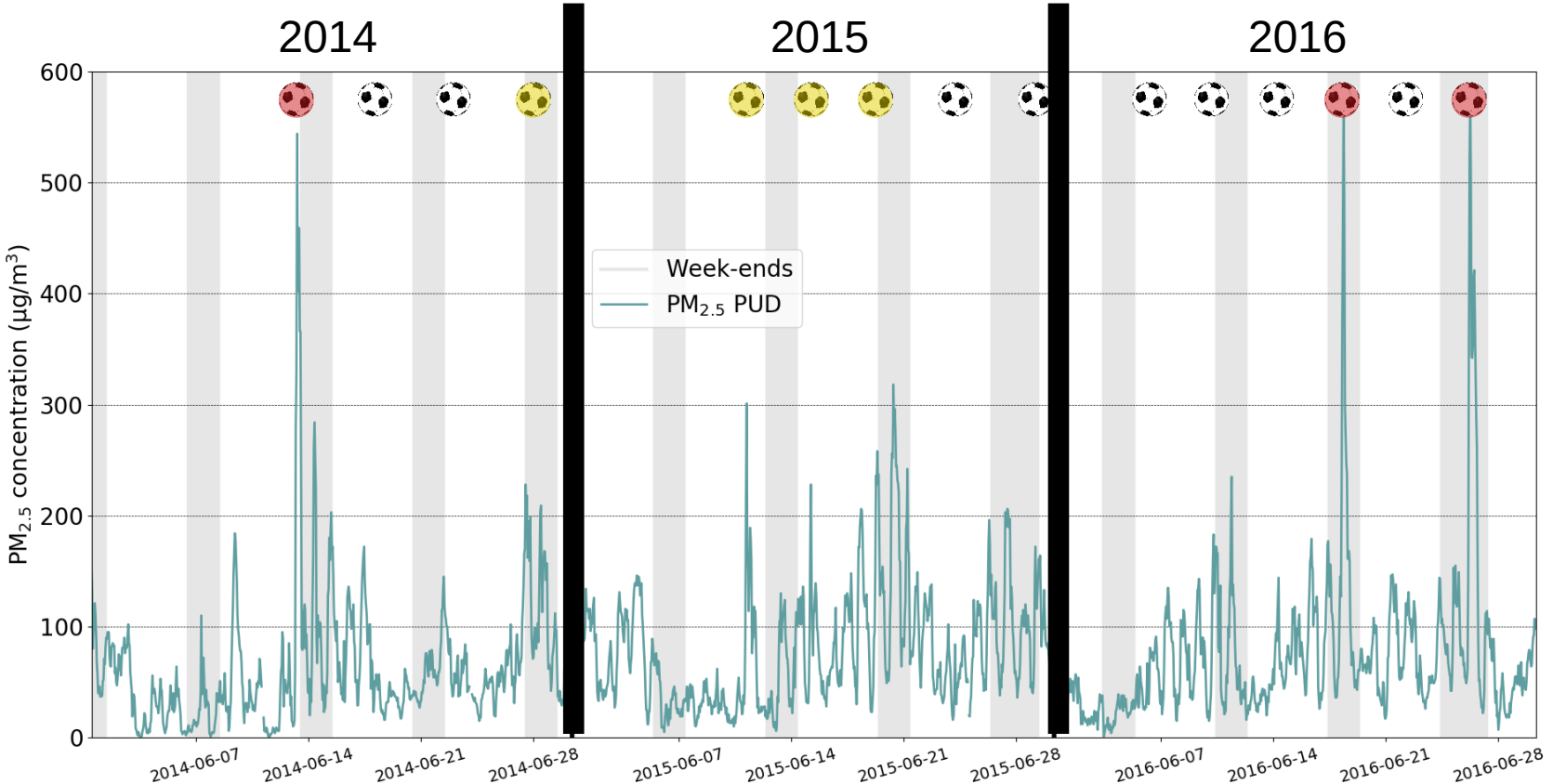

Figure Fine particle concentrations at Pudahuel (blue line) and soccer games kickoff. June 2014, 2015 and 2016.

A correlation with soccer games of *La Roja*



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A correlation with soccer games of *La Roja*

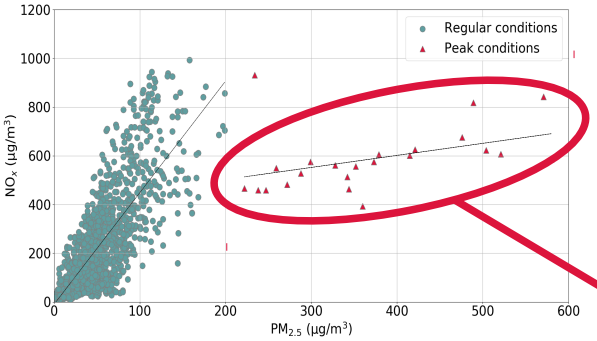






Barbecues? - independent pieces of evidence

1st piece of evidence: Emission factors



	$NO_x/PM_{2.5}$	NO_x/CO
Traffic	1750%	7.5%
Residential	200%	19%
Industry	186%	20%
Barbecue	**41-62%**	**1.4-2.4%**
Observed	**73%**	**5%**

Table Emission ratios by source type in HTAP for Santiago and barbecue emission ratios from *Lee, 1999* and *Vicente et al., 2018*

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(a, b, c, d) exist!	**Observed**	**73%**	**5%**

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2nd piece of evidence: Modeling with CHIMERE

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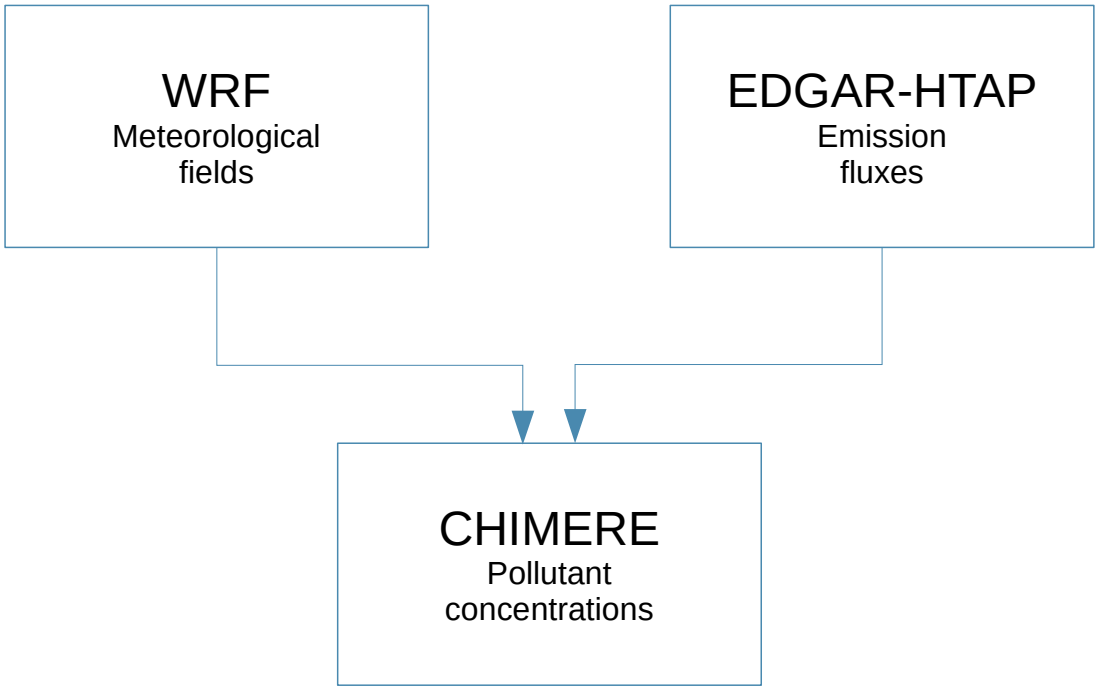

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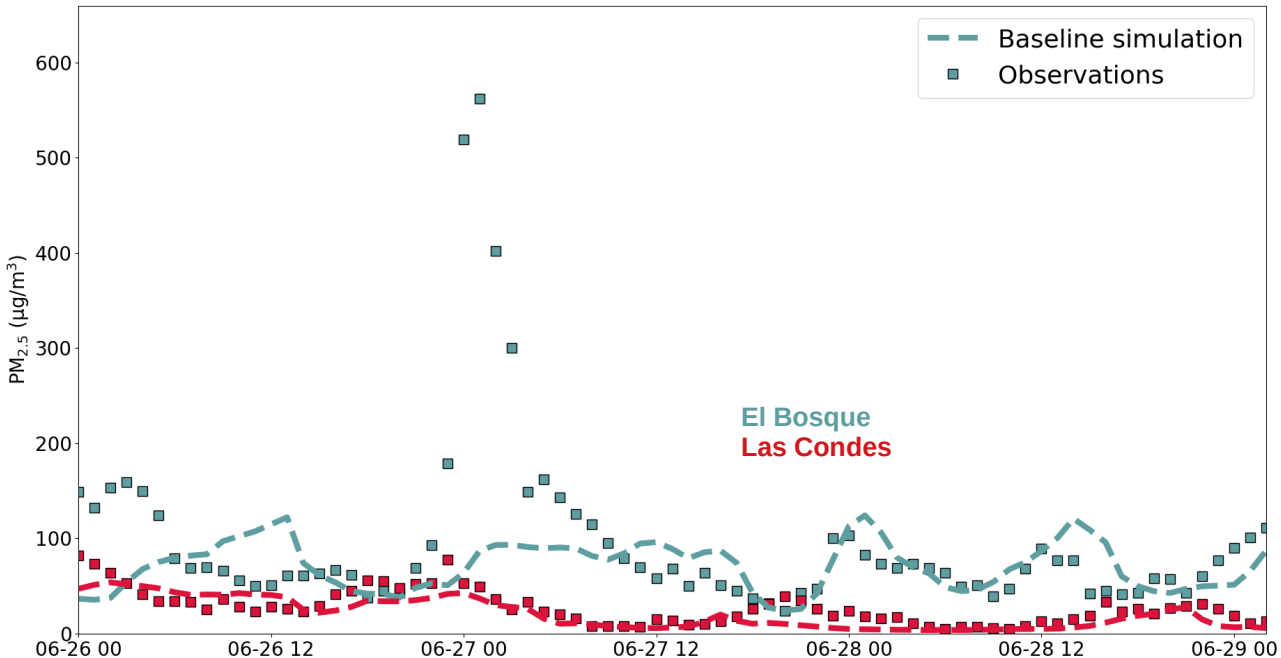

Figure Observed and modeled fine particle concentrations on June 26, without barbecue emissions

Barbecues? - independent pieces of evidence

2nd piece of evidence: Modeling with CHIMERE

- 2016 poll: 29% of Santiaguinos would cook a barbecue for the final game
- Around **100,000 fires** i.e. **+2ton/h of $PM_{2.5}$**
- Included in CHIMERE, at the time of the game, spatially distributed based on population

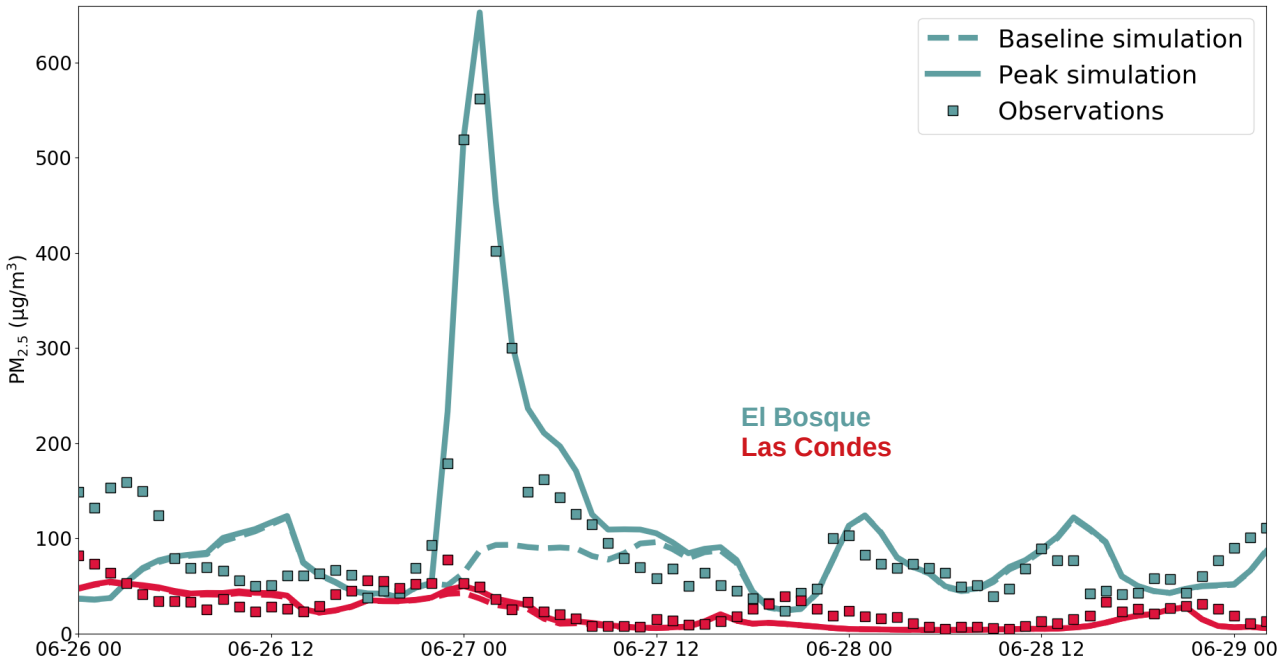

Figure Observed and modeled fine particle concentrations on June 26, with and without barbecue emissions

Conclusions

>500µg/m³

$PM_{2.5}$ for a few hours

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Barbecue

emissions during soccer games explain these levels

Conclusions

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Barbecue

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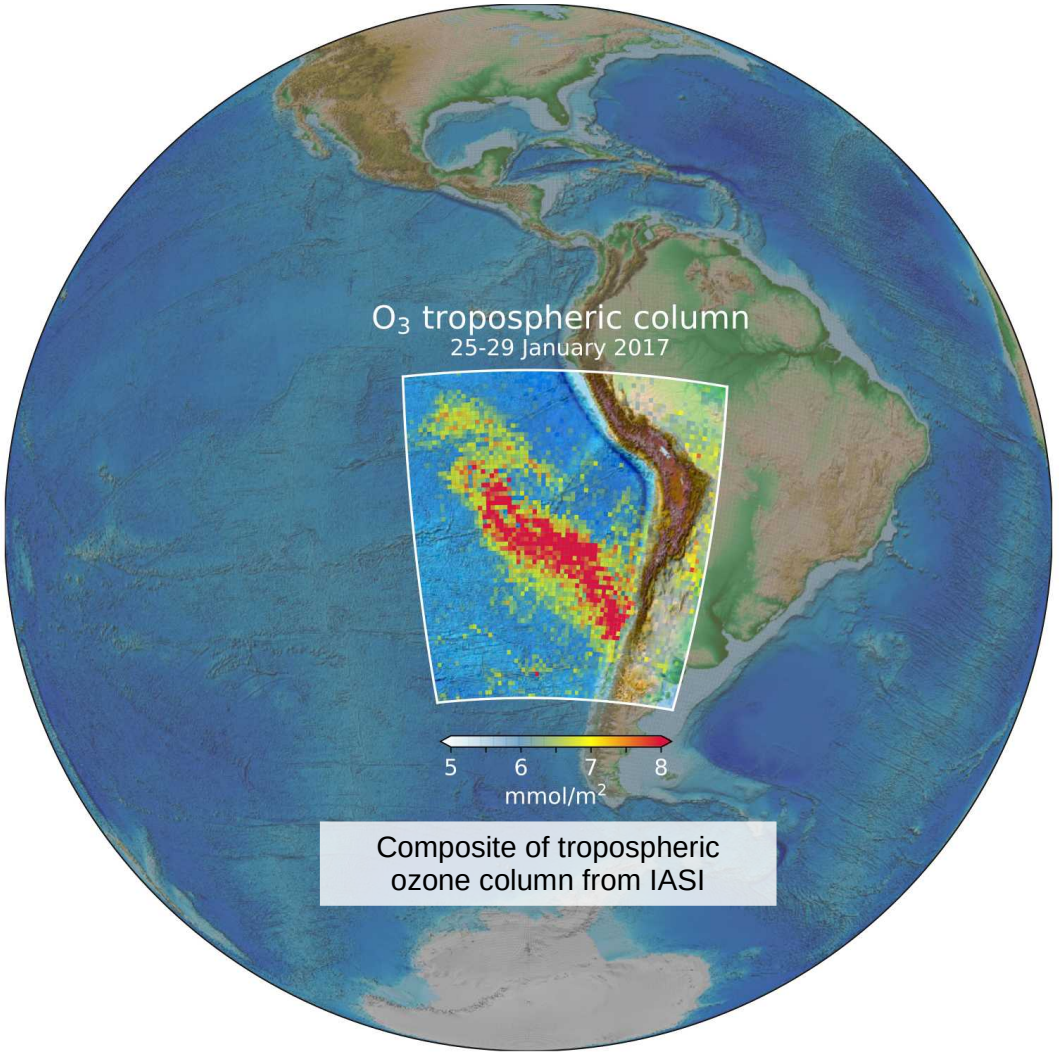
>50%

of days with >200µg/m^3 due to the “game effect”

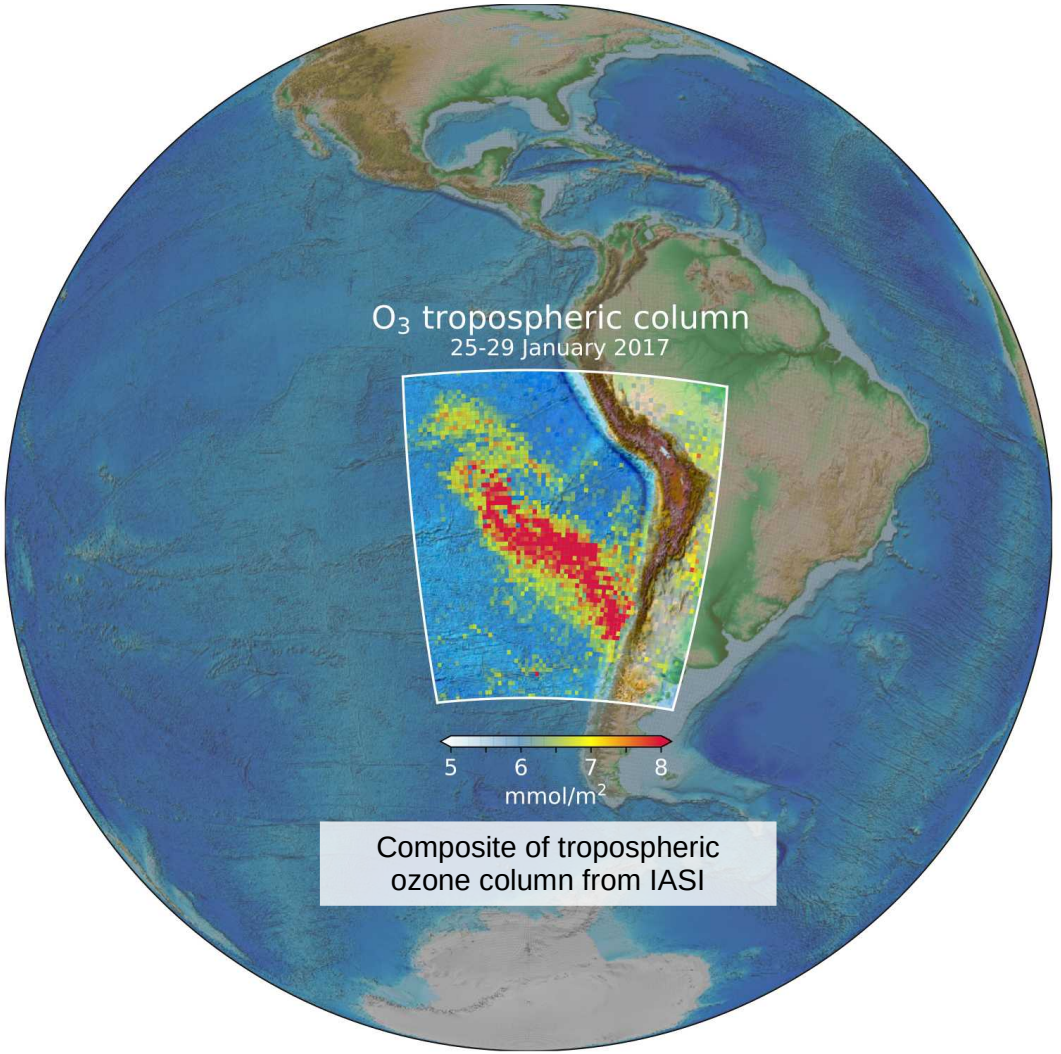
1 - Air pollution extreme events

1.a Winter 2016 *(Lapere et al., 2020)*

1.b Summer 2017 *(Lapere et al., 2021b)*



Composite of tropospheric ozone column from IASI



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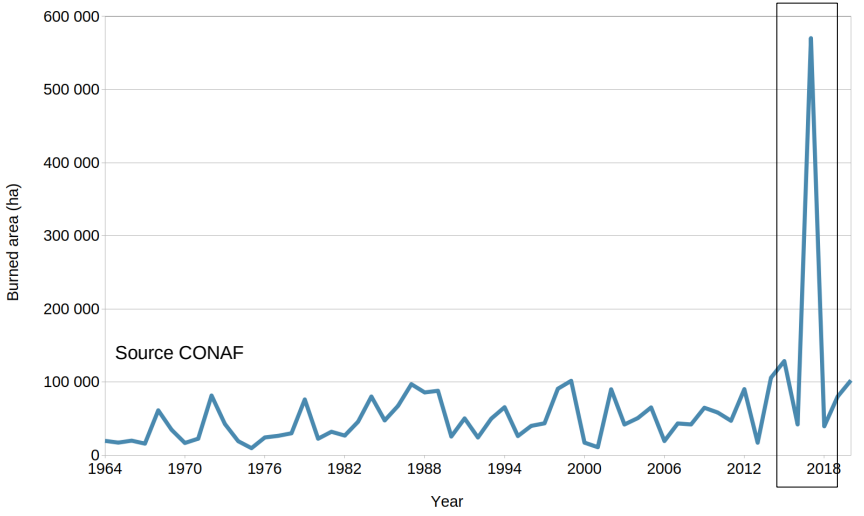

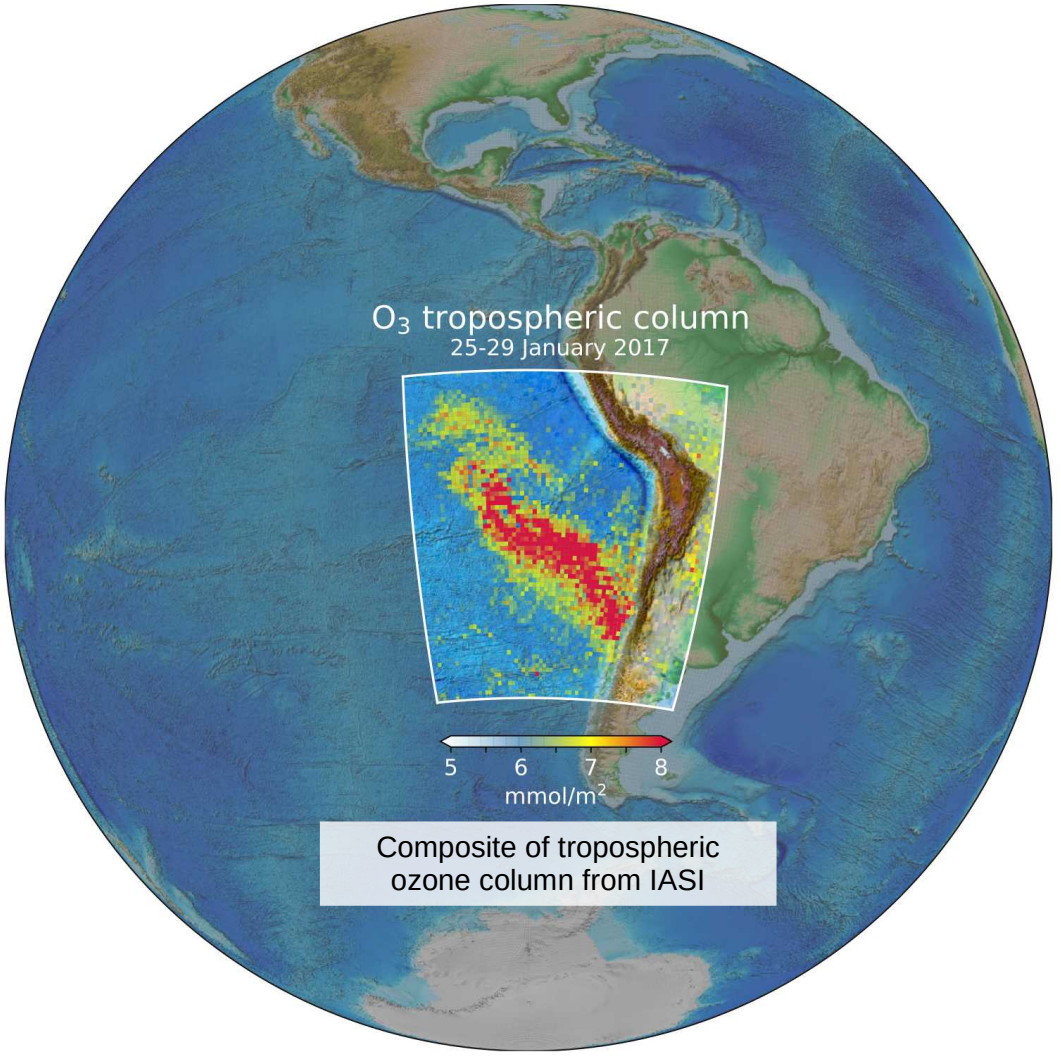

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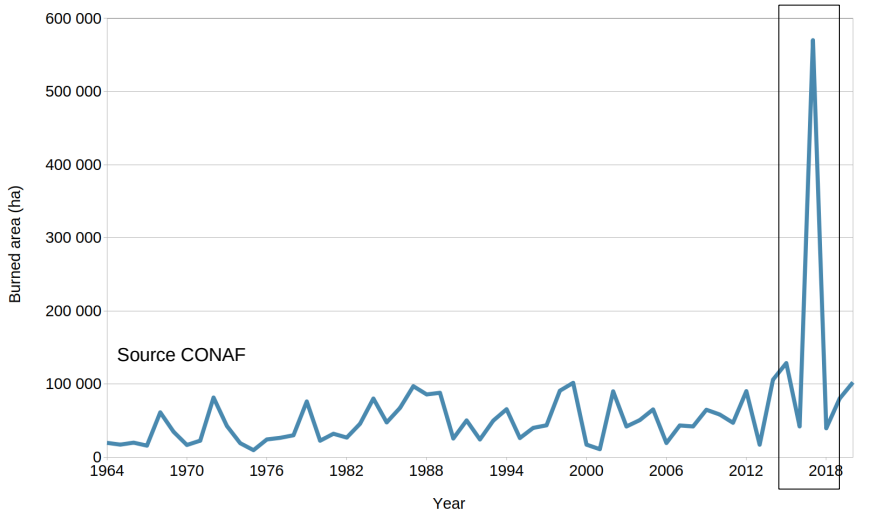
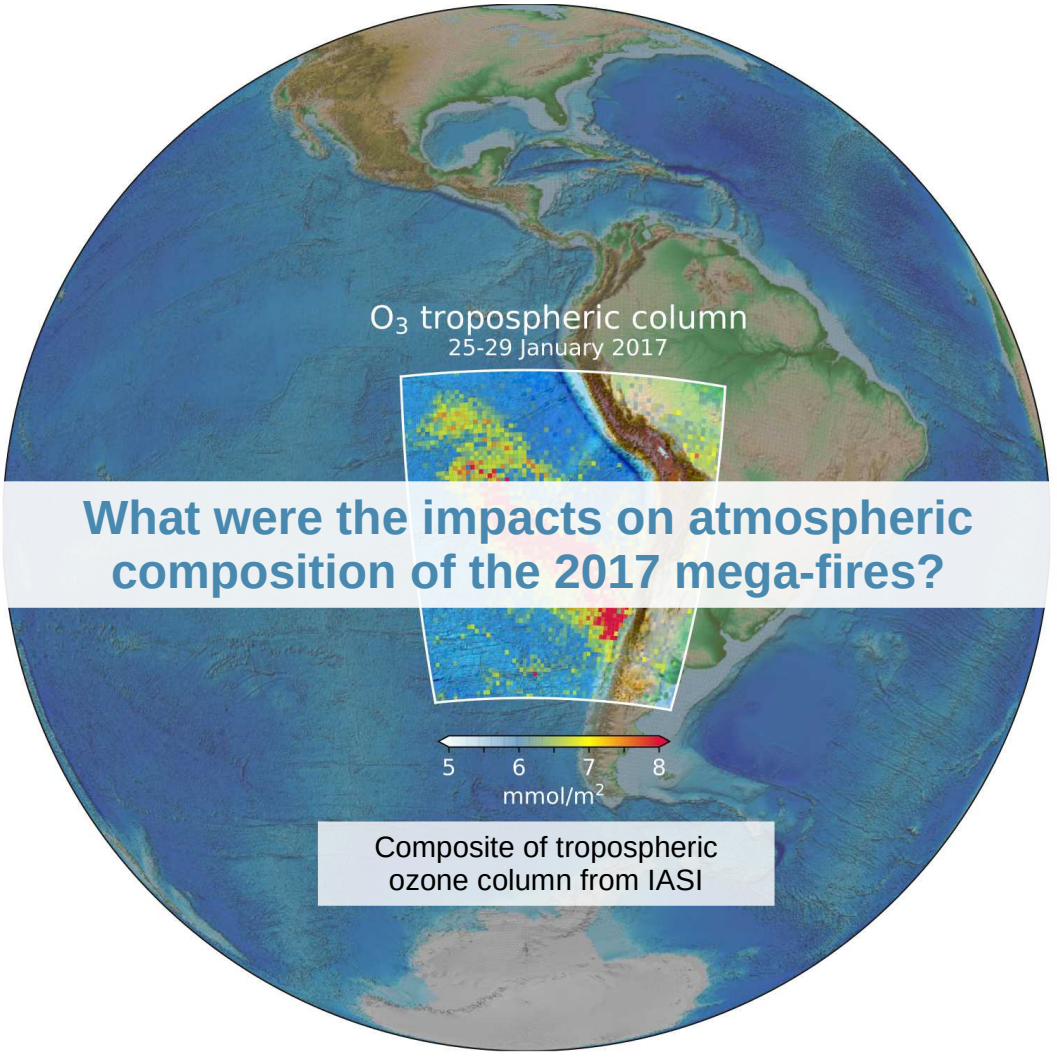
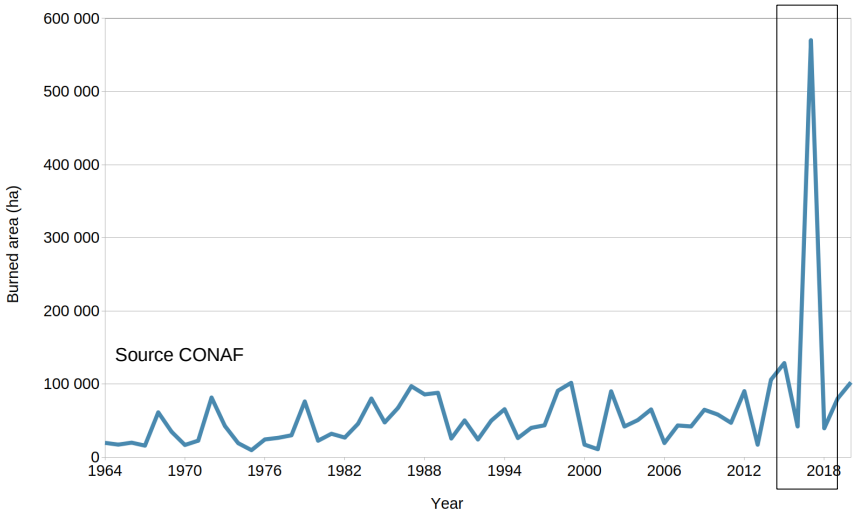


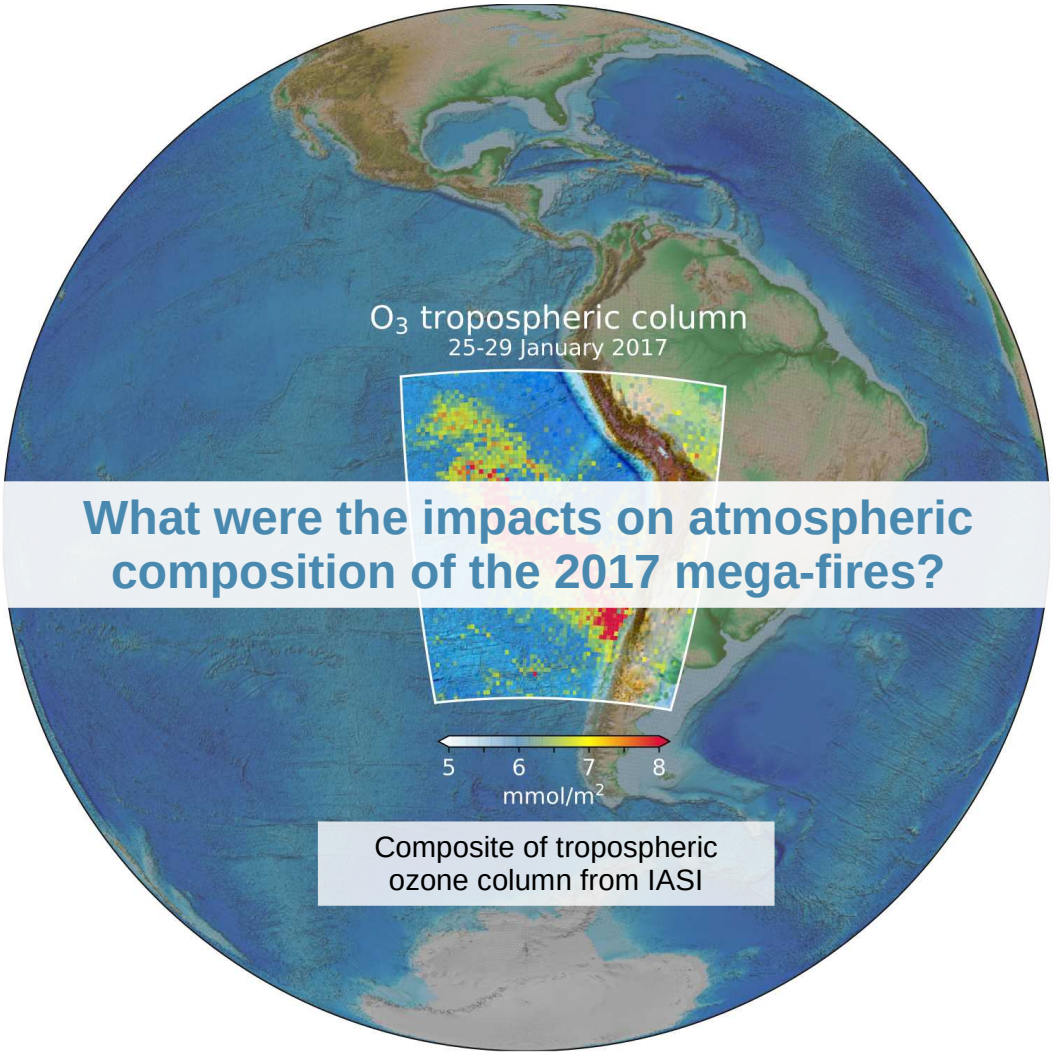


What were the impacts on atmospheric composition of the 2017 mega-fires?

Composite of tropospheric ozone column from IASI





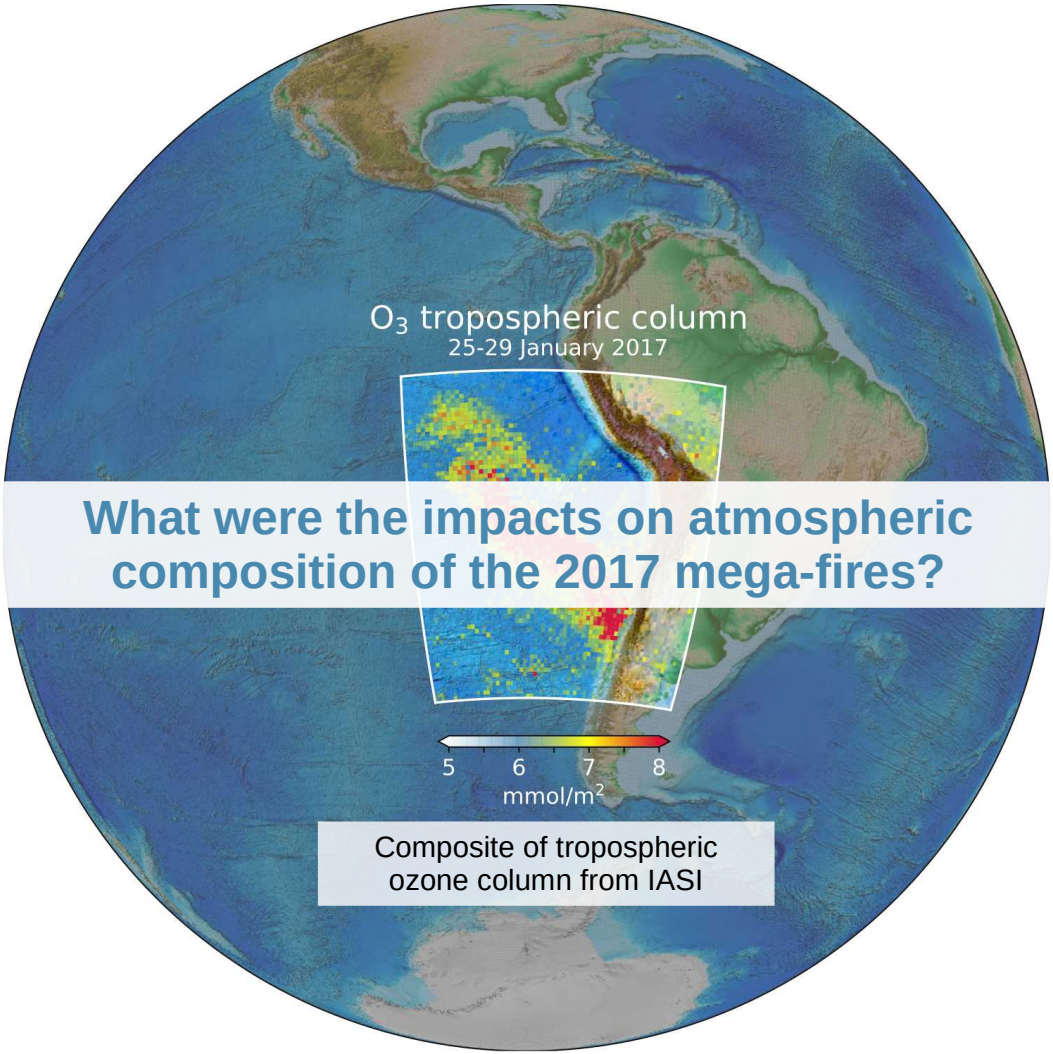


What were the impacts on atmospheric composition of the 2017 mega-fires?

Composite of tropospheric ozone column from IASI

Corrected reflectance from SUOMI on 01-26-2017

100km



Composite of tropospheric ozone column from IASI

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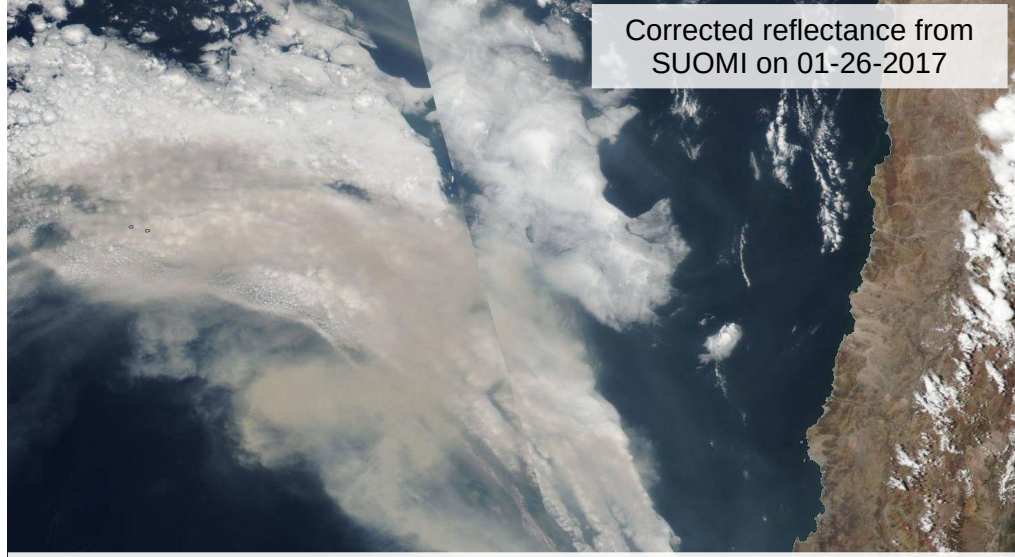


Corrected reflectance from SUOMI on 01-26-2017

Can we estimate the indirect aerosol feedback of this event?



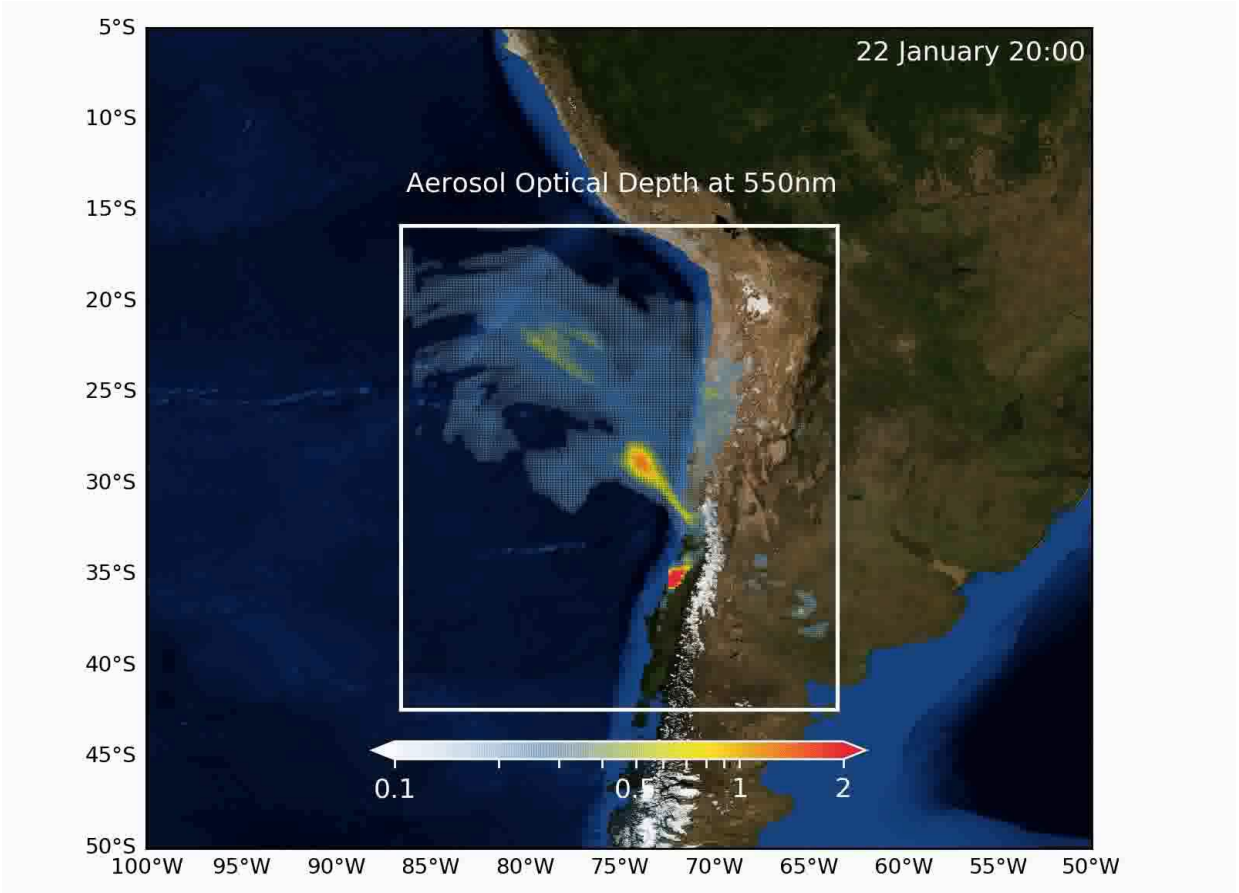


Figure AOD evolution during the fire event as simulated by CHIMERE (time in UTC).

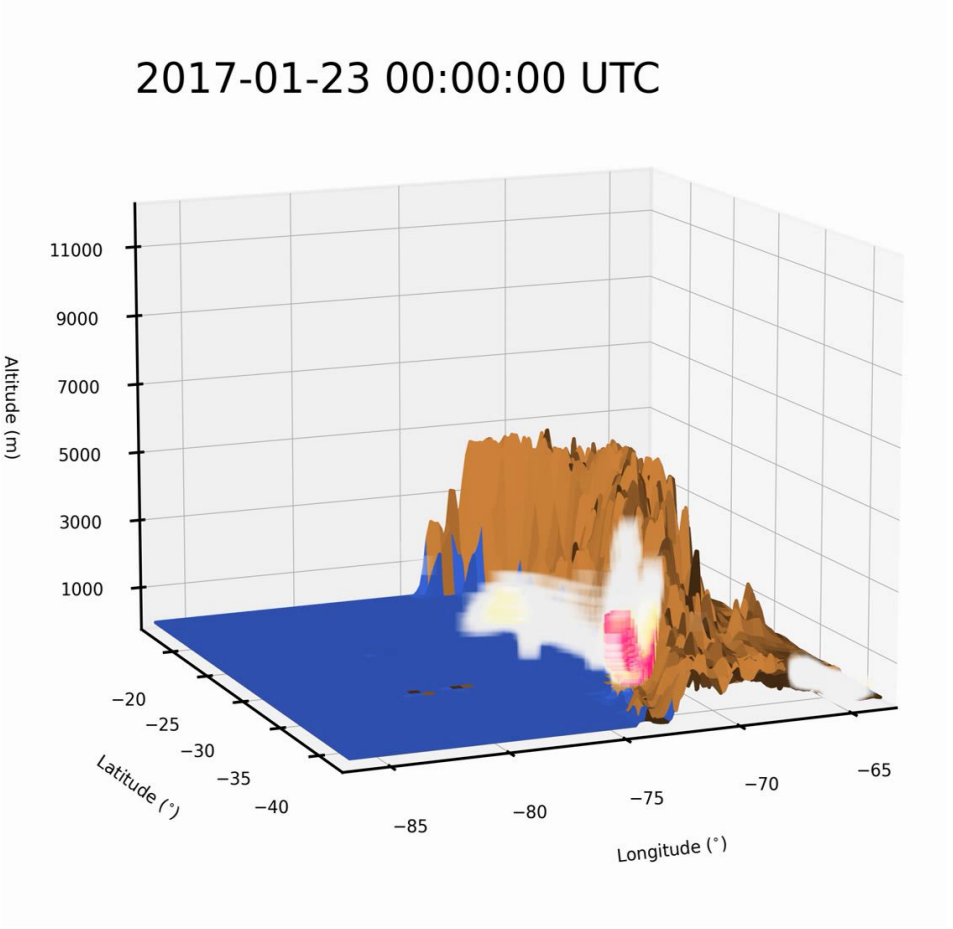
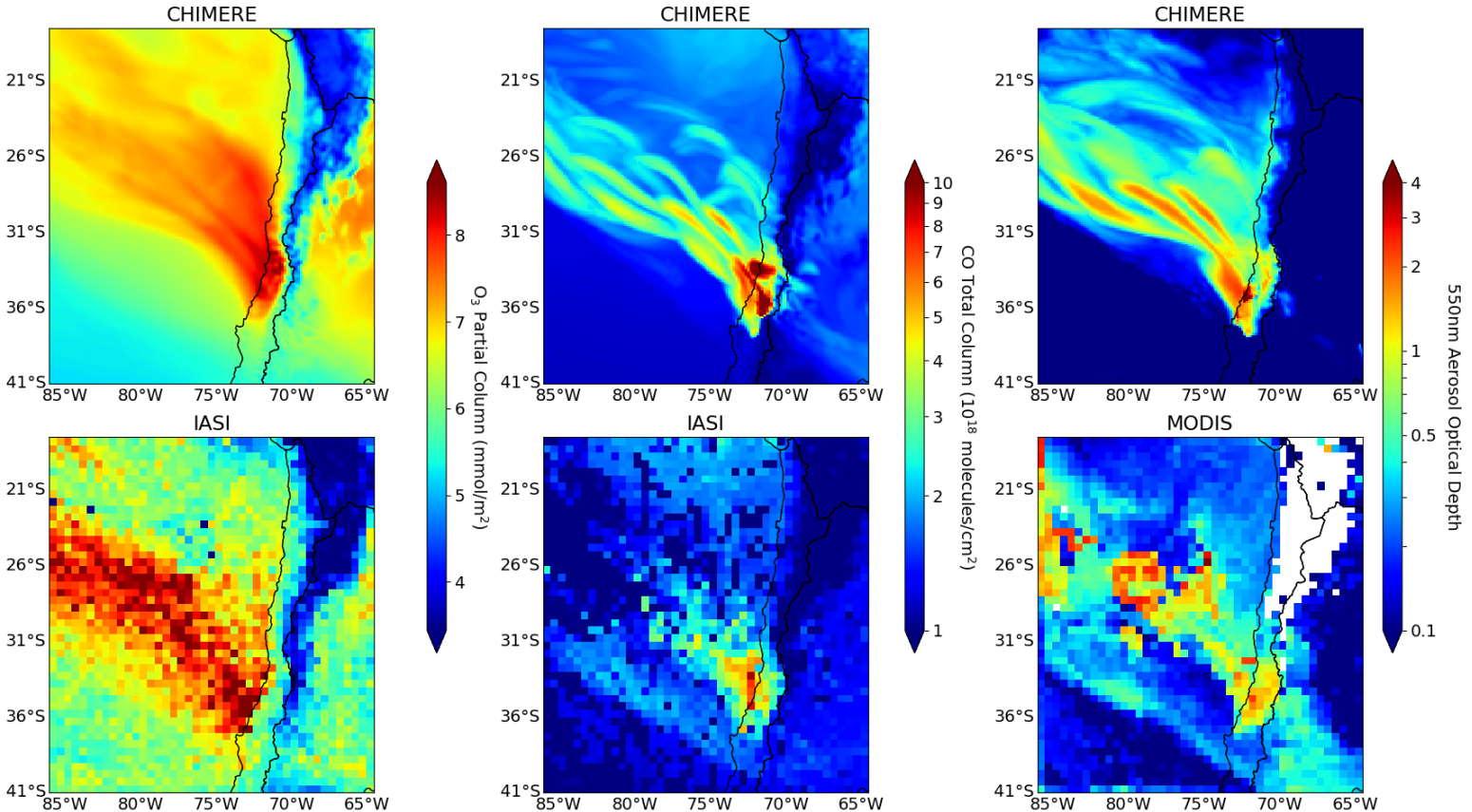


Figure PM_{10} evolution during the fire event as simulated by CHIMERE (time in UTC).

Horizontal plume



- CHIMERE and satellite data offer a similar mean picture of the event both in **spatialization** and **intensity**
- **Increase of $PM_{2.5}$ and CO in Santiago** by 150% (30µg/m^3) and 50% (200ppb), resp.
- Direct **aerosol feedback**
  -100W/m^2 radiation
  -1°C surface temperature
  -100m PBL height

Figure Comparison of the simulated horizontal plume with satellite data. Composite for January 25 to 29.

Vertical plume

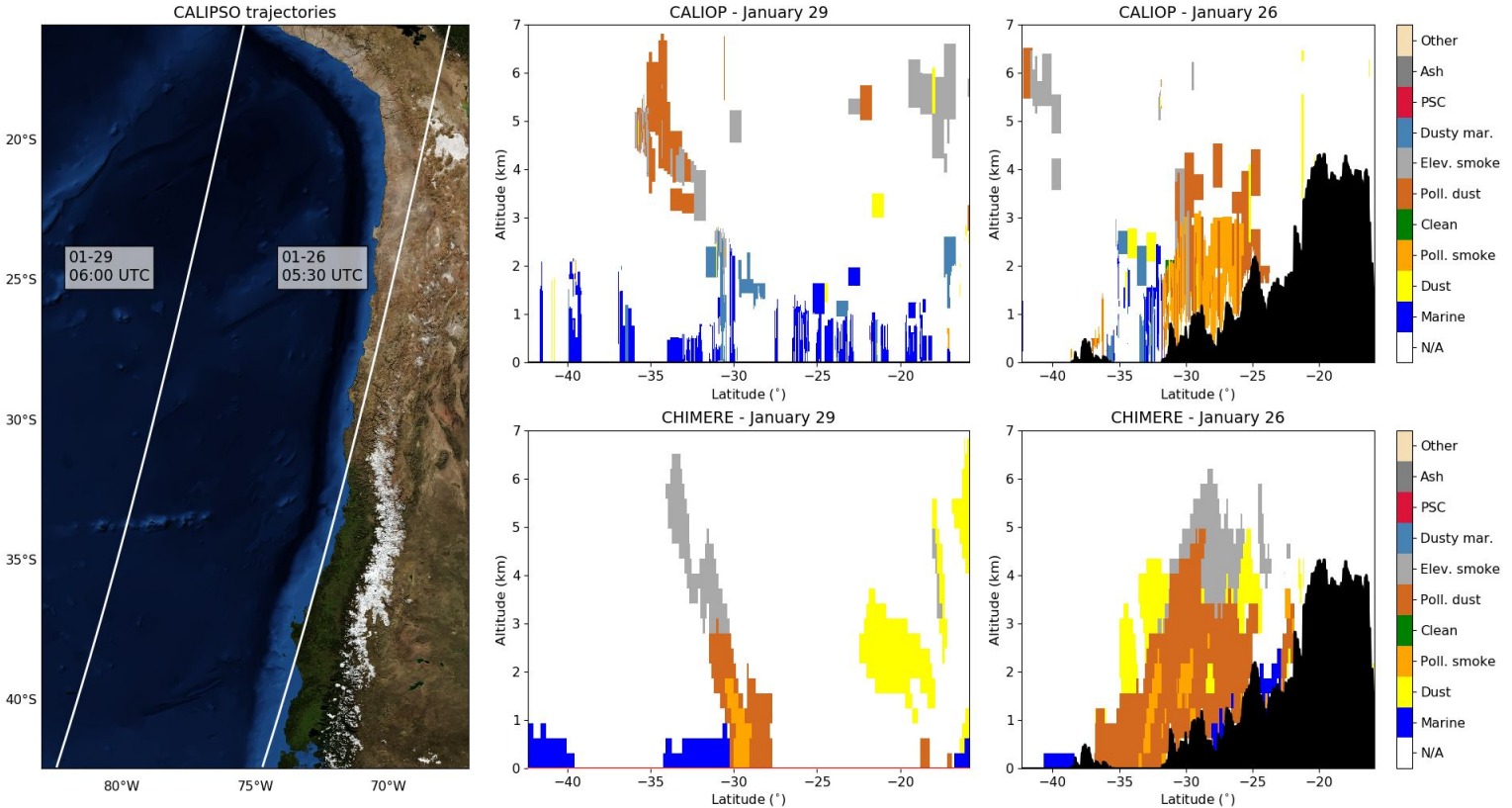
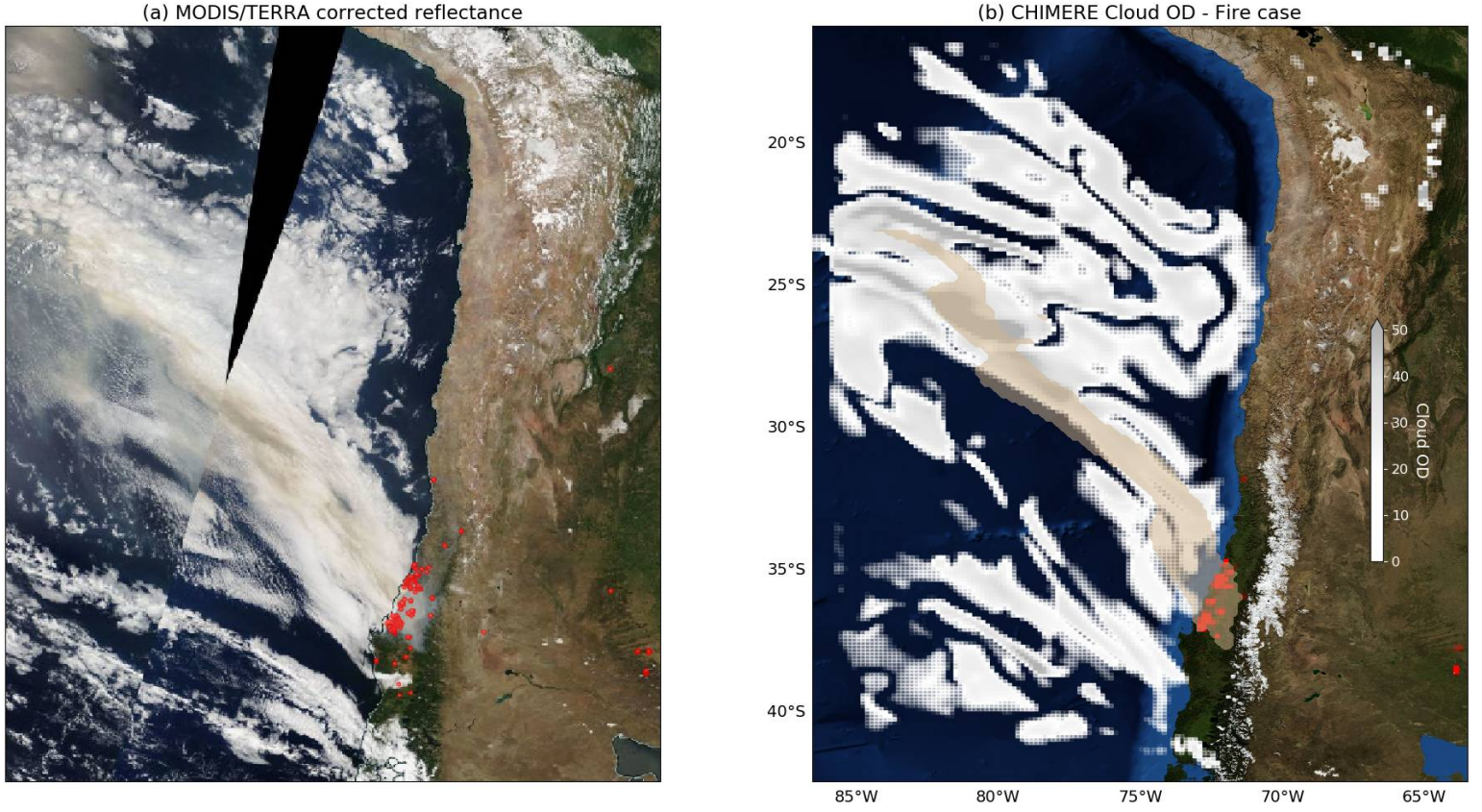


Figure Comparison of the CHIMERE simulated vertical plume composition with CALIOP for 2 trajectories.

- Vertical transects are in agreement with CALIOP
- Injection of aerosols up to **7km in altitude**

Cloud feedback



- Reasonable reproduction of cloud cover and smoke plume

Figure Fire locations, cloud cover and main smoke plume as seen by (a) MODIS and (b) CHIMERE. January 27

Cloud feedback

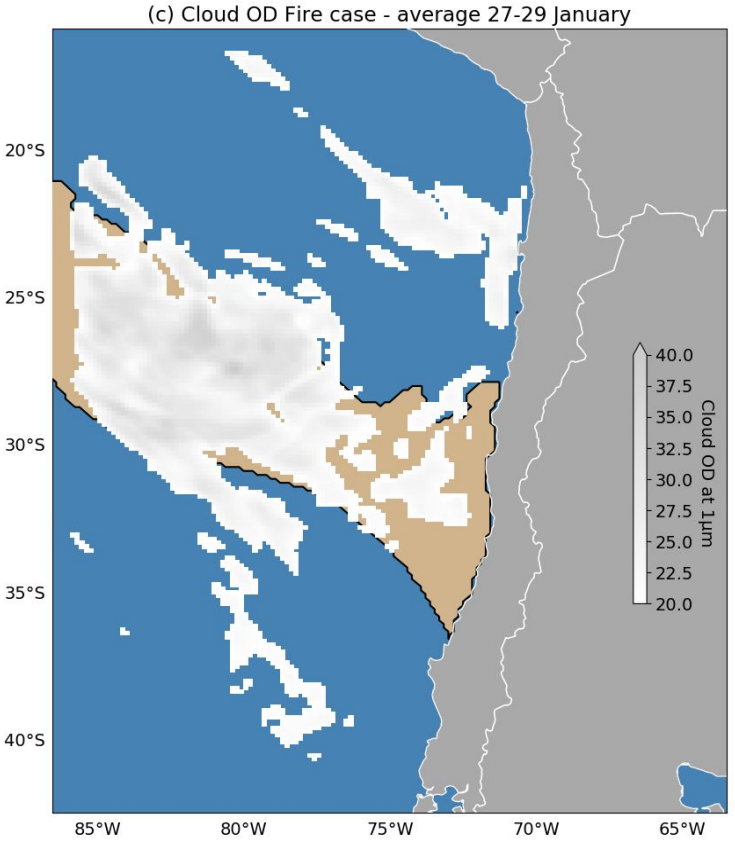


Figure Cloud cover during the fire event (left) and effect of fire emissions on cloud optical depth (right)

Cloud feedback

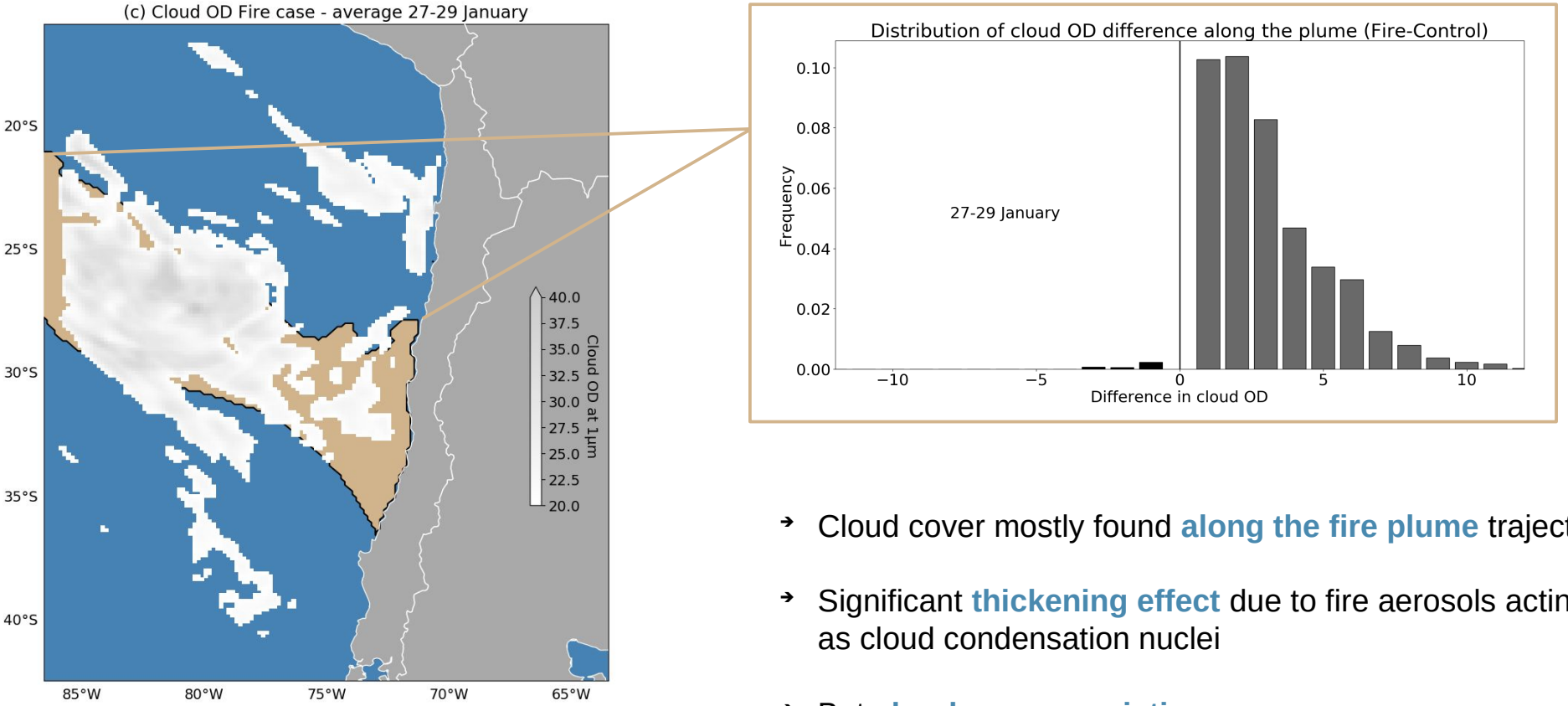
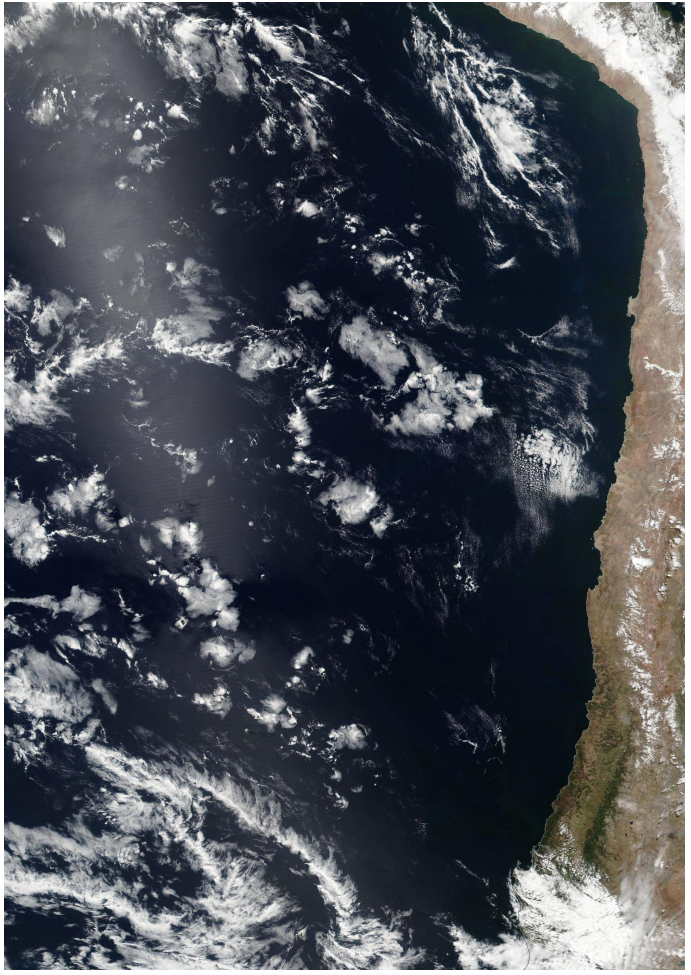


Figure Cloud cover during the fire event (left) and effect of fire emissions on cloud optical depth (right)

- Cloud cover mostly found **along the fire plume** trajectory
- Significant **thickening effect** due to fire aerosols acting as cloud condensation nuclei
- But **clouds are preexisting**

Initial working hypothesis



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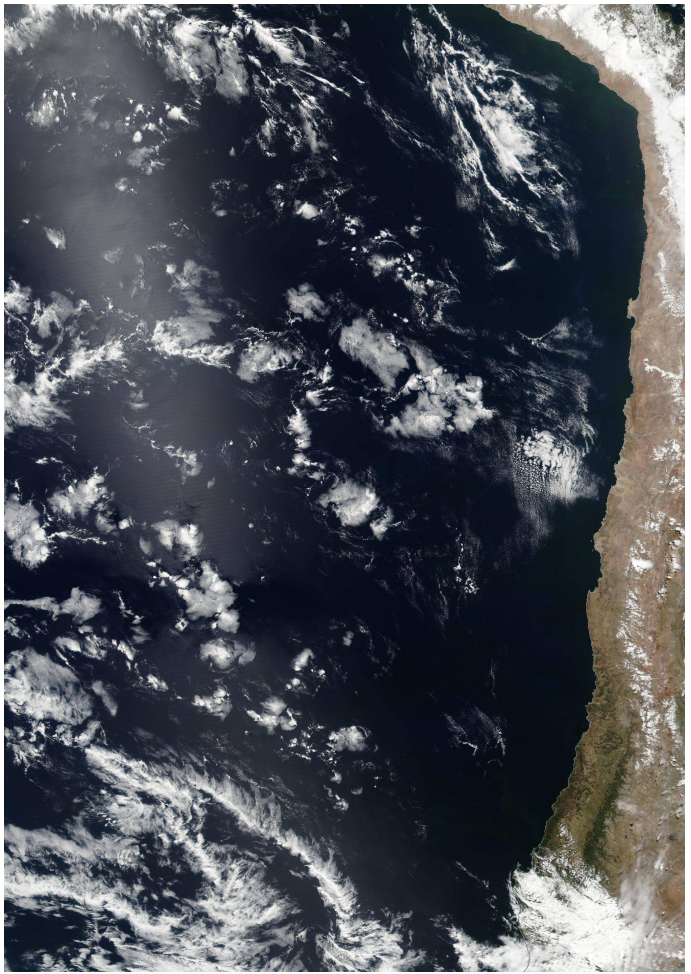
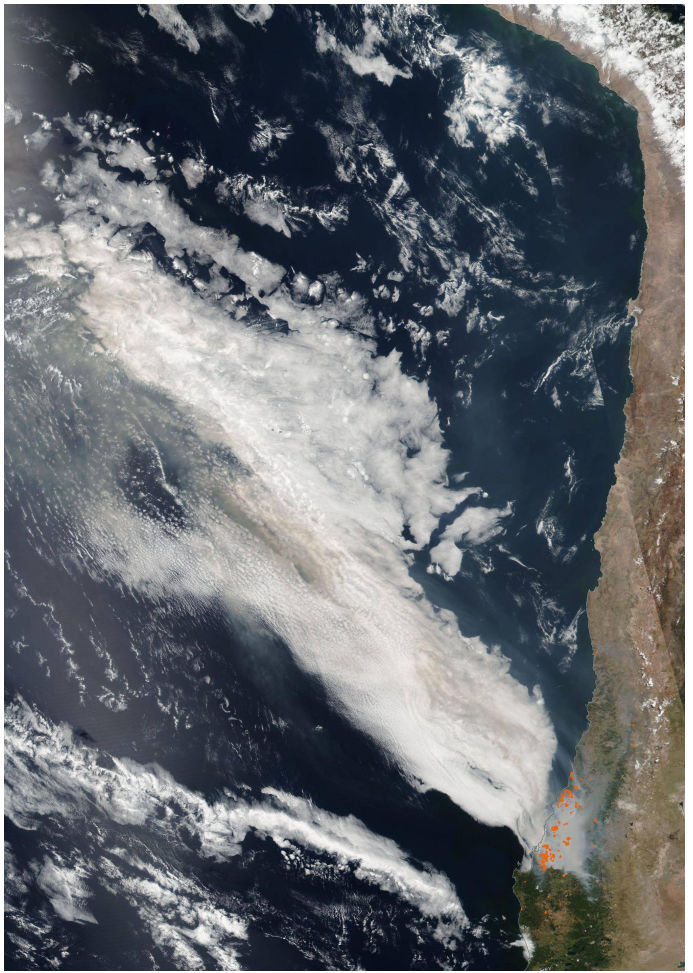
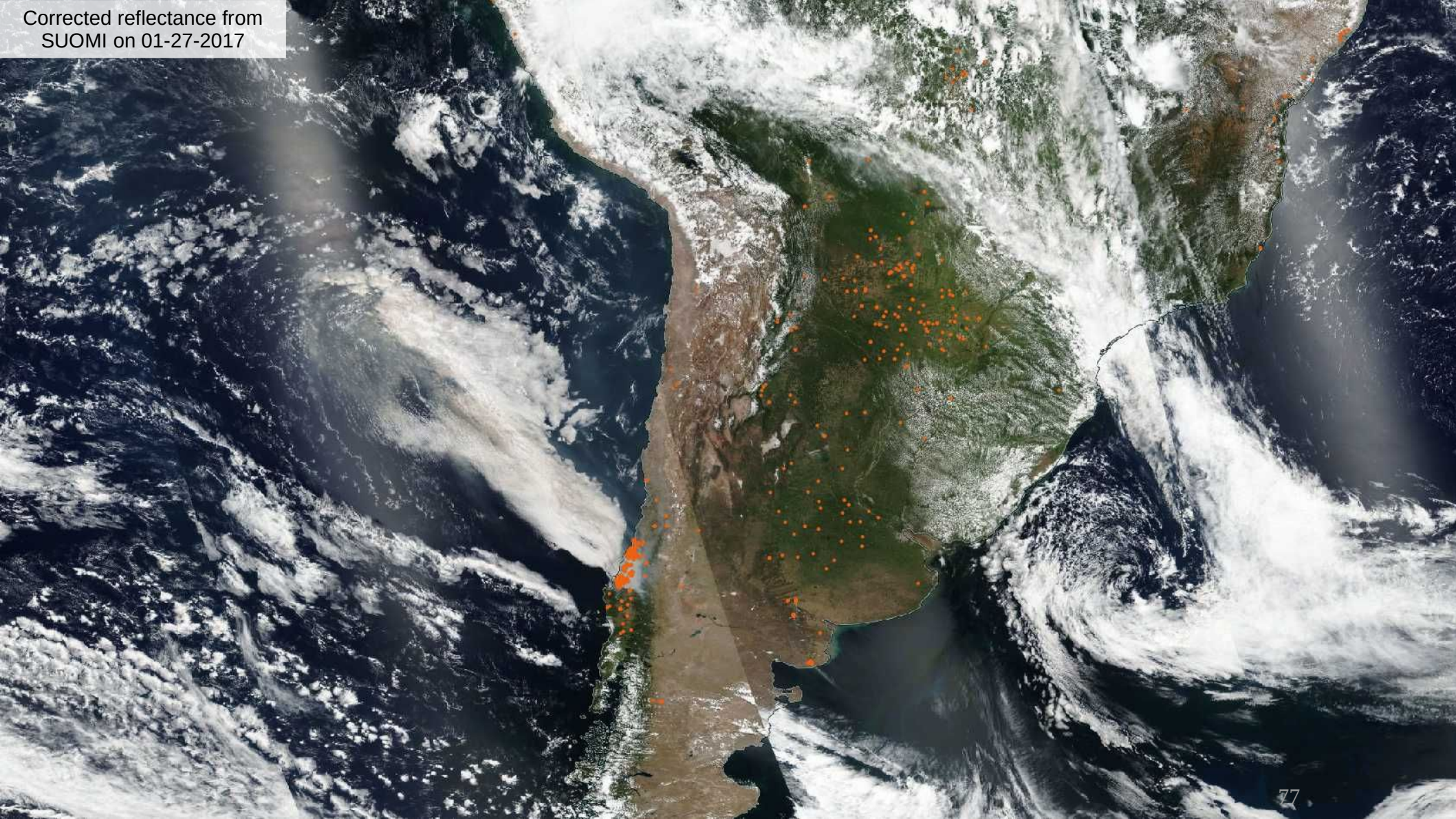


+ FIRES =>

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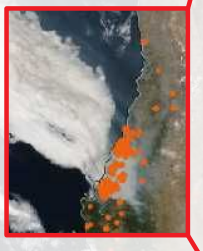
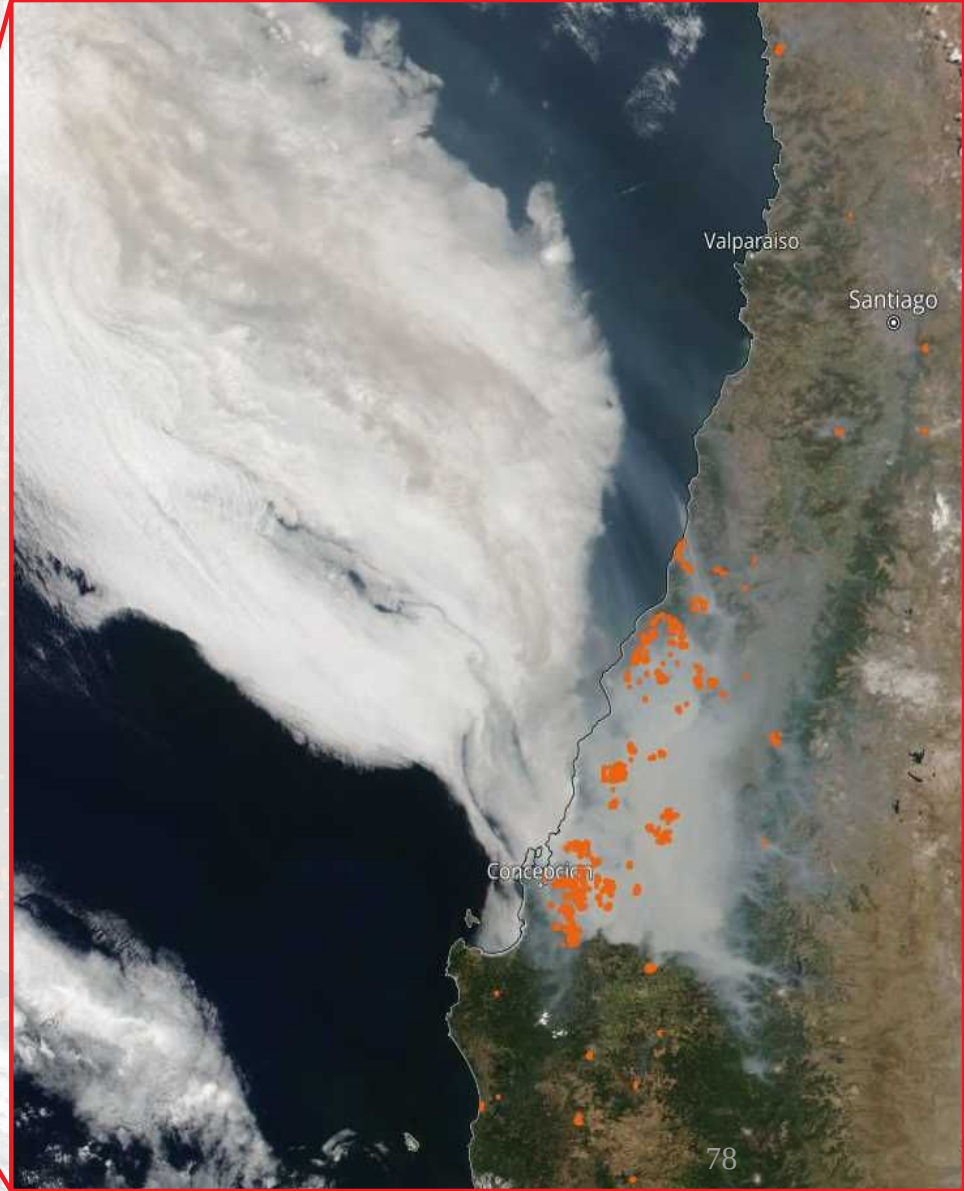
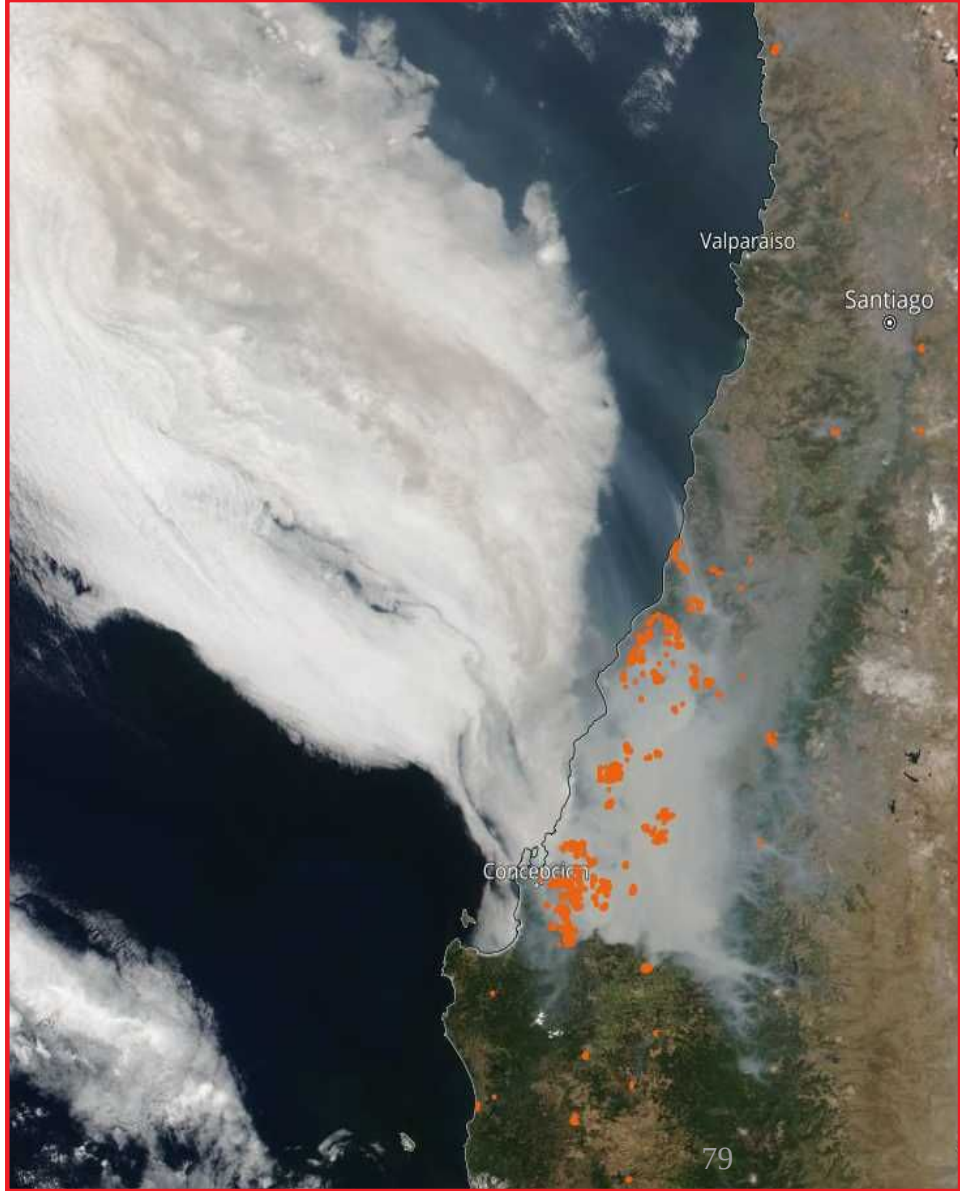




Why not the **entire** cloud formation?



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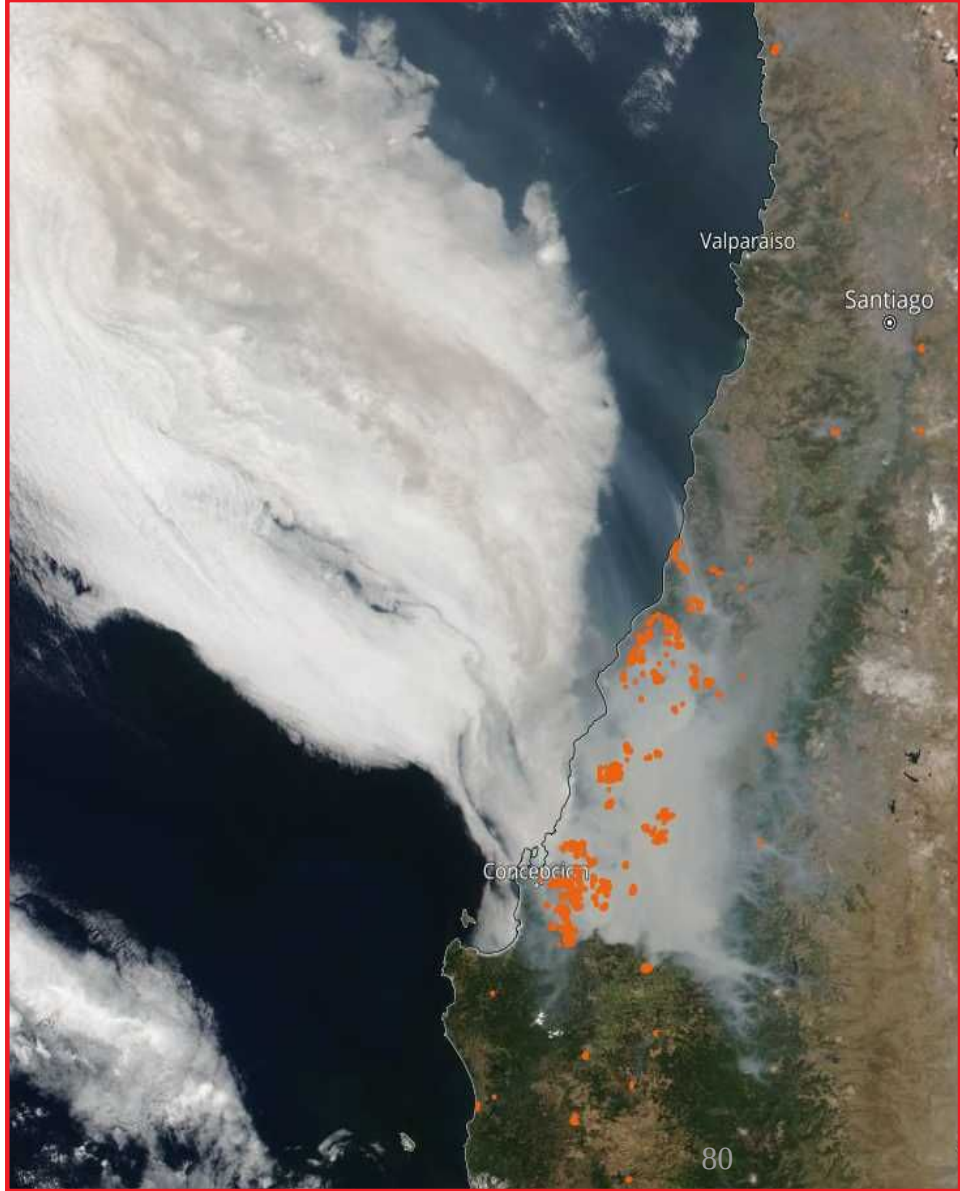
Raises the question: can we **properly study** such a feedback?

(i) Aerosol indirect effect is not perfectly modeled



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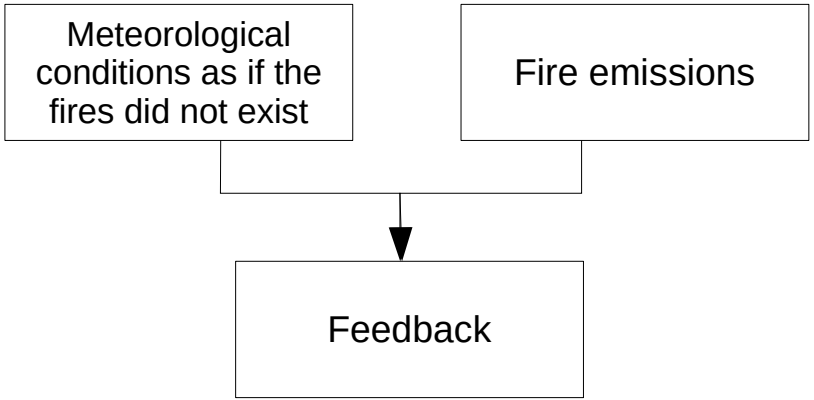

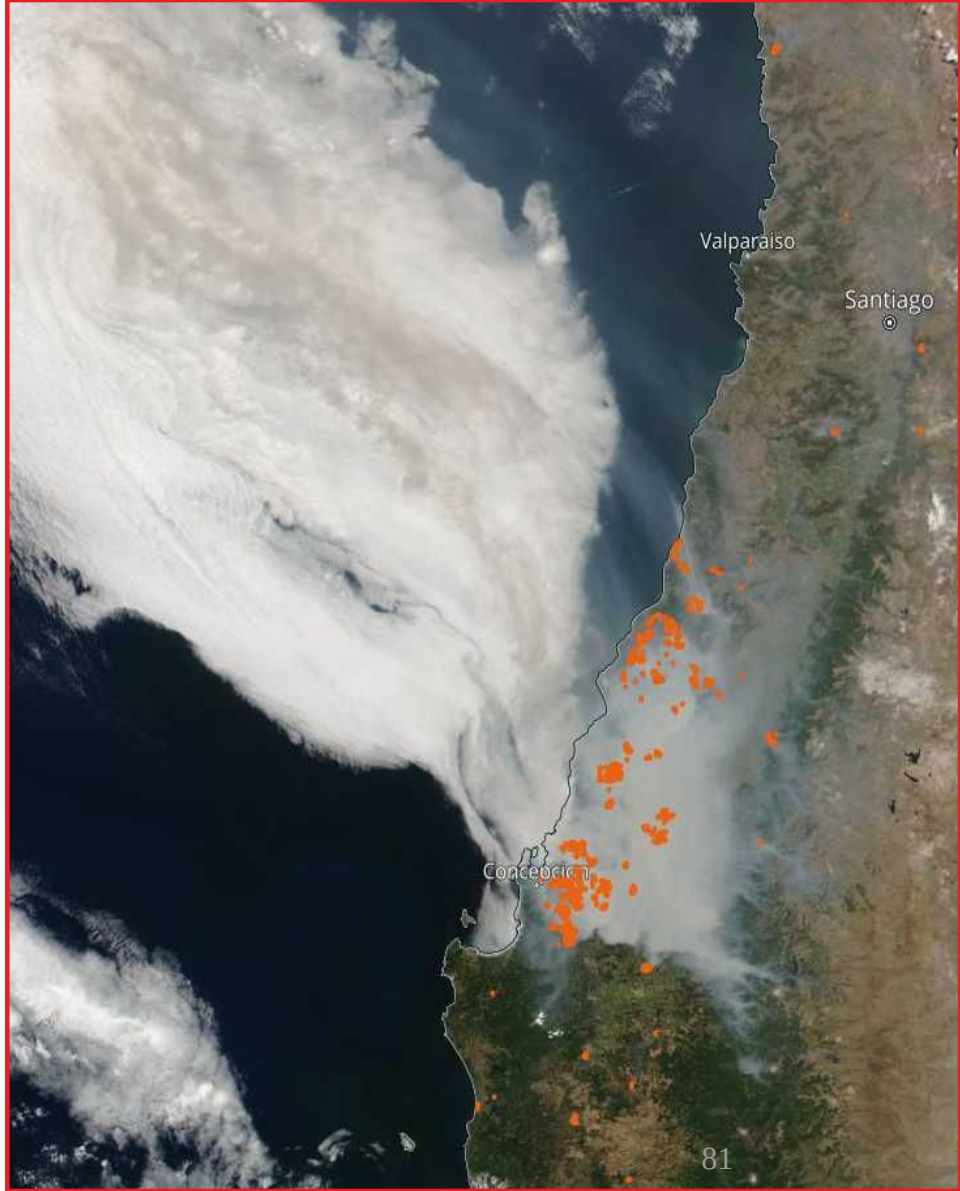

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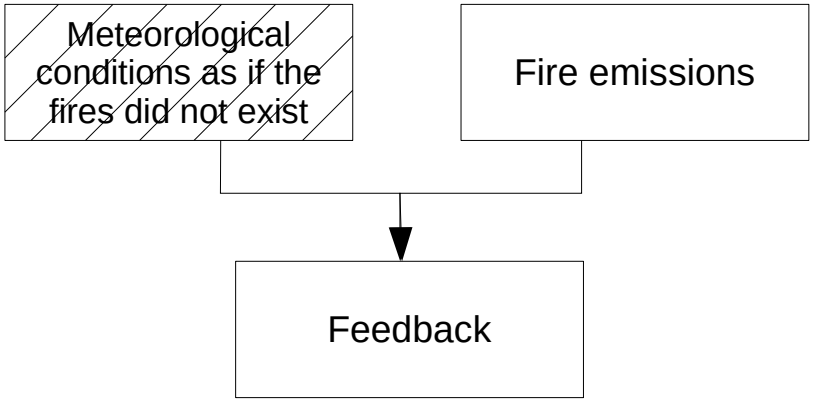
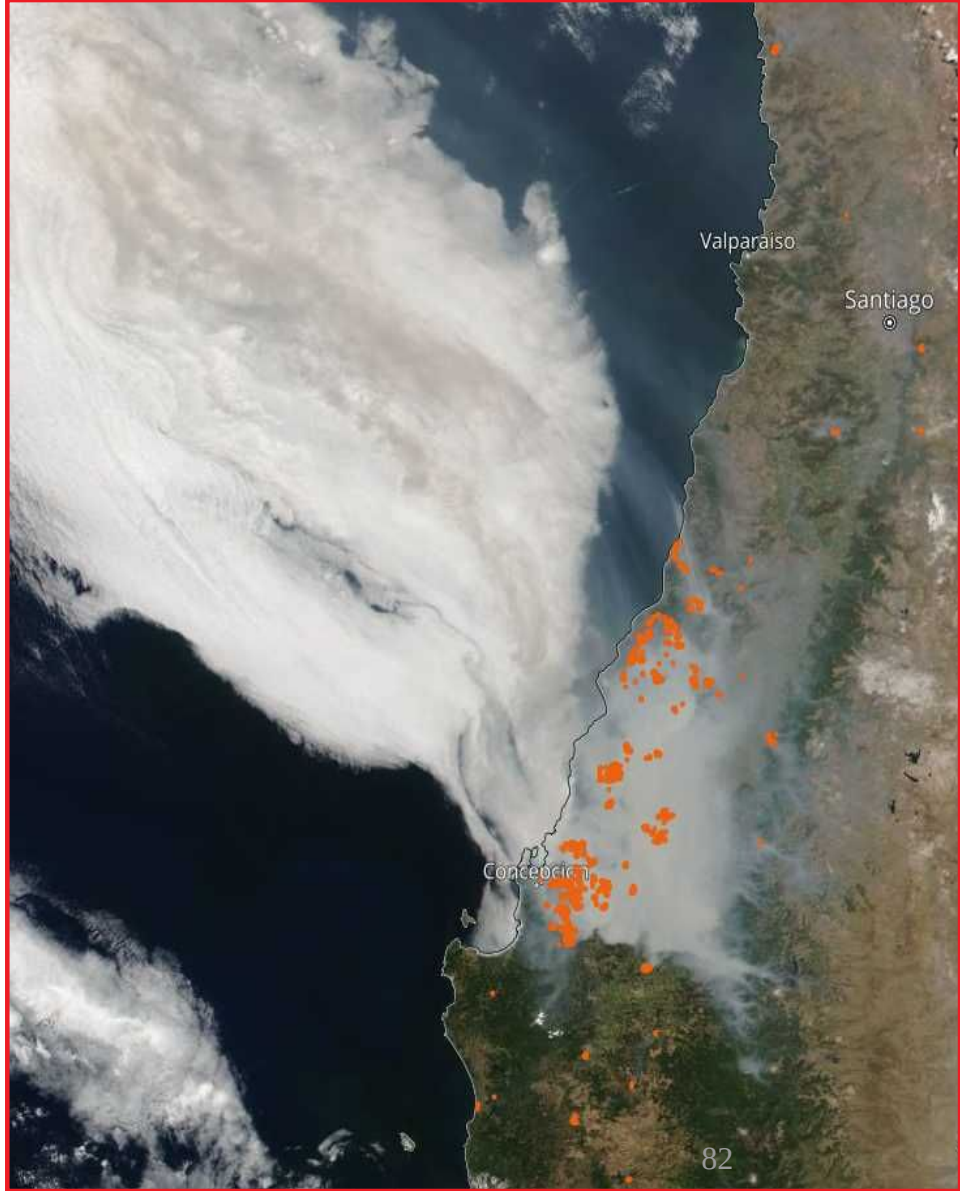
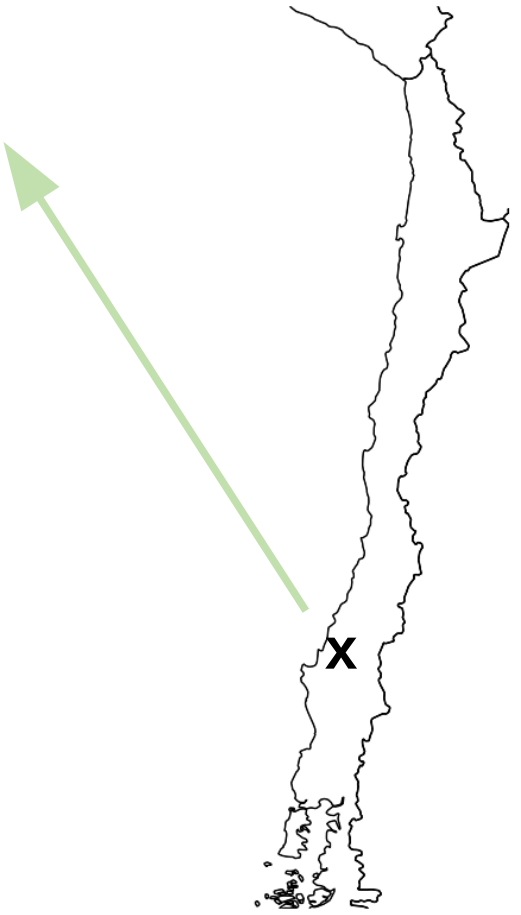




Conclusions

>2000km

plume from the 2017 mega-fires



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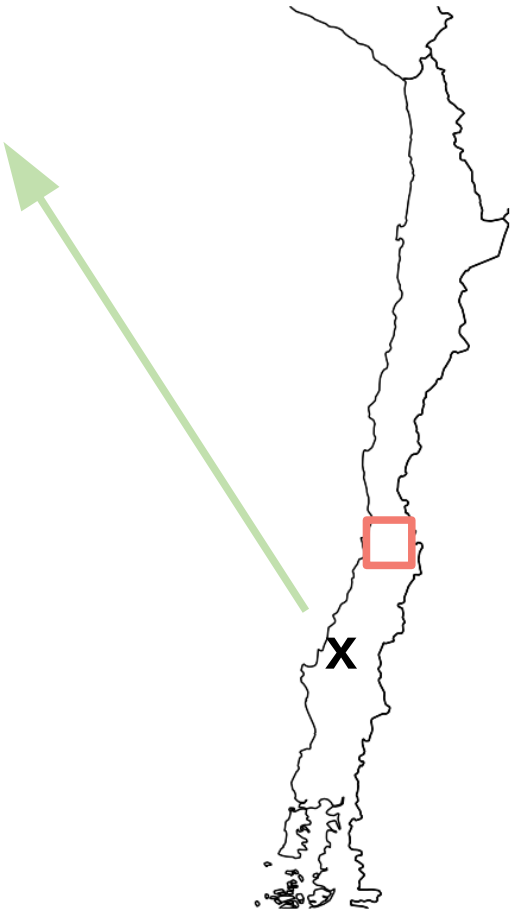
$PM_{2.5}$ in Santiago



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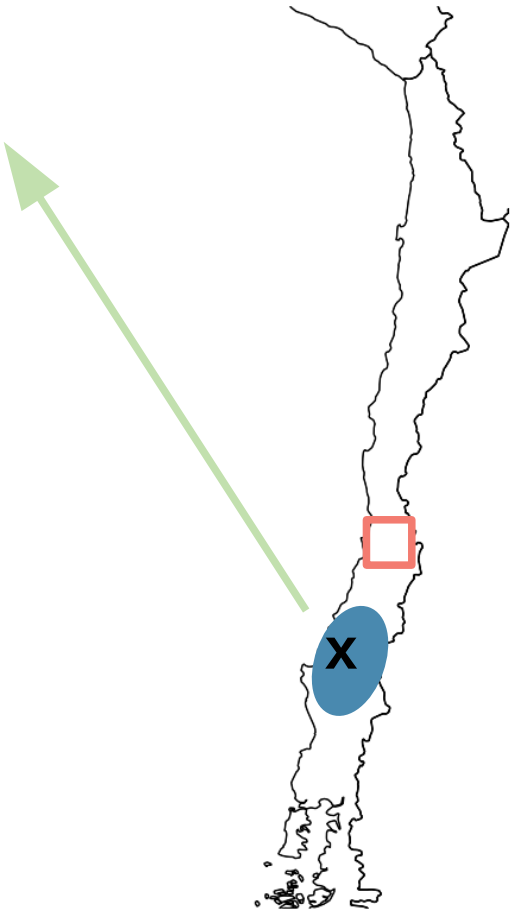

Conclusions

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1°C

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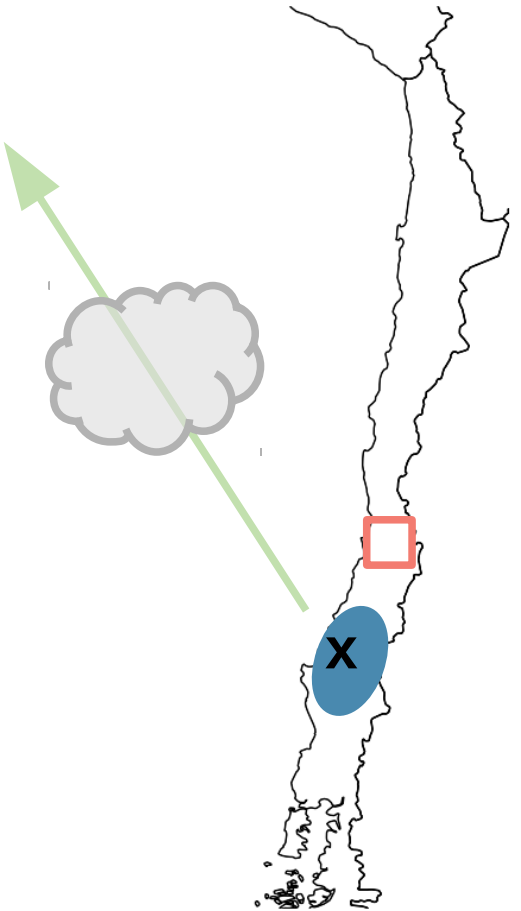
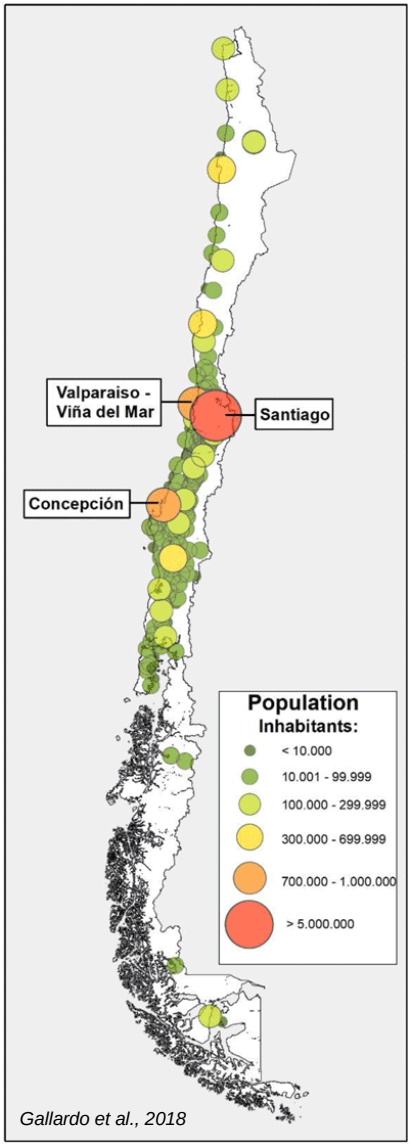
Cloud feedback

significant but hard to properly assess

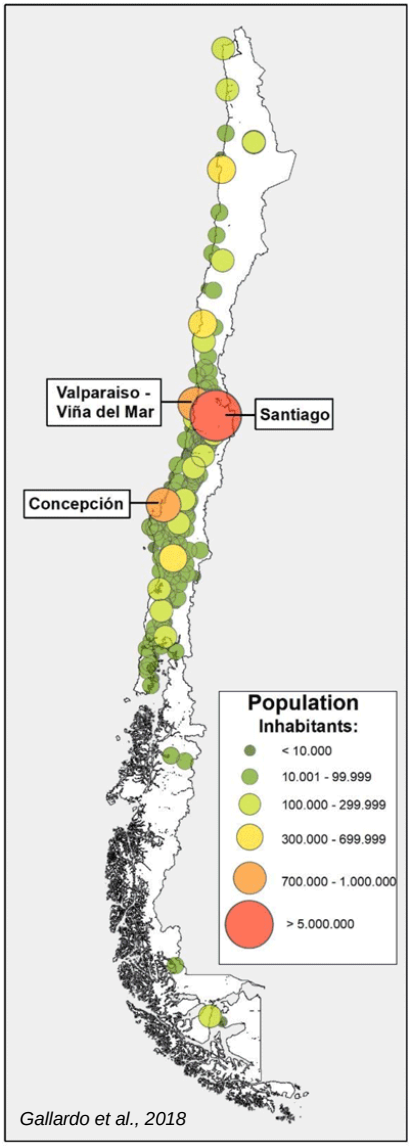


2 – Regional transport of air pollutants in central Chile

(Lapere et al., 2021c)



Gallardo et al., 2018



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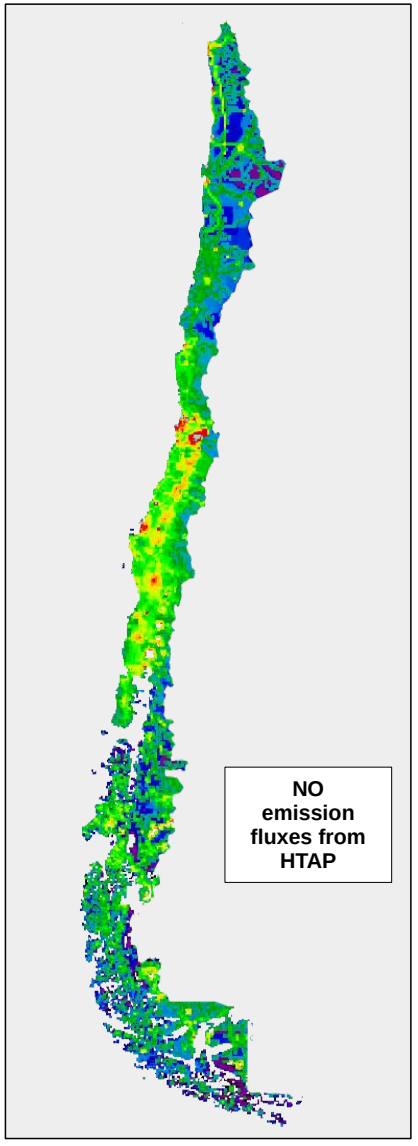

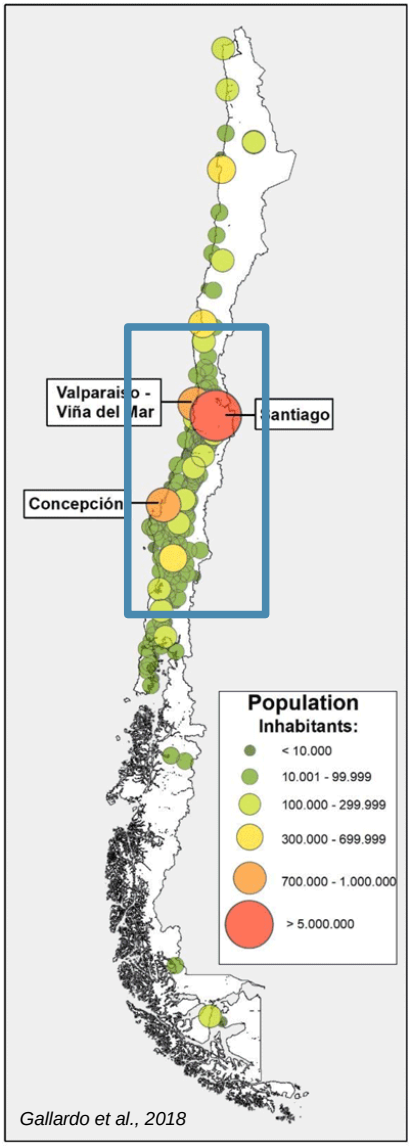

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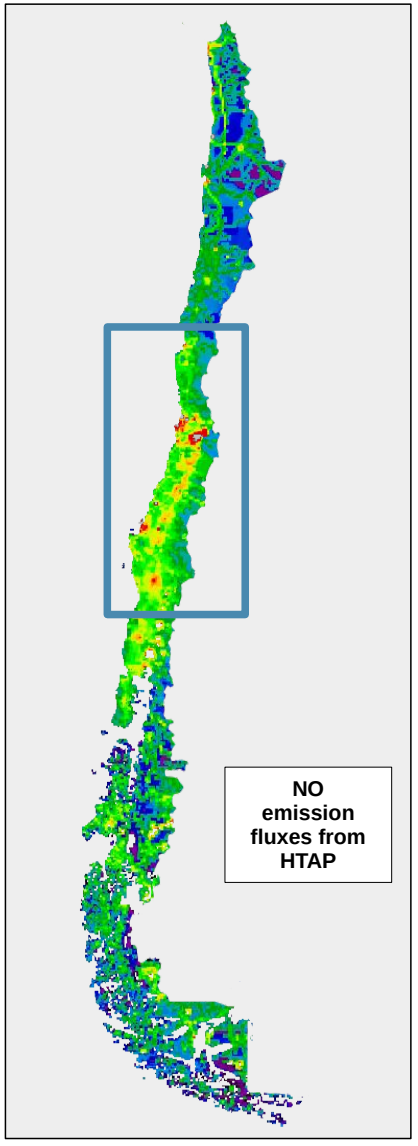


Where and how do air pollutants transport across central Chile in summer and winter?

Seasonal transport – effect on surface concentrations

Figure Relative contribution of Santiago emissions to surface mean $PM_{2.5}$ – **Winter**

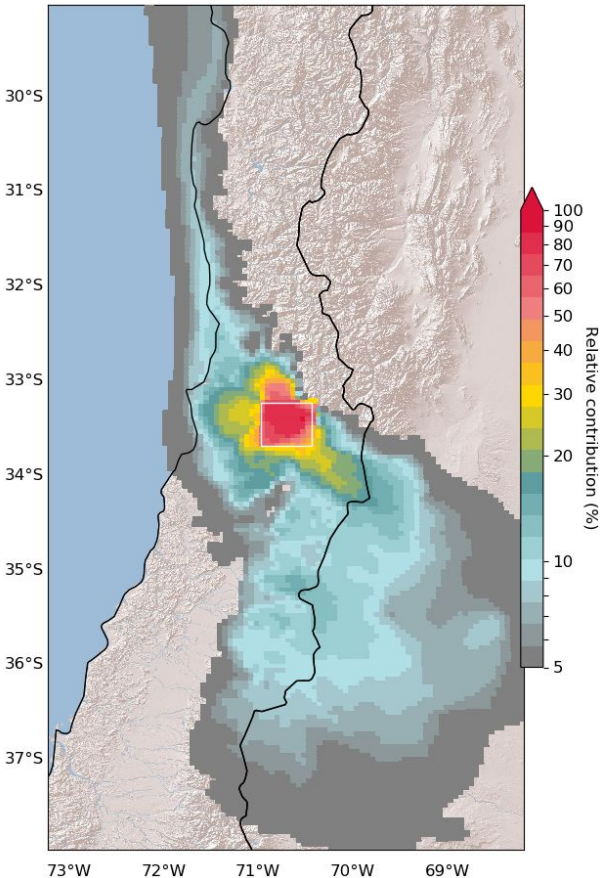
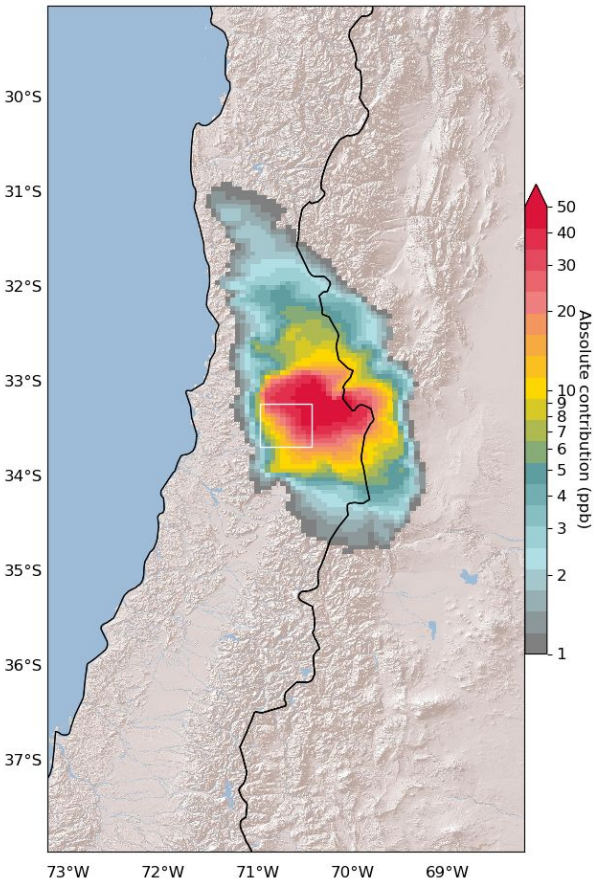
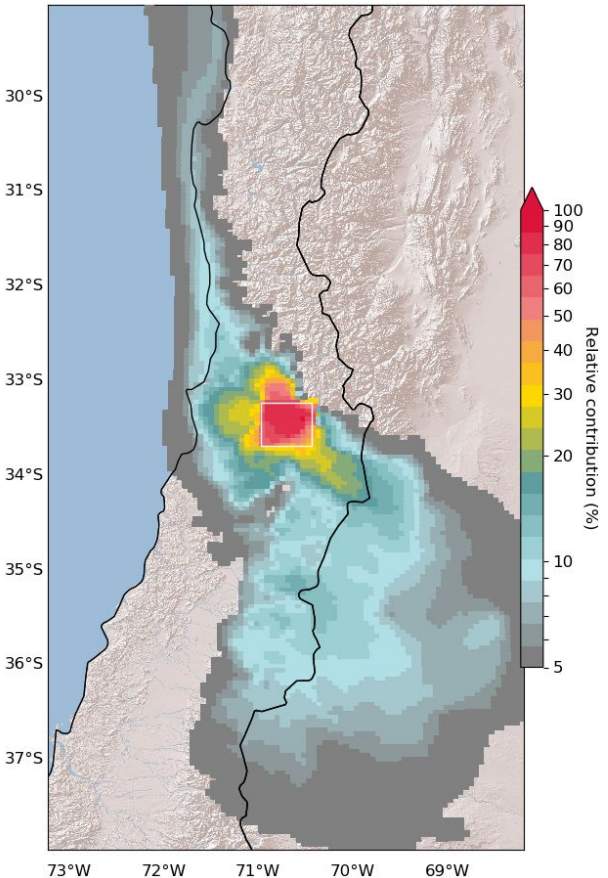


Figure Absolute contribution of Santiago emissions to surface daily max O_3 – **Summer**



Seasonal transport – effect on surface concentrations

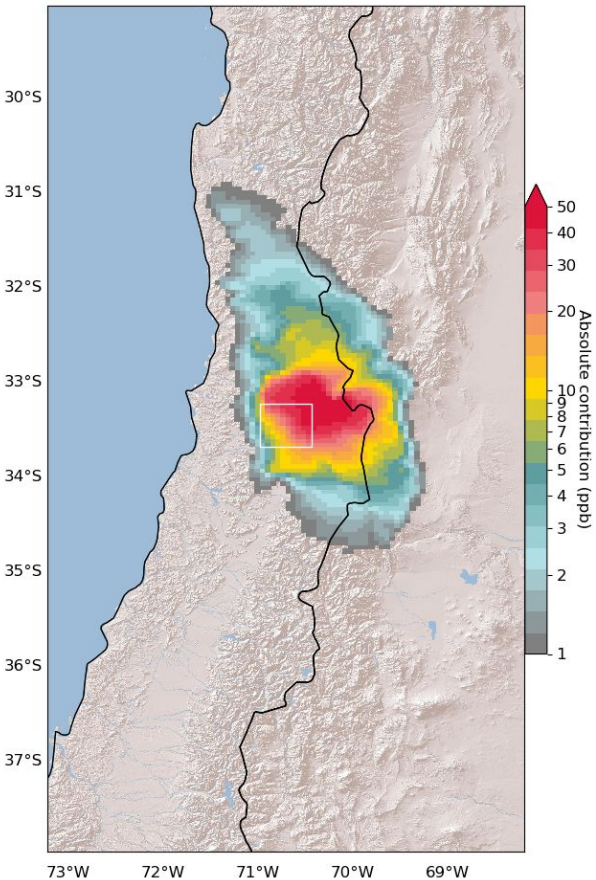
Figure Relative contribution of Santiago emissions to surface mean $PM_{2.5}$ – **Winter**



Santiago emissions are responsible for:

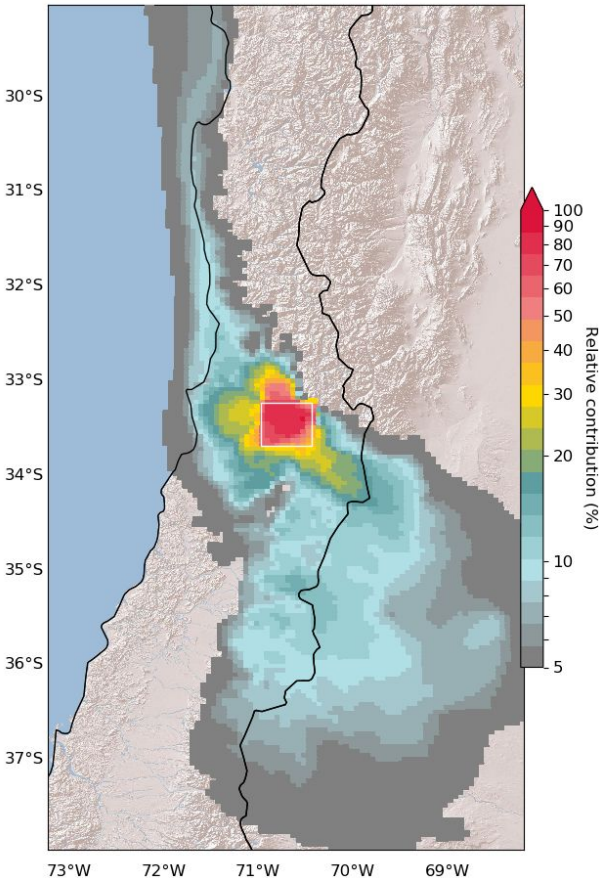
- A plume of $PM_{2.5}$ **extending over 1000km**
- Ozone peaks **in the Andes** as far as Argentina

Figure Absolute contribution of Santiago emissions to surface daily max O_3 – **Summer**



Seasonal transport – effect on surface concentrations

Figure Relative contribution of Santiago emissions to surface mean $PM_{2.5}$ – **Winter**



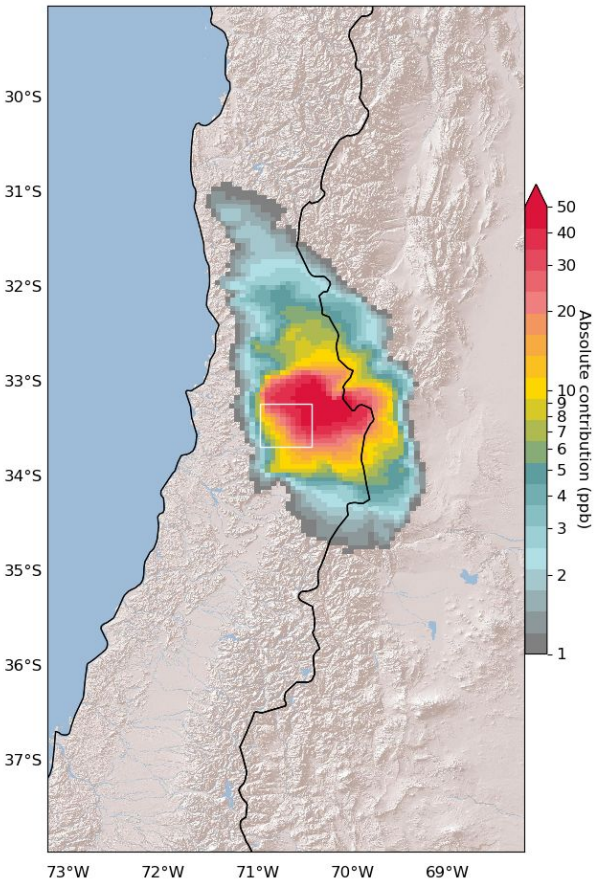
Santiago emissions are responsible for:

- A plume of $PM_{2.5}$ **extending over 1000km**
- Ozone peaks **in the Andes** as far as Argentina

In turn, **regional emissions** account for:

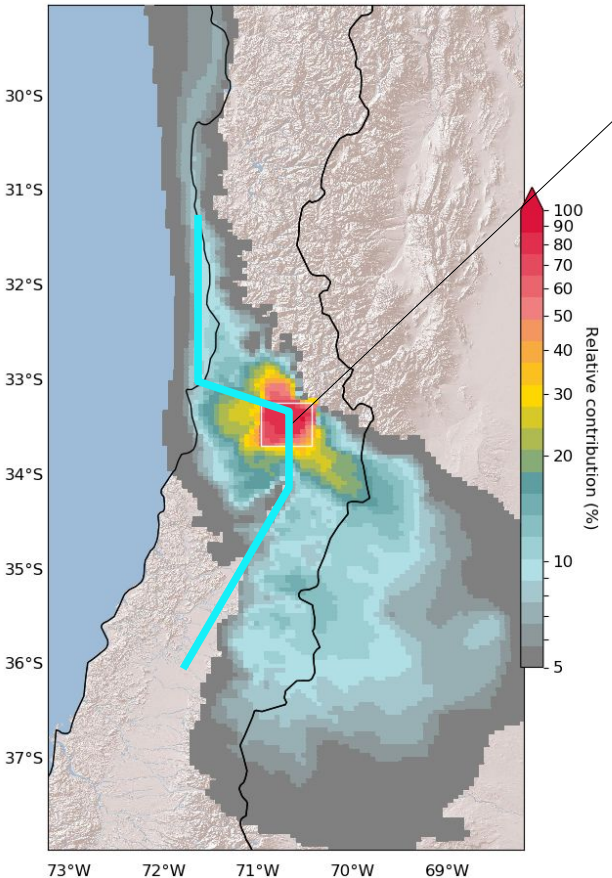
- **13 to 15% of $PM_{2.5}$** in Santiago in winter
- **No contribution to Ozone** formation in summer

Figure Absolute contribution of Santiago emissions to surface daily max O_3 – **Summer**



Seasonal transport – advection processes

Figure Relative contribution of Santiago emissions to surface mean $PM_{2.5}$ – **Winter**



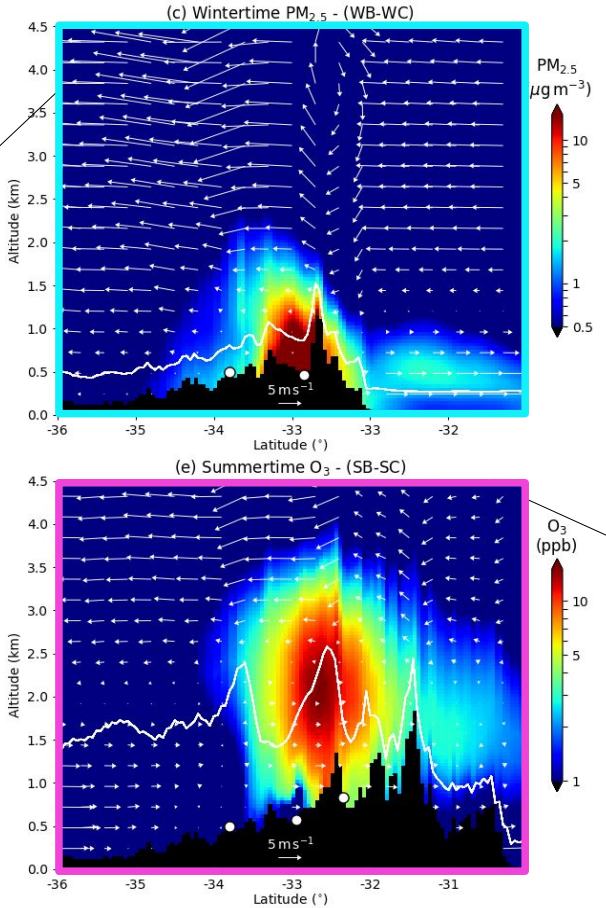
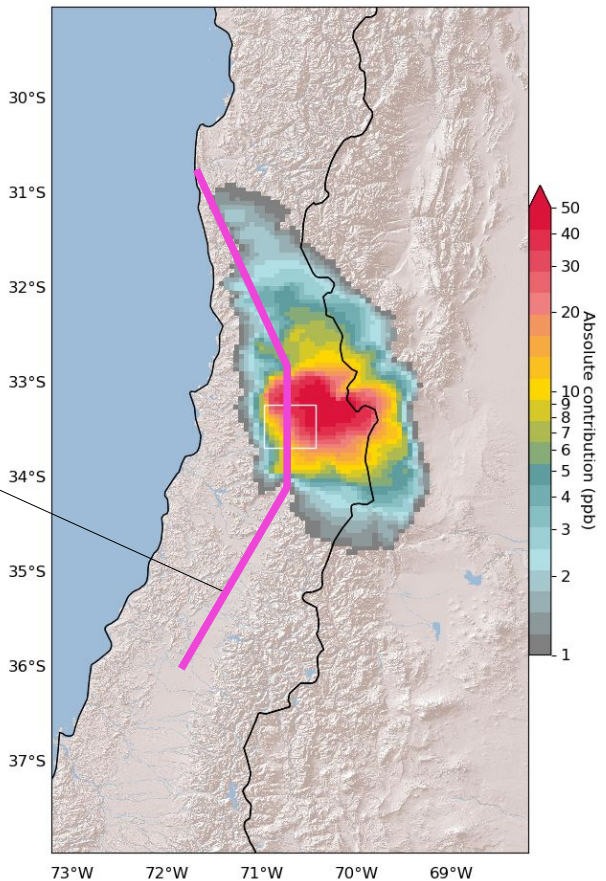


Figure Mean transport of Santiago emissions. White line in (c) and (e) is the average PBLH.

Figure Absolute contribution of Santiago emissions to surface daily max O_3 – **Summer**



Seasonal transport – advection processes

- Wintertime PM transport is contained **within the boundary layer**, except above the **ocean** where it is injected into the free troposphere

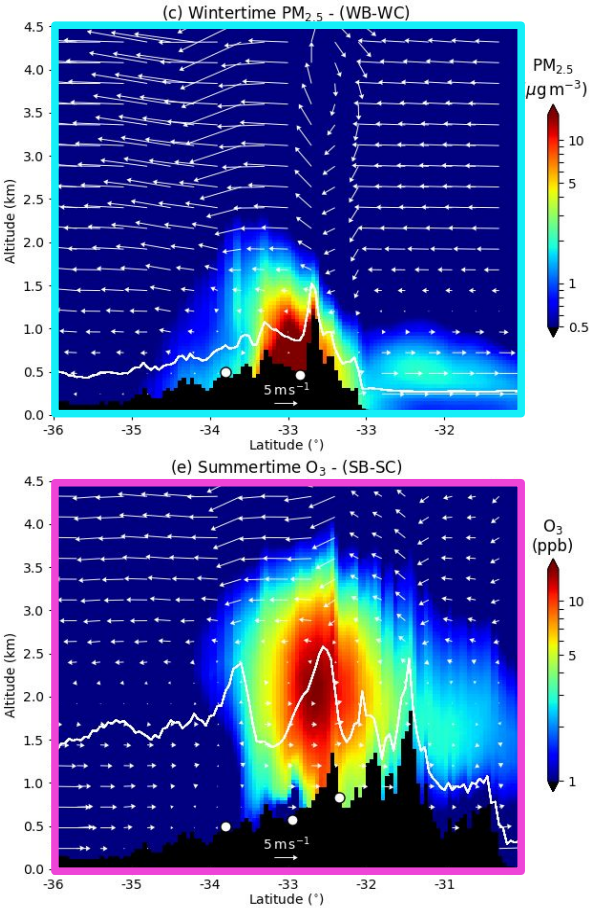


Figure Mean transport of Santiago emissions. White line in (c) and (e) is the average PBLH.

- Ozone export from Santiago creates a **persistent bubble in altitude**

Ozone chemistry and mountain dynamics

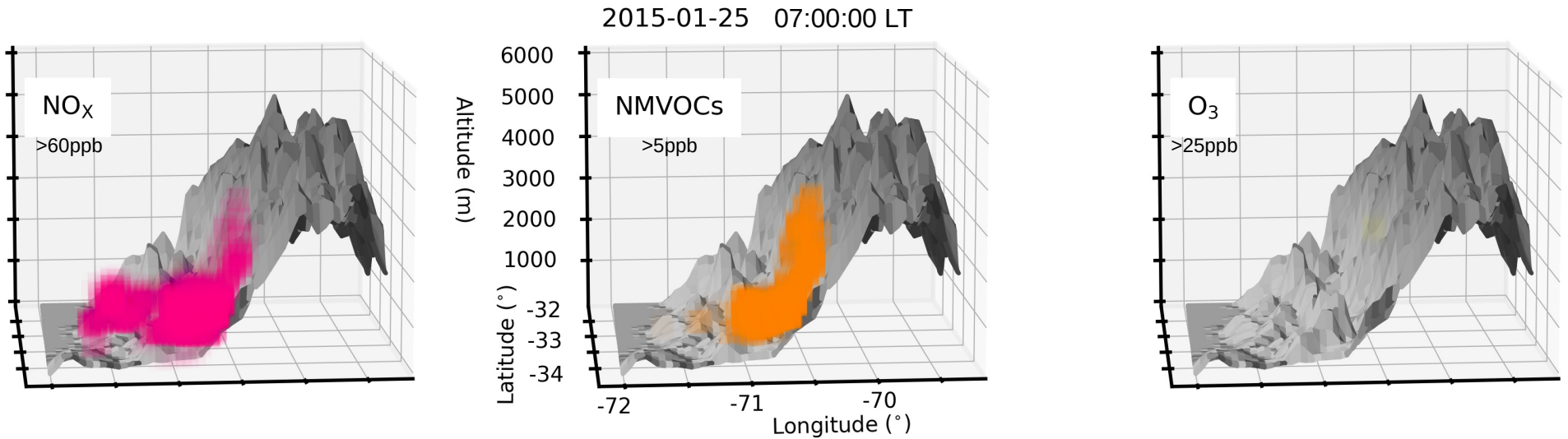
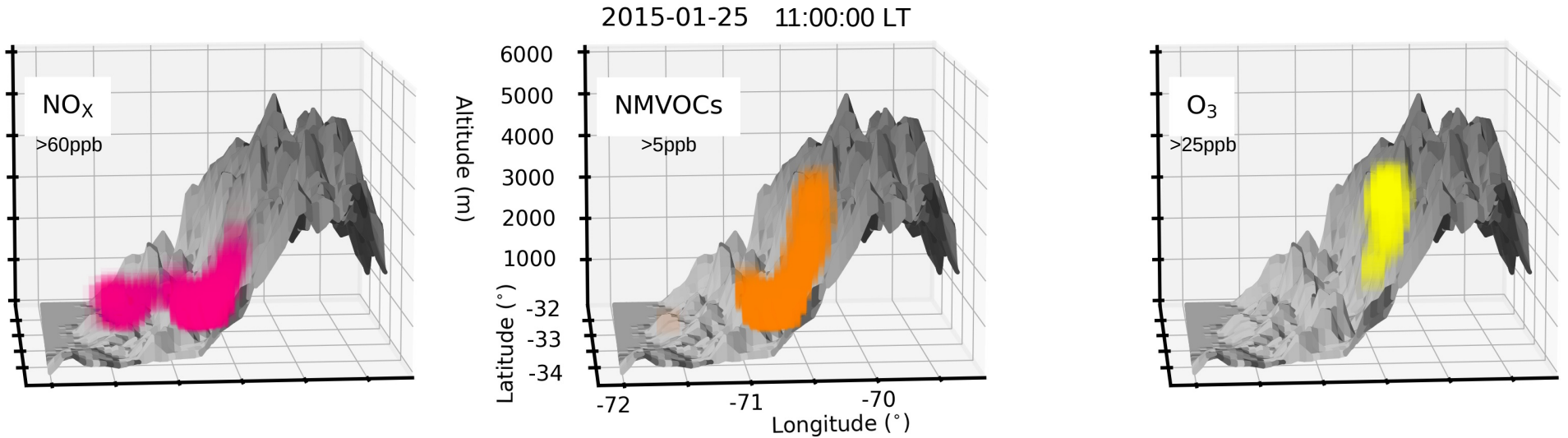


Figure Summertime venting of Ozone and its precursors

Ozone chemistry and mountain dynamics



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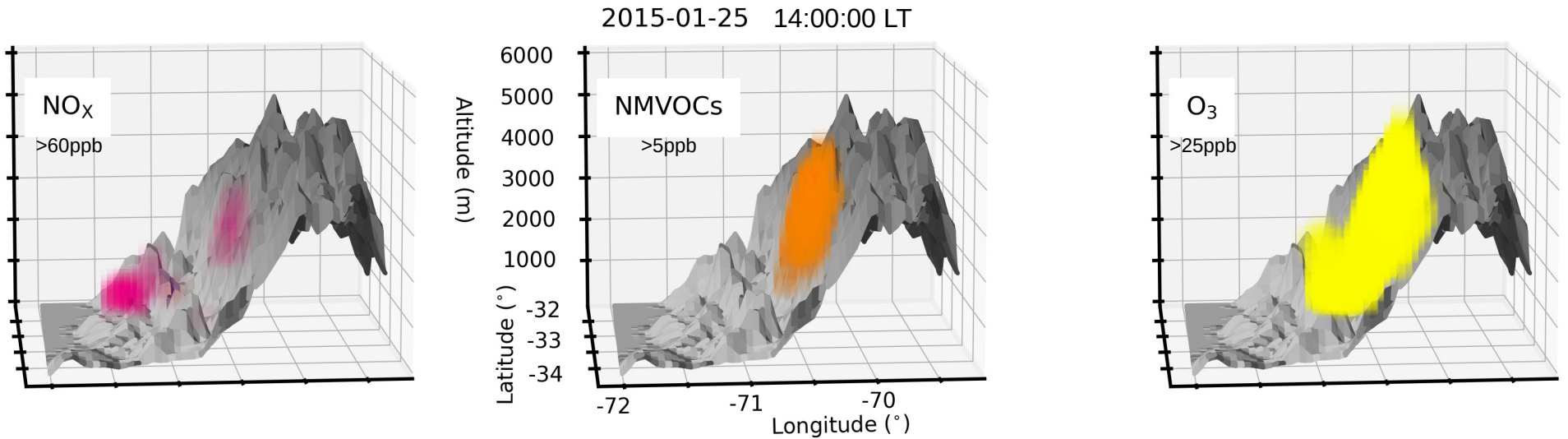

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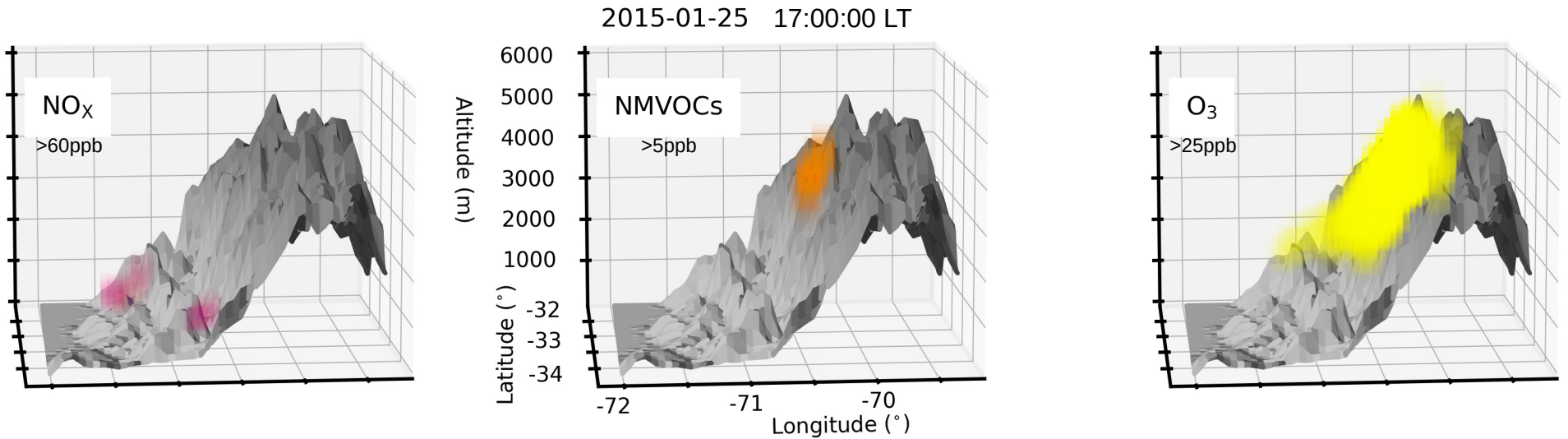

Figure Summertime venting of Ozone and its precursors

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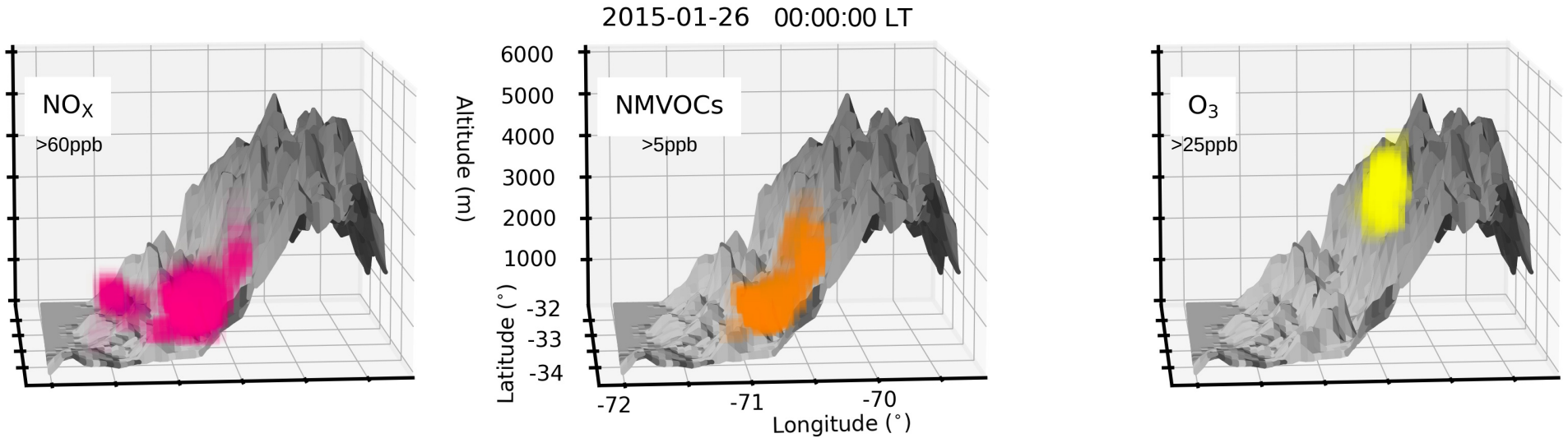
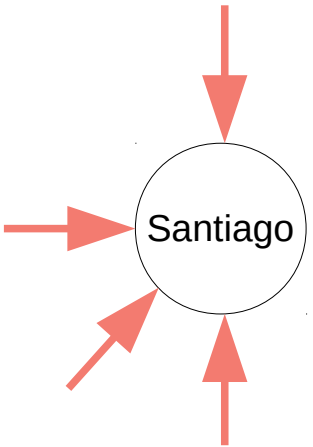


Figure Summertime venting of Ozone and its precursors

Conclusions

15%

of winter $PM_{2.5}$ in Santiago from transport



Conclusions

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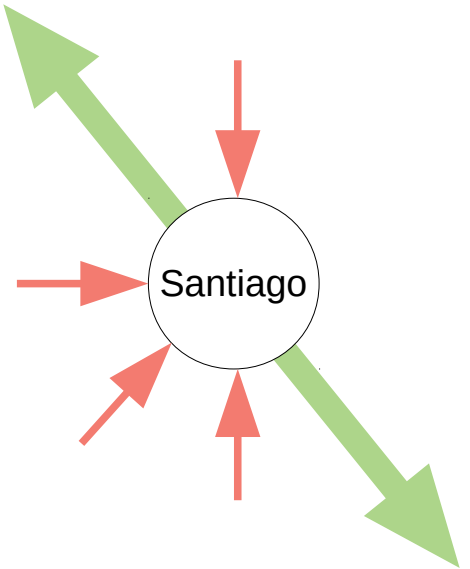
1000km

traveled by $PM_{2.5}$ from Santiago in winter



Conclusions

15%

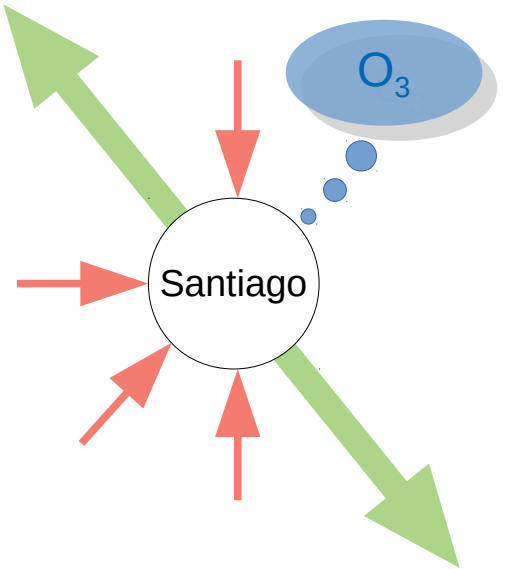
of winter $PM_{2.5}$ in Santiago from transport

1000km

traveled by $PM_{2.5}$ from Santiago in winter

20ppb

excess ozone persistent “bubble” in altitude above Santiago



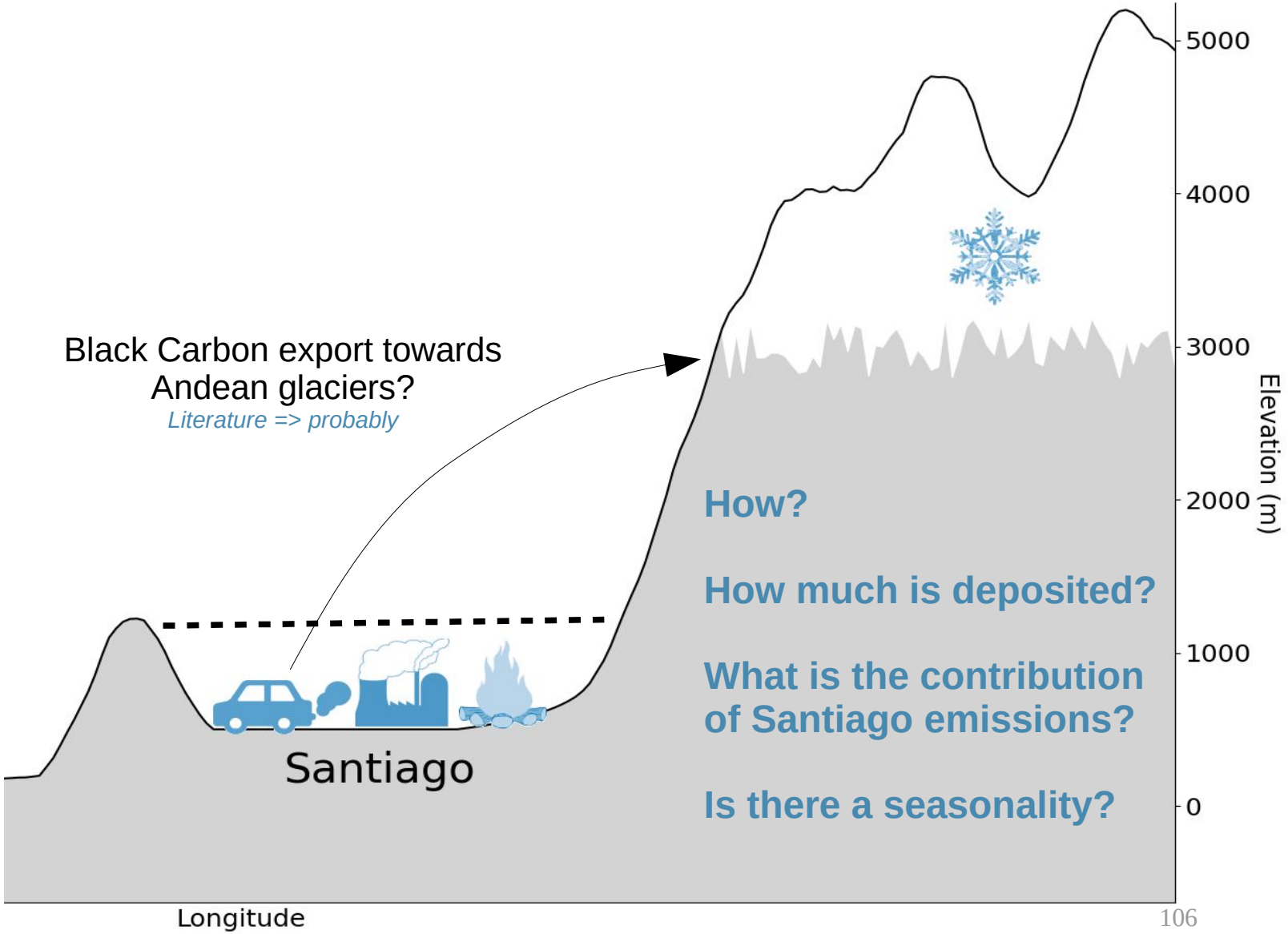
3 – Anthropogenic black carbon and Andean glaciers

(Lapere et al., 2021)
(Huneeus, Lapere et al., 2021)

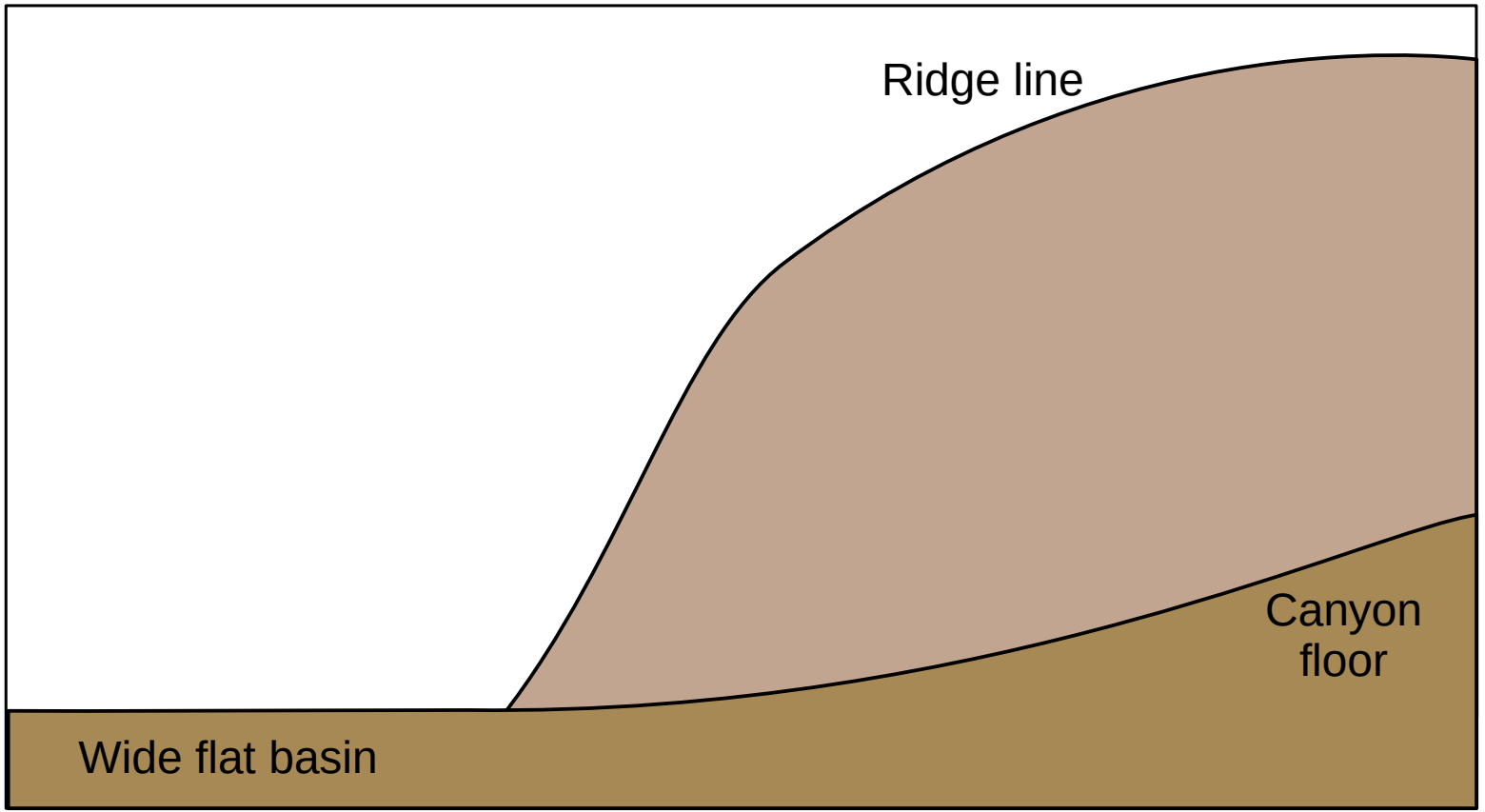


The *Olivares Alfa* glacier in Chile lost 66% of its ice mass in 60 years *(Malmros et al., 2016)*

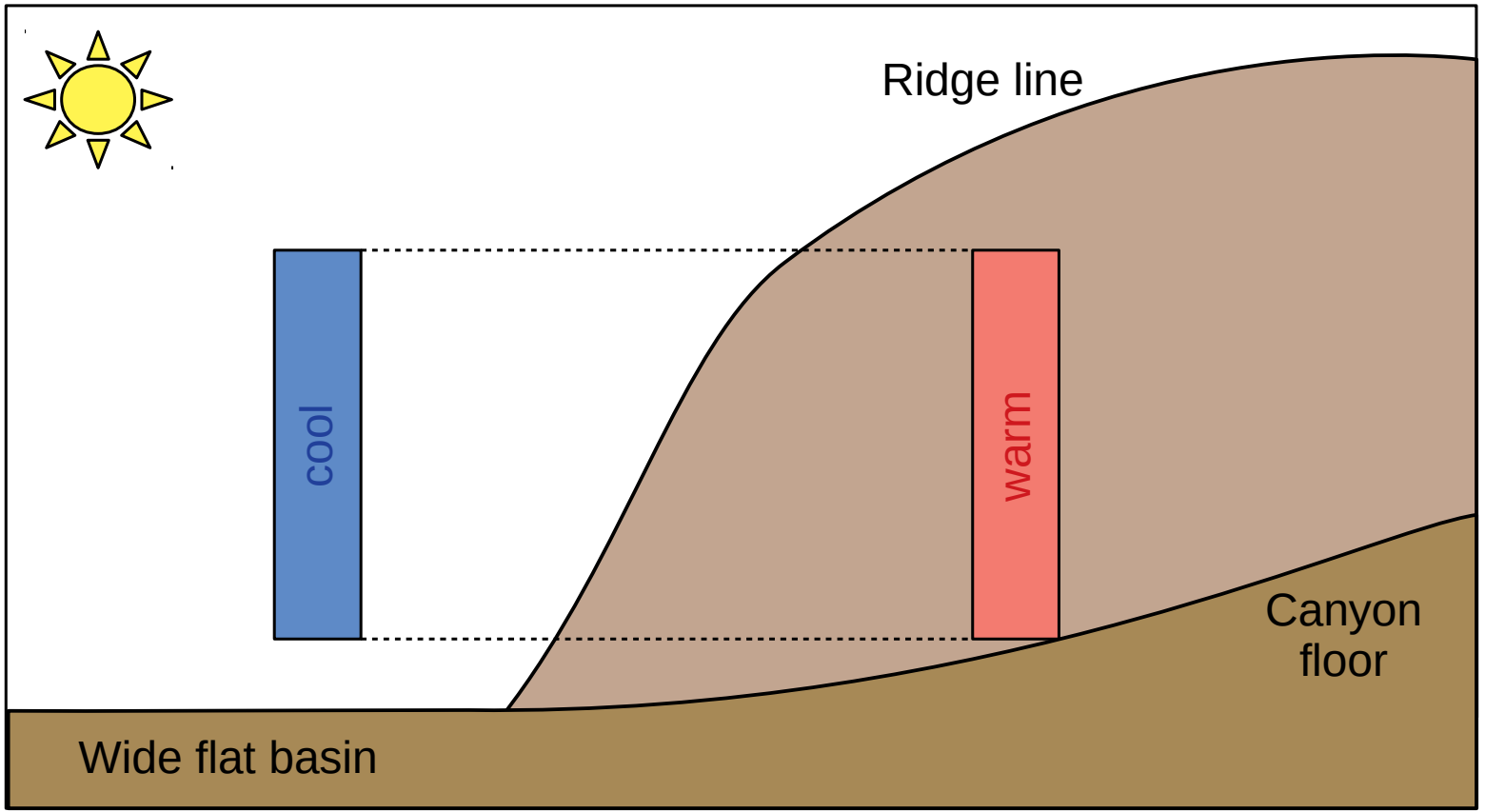



Black Carbon export towards
Andean glaciers?
Literature => probably
Santiago
How?
How much is deposited?
What is the contribution
of Santiago emissions?
Is there a seasonality?
5000
4000
3000
2000
1000
0
Elevation (m)
Longitude

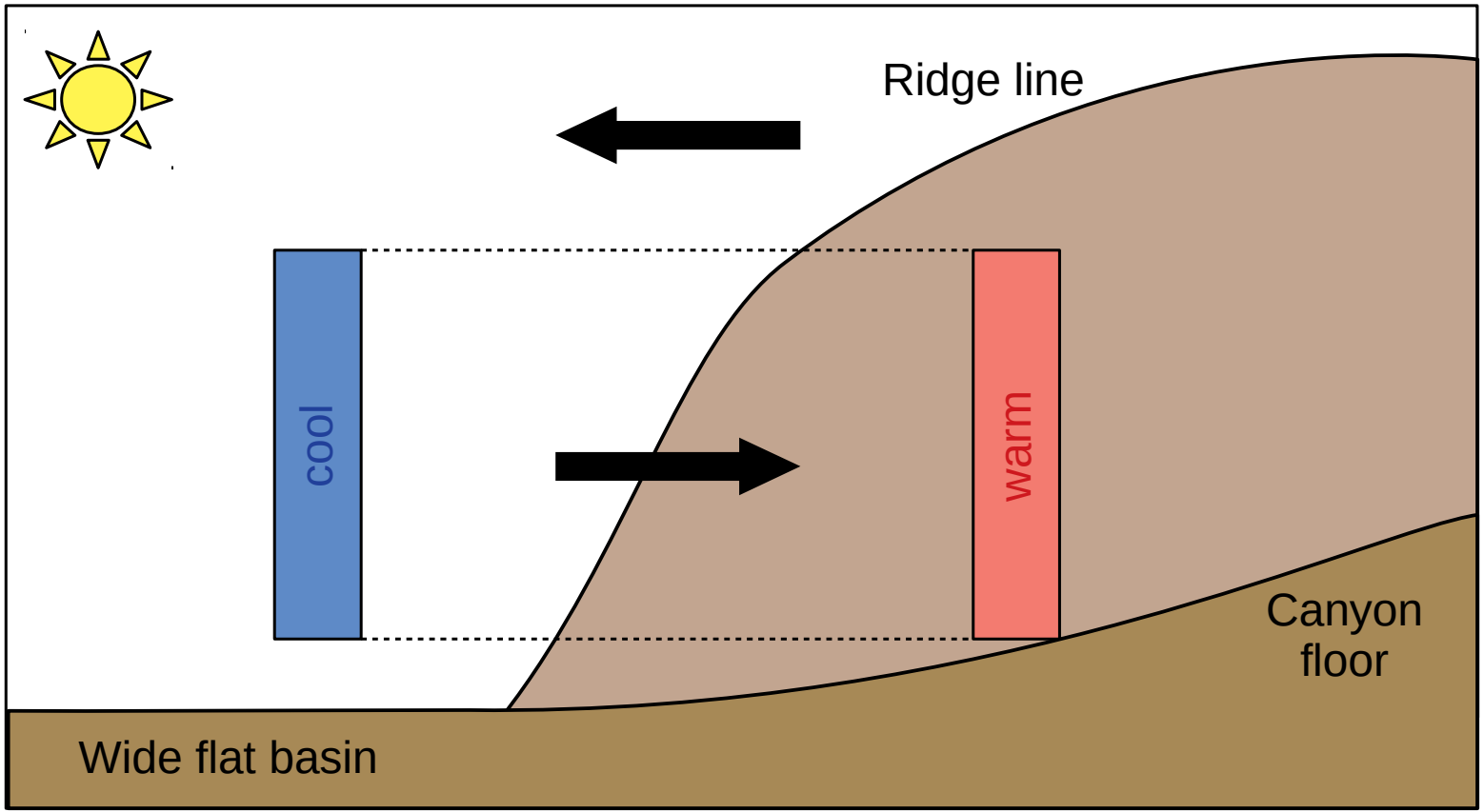
Mountain-valley circulation



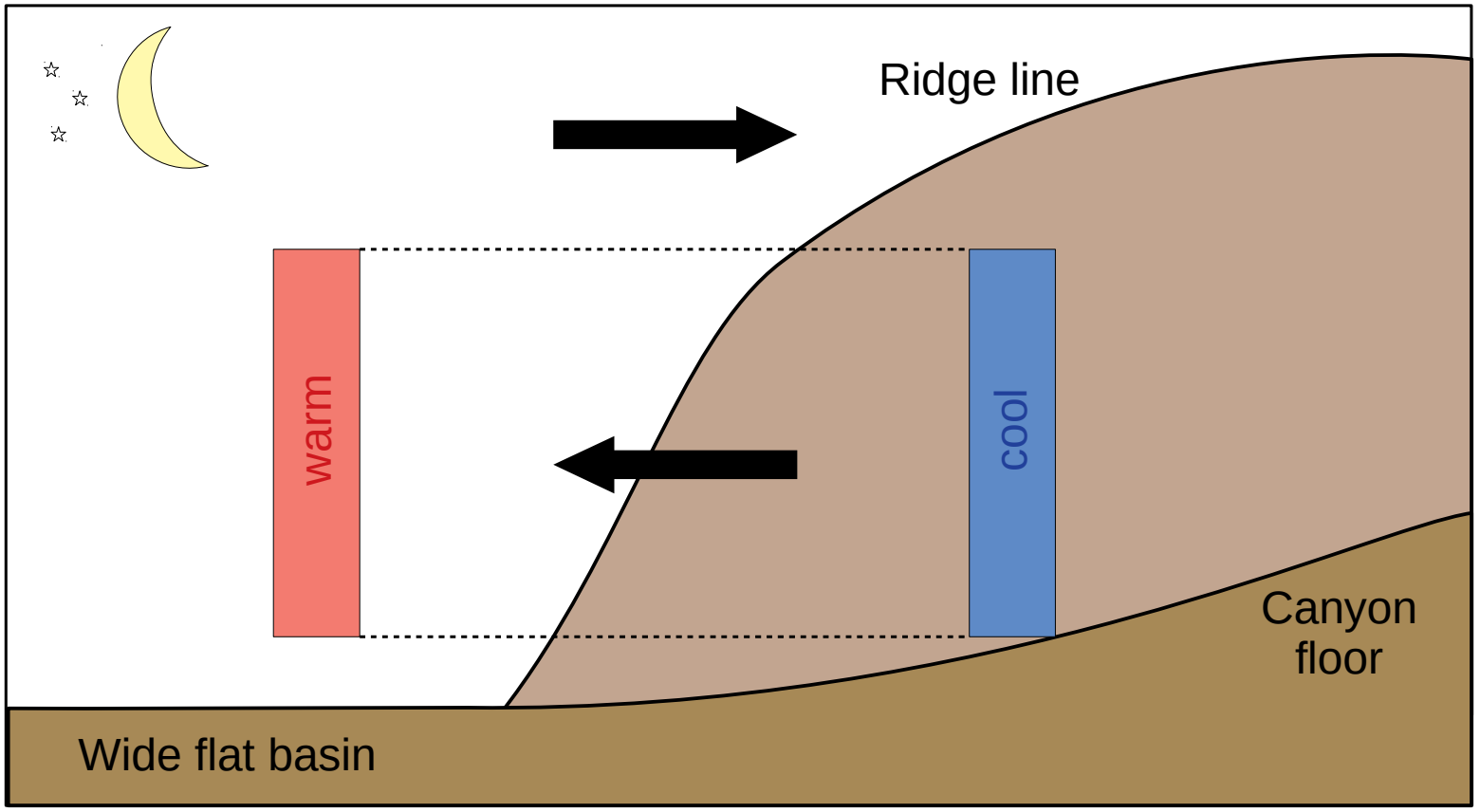
Mountain-valley circulation



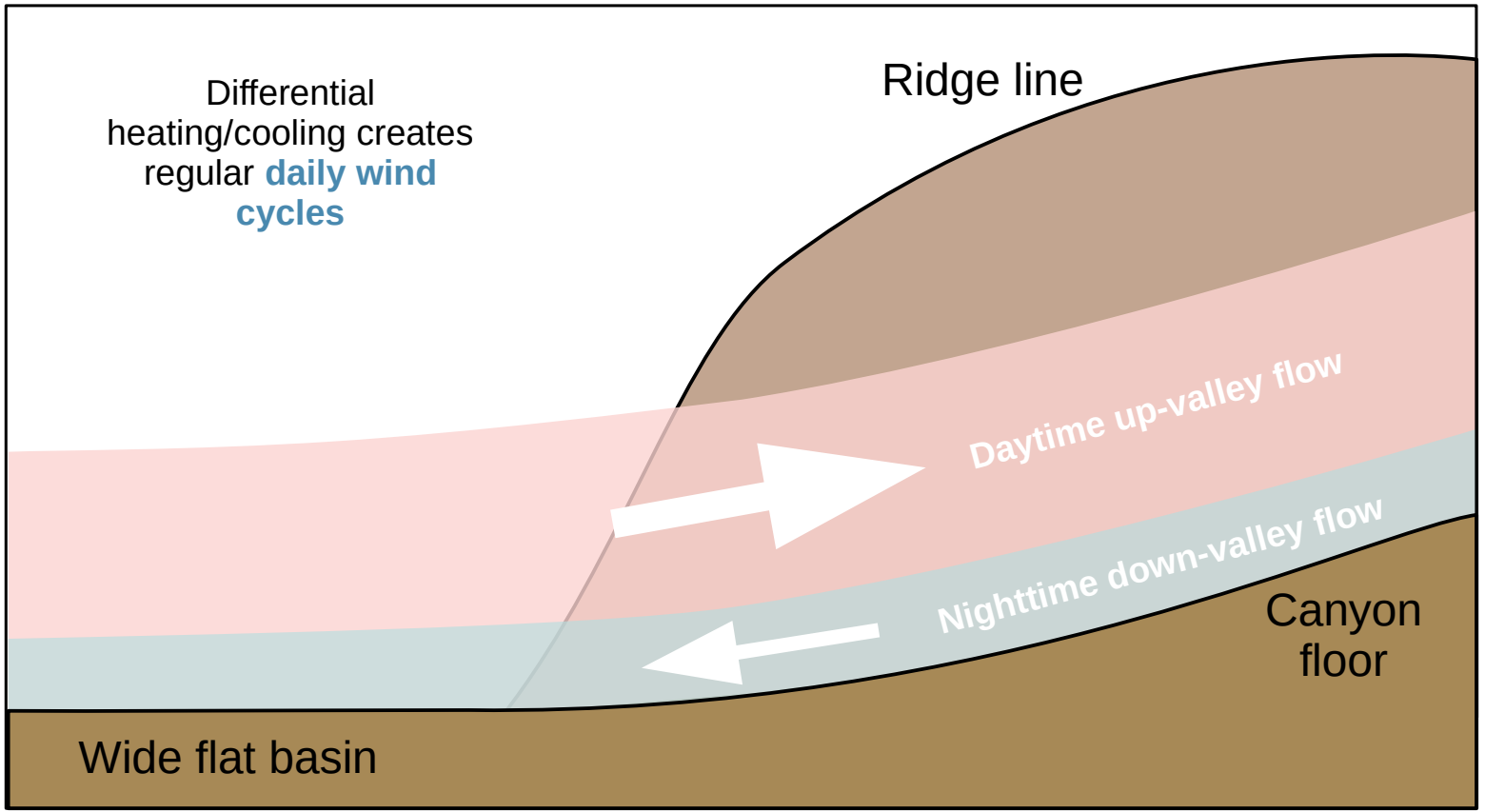
Mountain-valley circulation



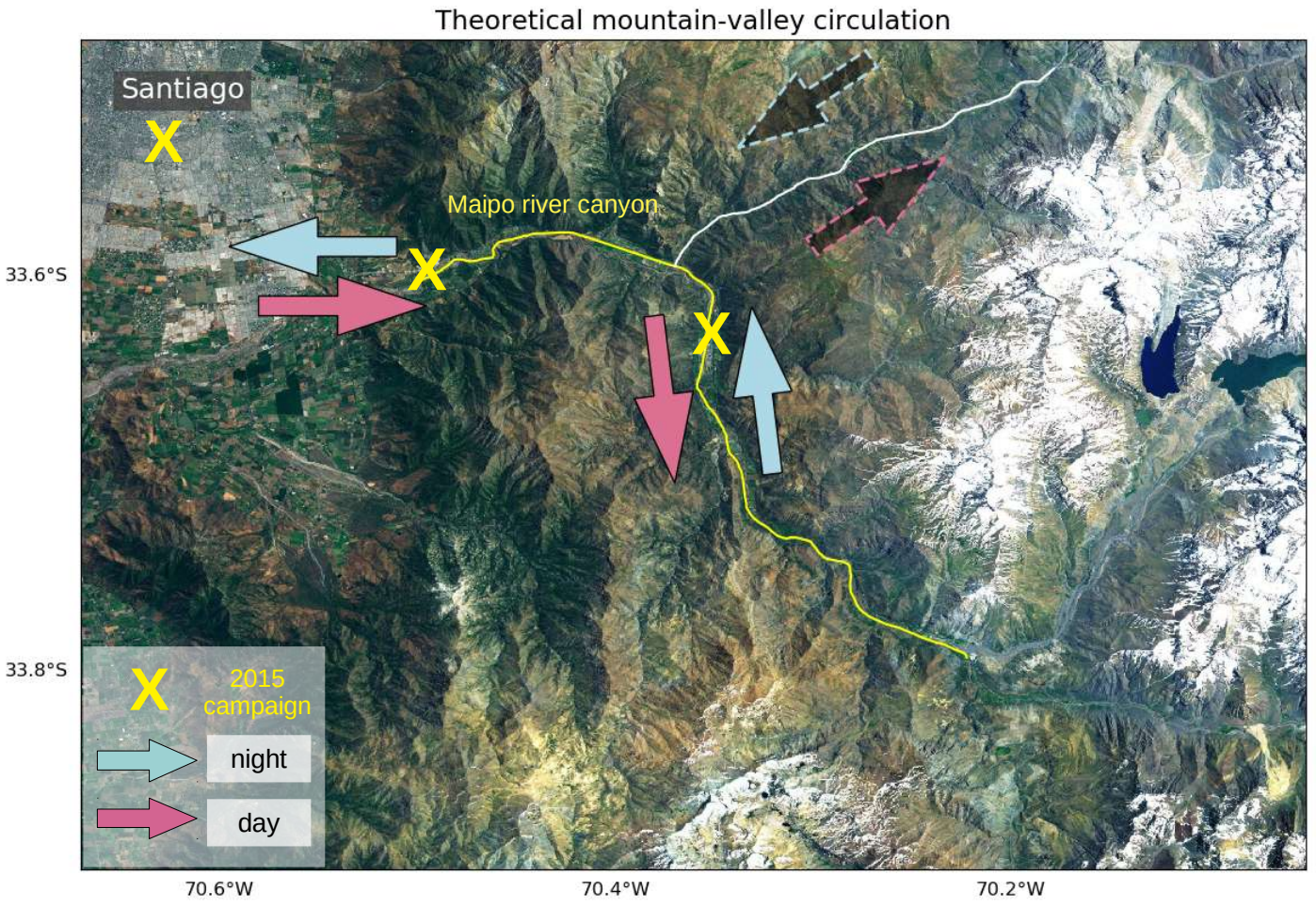
Mountain-valley circulation



Mountain-valley circulation



Mountain-valley circulation



Consequence:

Intrusions of urban air pollutants **into canyons** during the day

Mountain-valley circulation

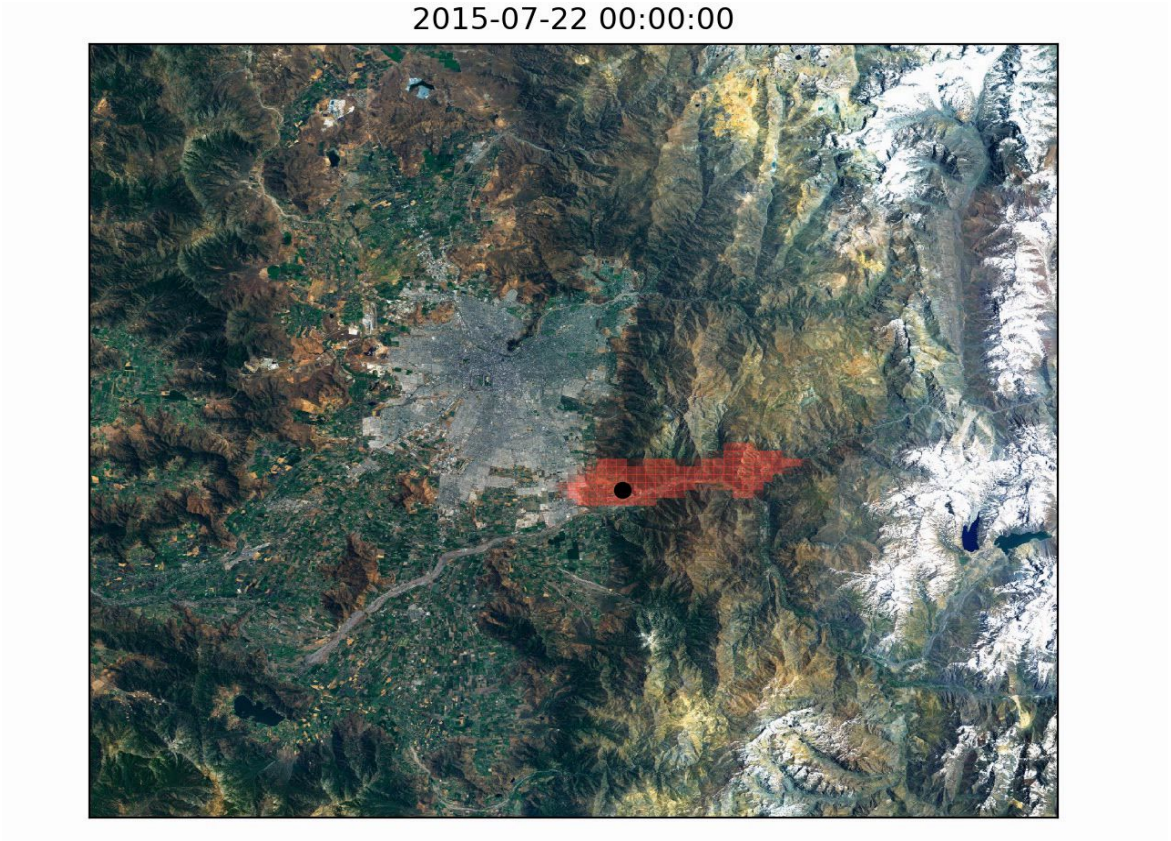


Figure Concentration of a passive tracer as simulated with CHIMERE during a winter period

Wintertime pathway

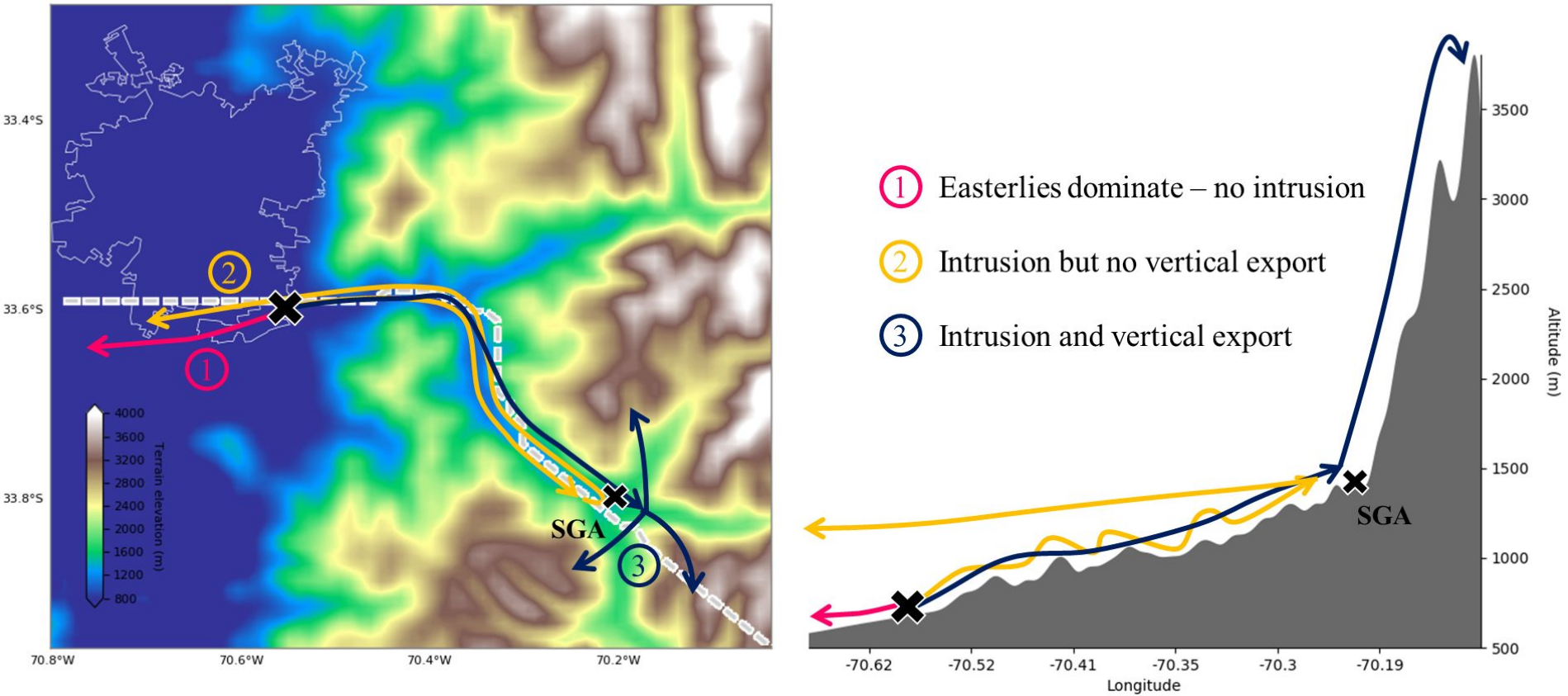


Figure Summary of possible export situations along the Maipo river canyon.

Wintertime pathway

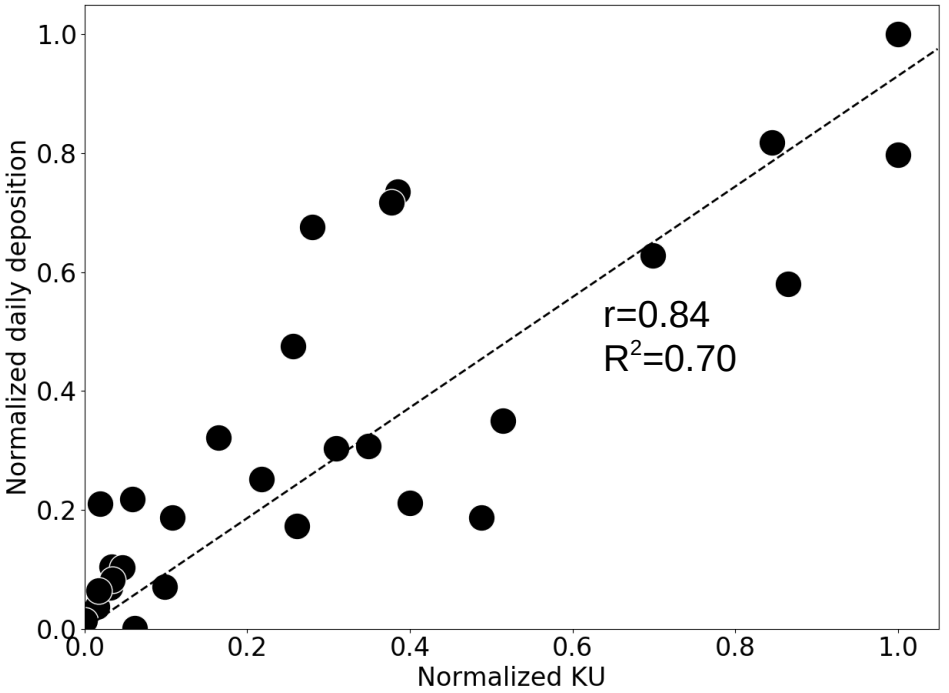


Figure Correlation between tracers deposition and a meteorological index based on zonal wind in Santiago and vertical diffusion deep into the Maipo river canyon.

- KU = index combining:
  (1) wind in Santiago
  (2) K_z in the Maipo canyon

- Explains **70% of the variability** in particles deposition on glaciers

Wintertime / Summertime

Emission fluxes and deposition rates

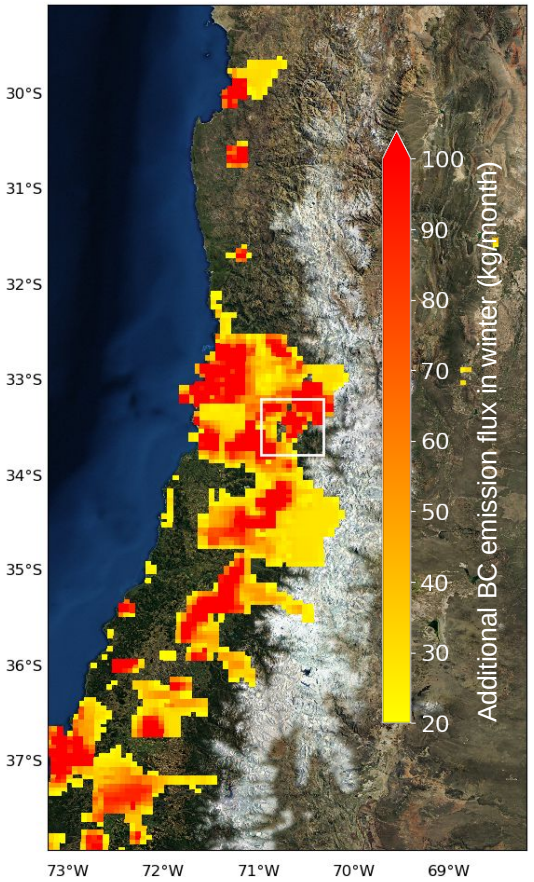
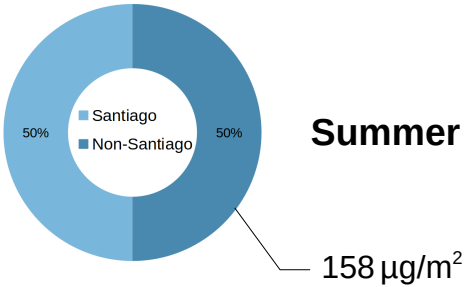
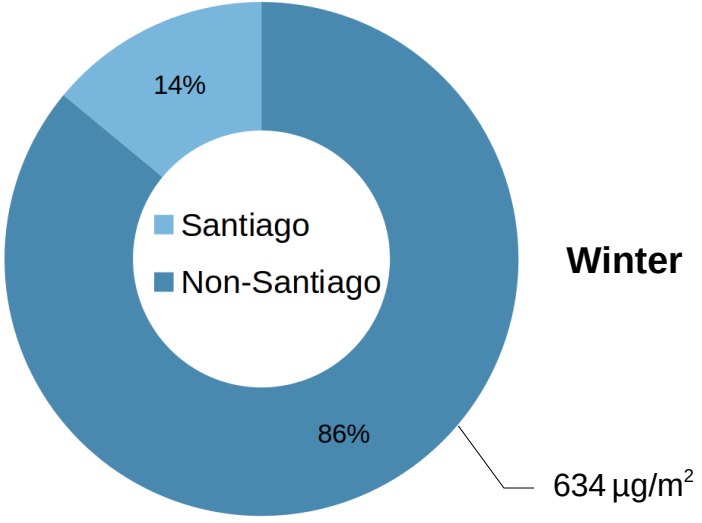


Figure BC emission rate difference winter-summer (HTAP)

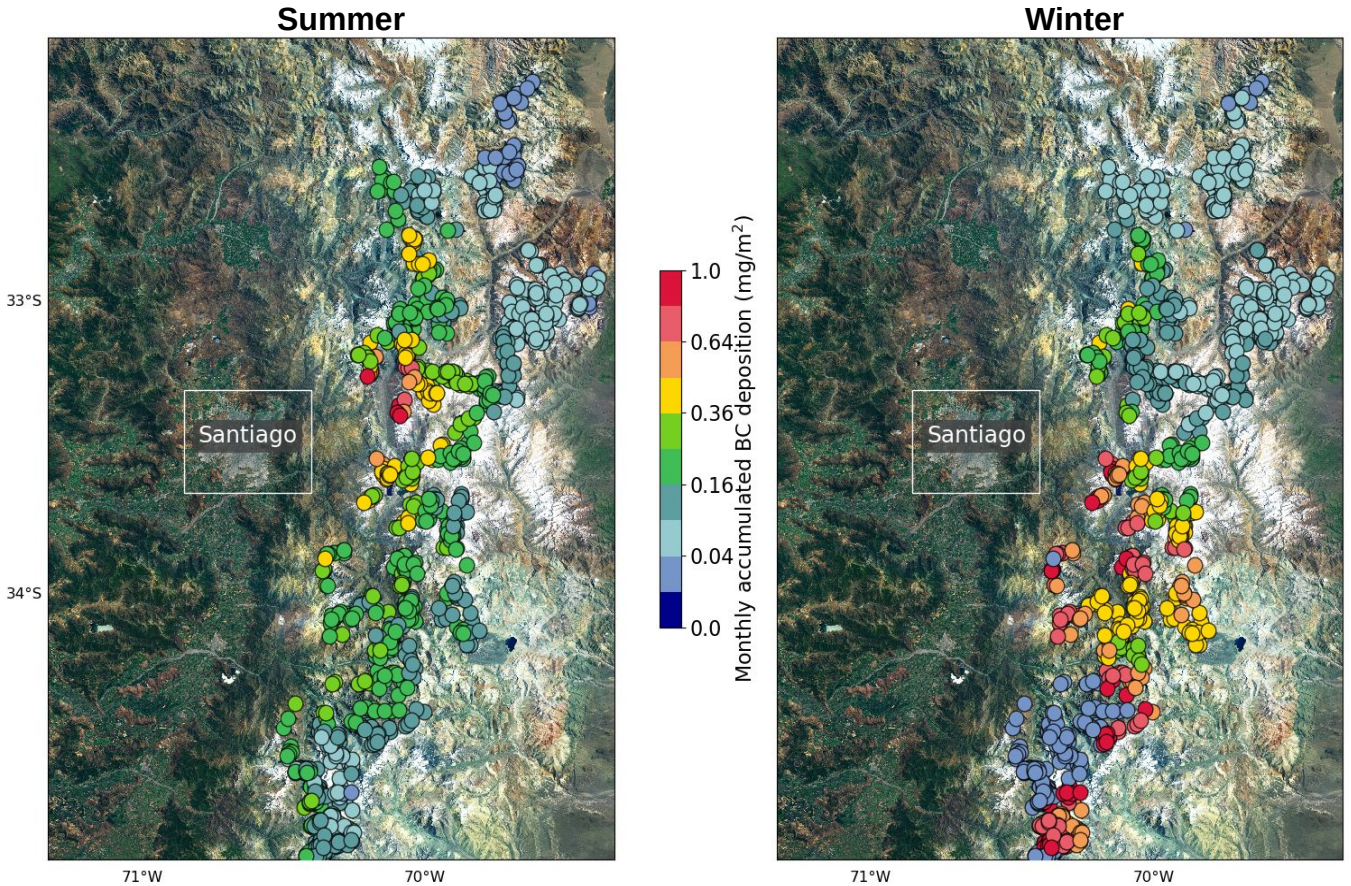


Average deposition fluxes (dry+wet) on glaciers of the central Andes simulated by CHIMERE



- Simulated deposition rates **larger than literature** *(Rowe et al., 2019)* and comparable to Tibetan Plateau *(Ji, 2016; Wang et al., 2017; Yan et al., 2019)*
- Possible **underestimation of radiative forcing** by a factor 2 to 4
- Santiago emissions **do not dominate** the winter signal

Deposition on individual glaciers



- Deposition reaches locally more than **1mg/m²/month**
- Impacted areas **change seasonally**
- Likely tied to different underlying **export mechanisms**

Figure Simulated BC accumulated deposition flux on glaciers (circles) for summer and winter 2015.

Synoptic circulation

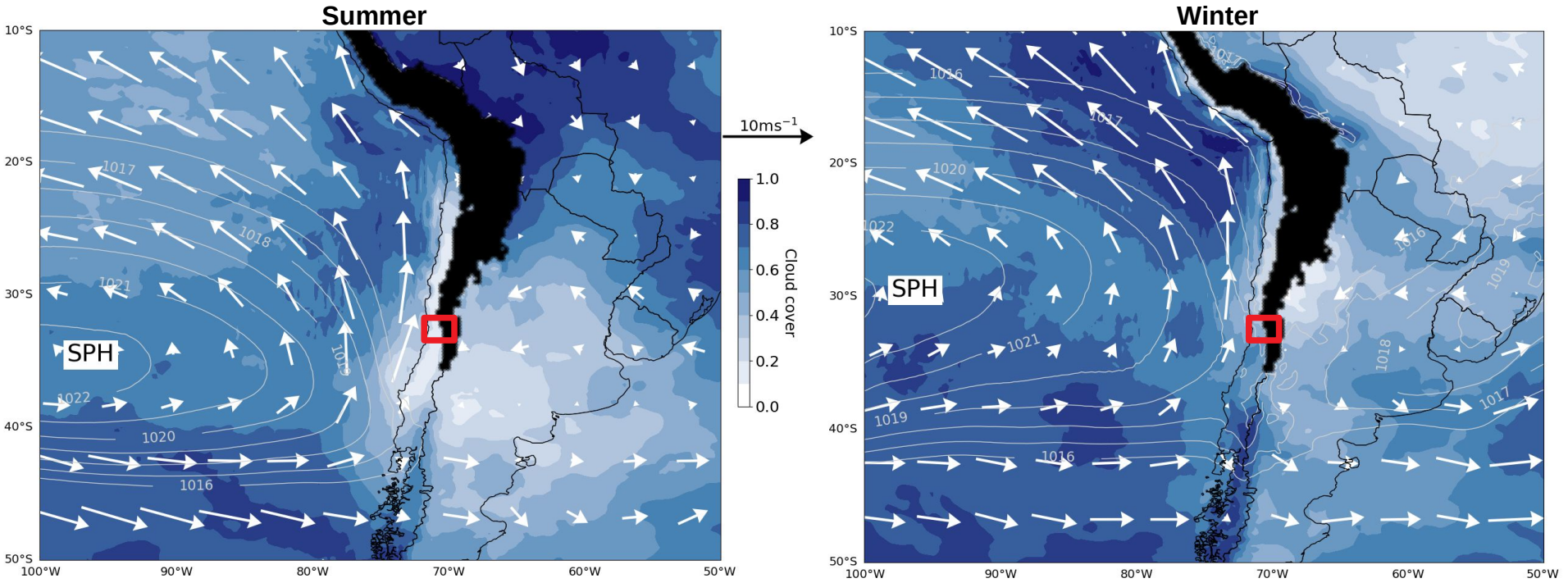


Figure Mean synoptic-scale circulation characteristics for summer and winter months. Data from ERA5 for 2010-2020.

Summer	Consistent synoptic forcing No cloud cover
Winter	Weak synoptic winds Average cloud cover ~40%

Wind profile in Santiago

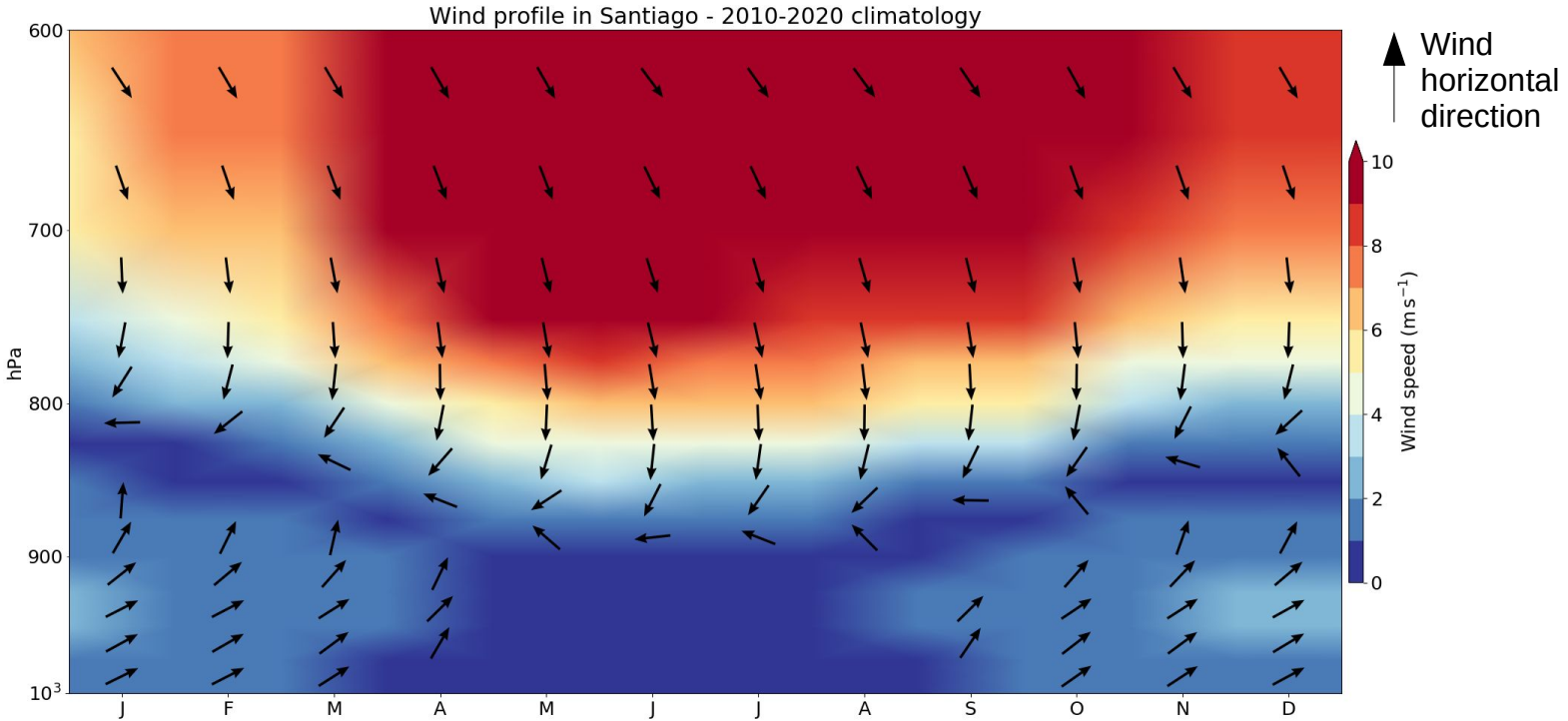
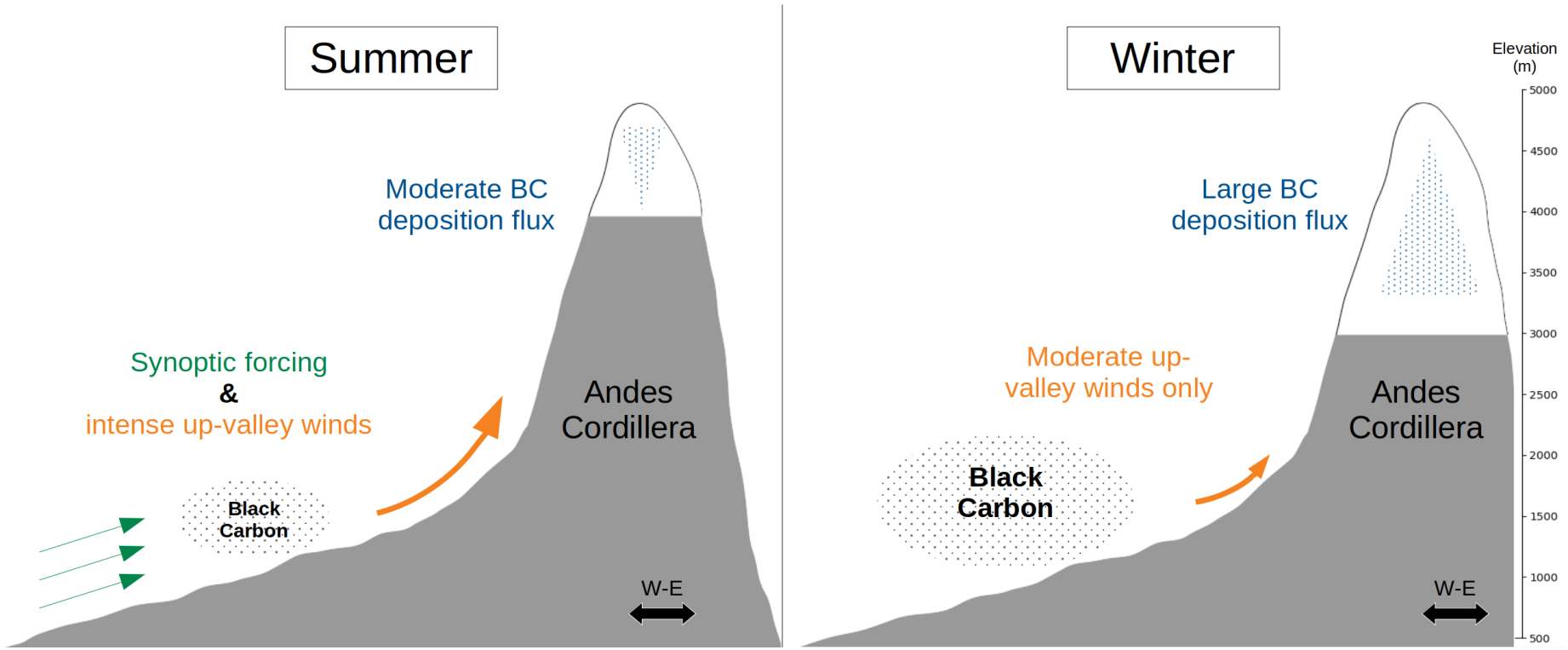


Figure Monthly mean wind vertical profile. Data from ERA5 for 2010-2020.

Summer	Established wind throughout the deep boundary layer
Winter	Shallow boundary layer, average wind not significant

Summary


Summer
Moderate BC
deposition flux
Synoptic forcing
&
intense up-valley winds
Black
Carbon
Andes
Cordillera
W-E
Winter
Large BC
deposition flux
Moderate up-
valley winds only
Black
Carbon
Andes
Cordillera
W-E
Elevation
(m)
5000
4500
4000
3500
3000
2500
2000
1500
1000
500

Conclusions

2-4

possible underestimation of BC deposition at the scale of the central Andes

Conclusions

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14%-40% vs **50%-100%**

contribution of Santiago emissions to BC deposition on glaciers in winter vs summer

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14%-40% vs **50%-100%**

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70%

of deposition variability near Santiago in winter explained by mountain dynamics

Conclusions

2-4

possible underestimation of BC deposition at the scale of the central Andes

14%-40% vs **50%-100%**

contribution of Santiago emissions to BC deposition on glaciers in winter vs summer

70%

of deposition variability near Santiago in winter explained by mountain dynamics

51% emissions but **74%** deposition

in summer compared to winter near Santiago

General Conclusions

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Combining the dense Chilean **observation** network with **chemistry-transport modeling** led to new results

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Causes and **effects** of **short-lived** events were better constrained

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Additional knowledge on the dominant **transport mechanisms** and the associated **magnitudes** was obtained

General conclusions



Combining the dense Chilean **observation** network with **chemistry-transport modeling** led to new results

Causes and **effects** of **short-lived** events were better constrained

Additional knowledge on the dominant **transport mechanisms** and the associated **magnitudes** was obtained

Uncharted territory was **explored** with the **ozone bubble** and map of **BC deposition** on glaciers of the cordillera

Recommendations

Recommendations



Policy design needs to consider **sporadic acute events** induced by specific sources

Recommendations



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Air pollution mitigation initiatives should focus **not only on local emissions**

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Black carbon deposition on glaciers of the Andes should be considered a **greater threat**

Recommendations

Policy design needs to consider **sporadic acute events** induced by specific sources

Air pollution mitigation initiatives should focus **not only on local emissions**

Black carbon deposition on glaciers of the Andes should be considered a **greater threat**

Santiago emissions but also across **central Chile** should be monitored

Perspectives

Perspectives

Black carbon and the Andes:

- Include **dust** and **BC/dust interaction** in the scope
- Evaluate radiative feedback and melting with a **snow module** coupled to CHIMERE
- Simulate at **higher resolution** and on **longer time** scales

Work on methodologies to **better estimate the aerosol indirect feedback** during short-lived events

Appendix

Transverse themes

General limitations of this work

Fires

Cloud formation due to aerosol indirect effect is naturally slightly underestimated in CHIMERE: (i) grid-resolved clouds only (ii) no convection-generated clouds *(Menut et al., 2021)*

➔ Higher resolution could be beneficial to minimize the first effect

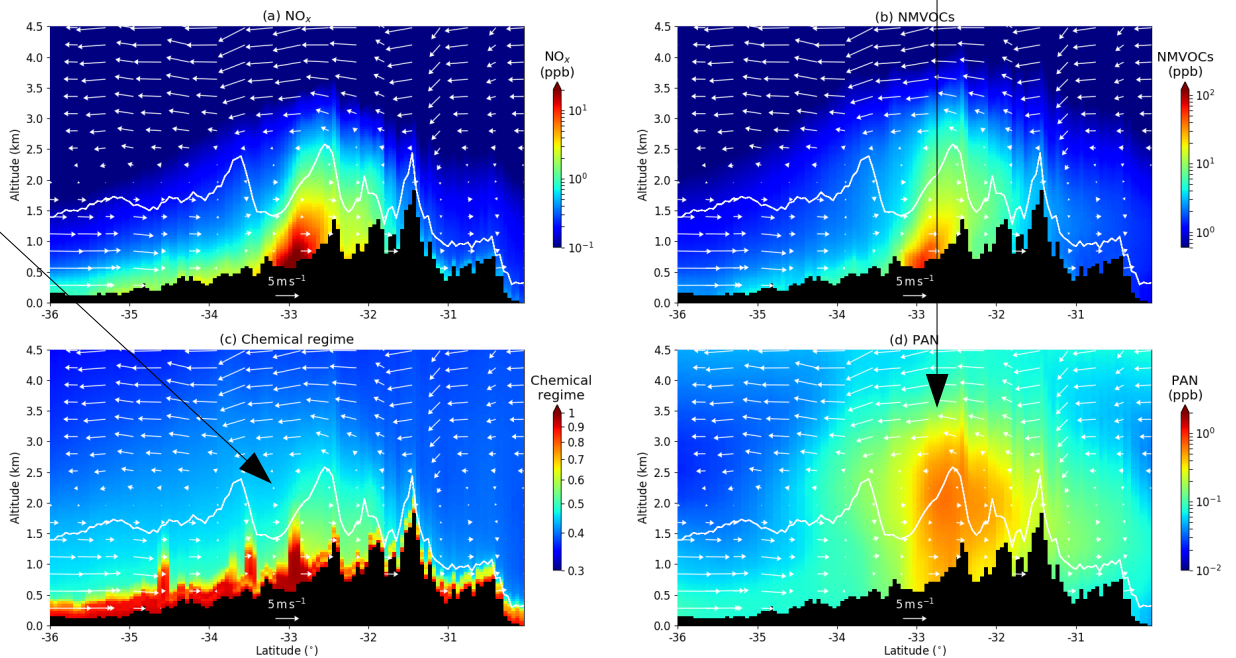
The studied event is likely included in the meteorological fields

➔ Using weather analogs *(Lorenz, 1969)* could be a suitable workaround

Meteorological uncertainty on Ozone

- Dry deposition on vegetation
- Natural sources of precursors
- **Temperature** bias => bias in PAN thermal decomposition => ~ppb O3 bias
- In a VOC-rich regime, water vapor - hence **relative humidity** - limits O3 levels (O3 removal by HOx)

(Lu et al., 2019)



General limitations of this work

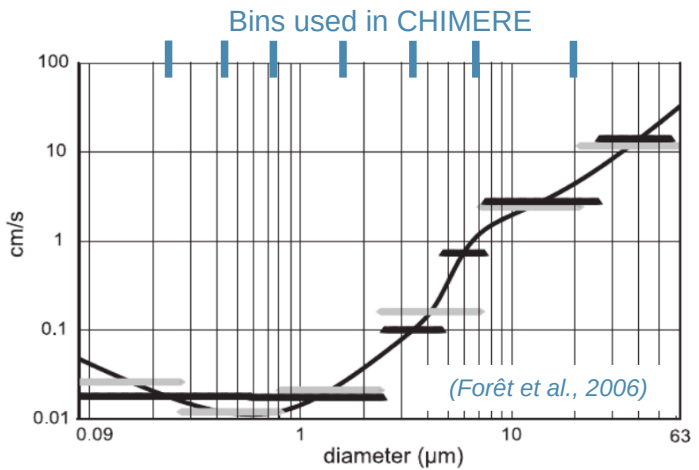
Black carbon export & deposition

Modeled deposition fluxes highly depend on the choice of the size distribution of particles *(Forêt et al., 2006)*

➔ Running sensitivity tests with increasing size bins would constrain this uncertainty

Wind is dominated by small-scale phenomena in winter and thus less well captured by WRF

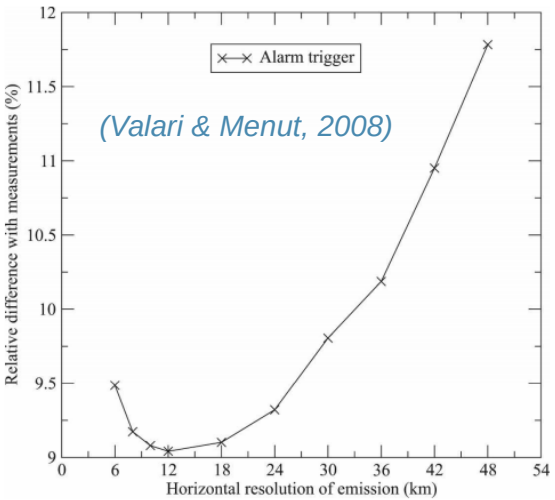
➔ WRF-DA with local wind data in the Andes could improve the performance *(Cazes Boezio & Ortelli, 2019)*



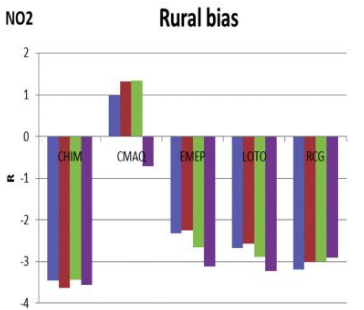
Impact of simulation resolution

- Increasing horizontal resolution improves simulations **up to a point** (~10km and mostly for urban regions) when input data resolution (emissions, meteorological fields…) becomes the limiting factor *(Schaap at al., 2015; Valari & Menut, 2008)*

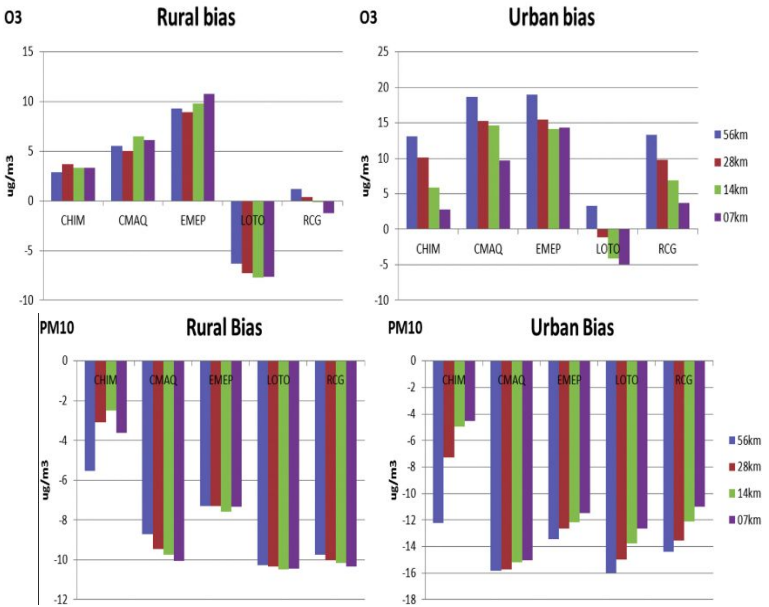
- Subgrid parameterization then becomes a better solution over resolution increase to gain accuracy *(Valari & Menut, 2008)*



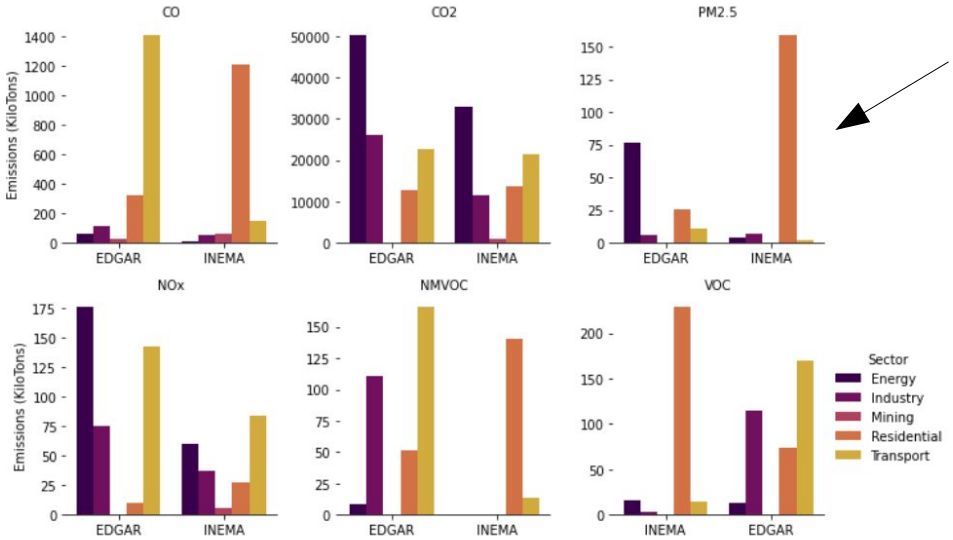
(Valari & Menut, 2008)



(Schaap at al., 2015)

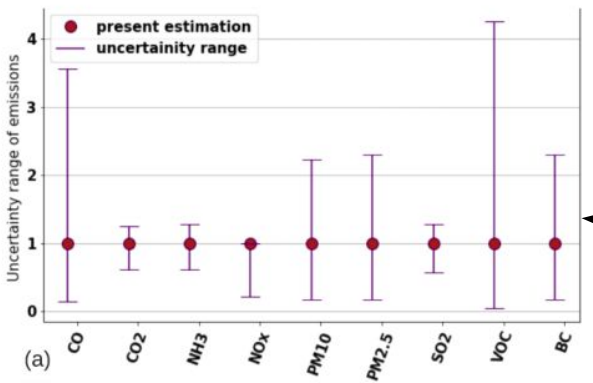


Uncertainty on emissions



Both over- and under-estimation in HTAP

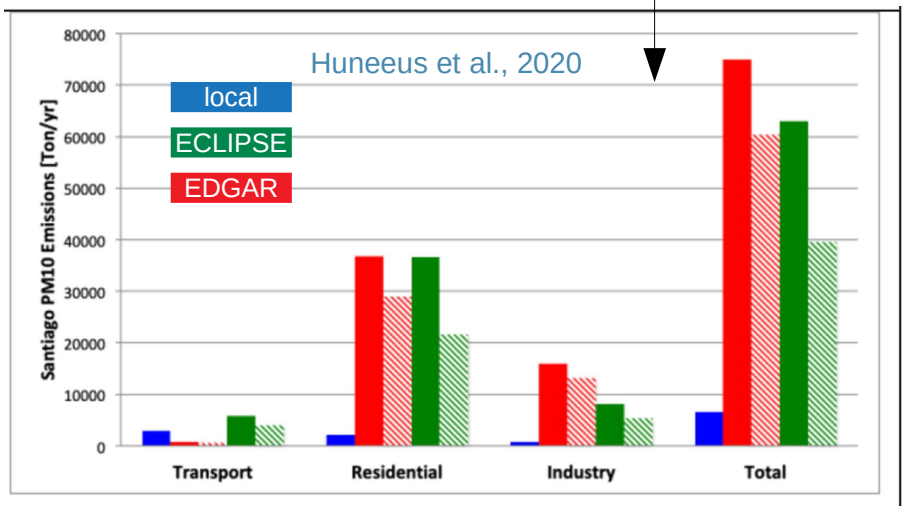
Álamos et al., 2021



Still large uncertainty in most recent inventories

Discrepancies not homogeneous throughout Chile

Huneeus et al., 2020



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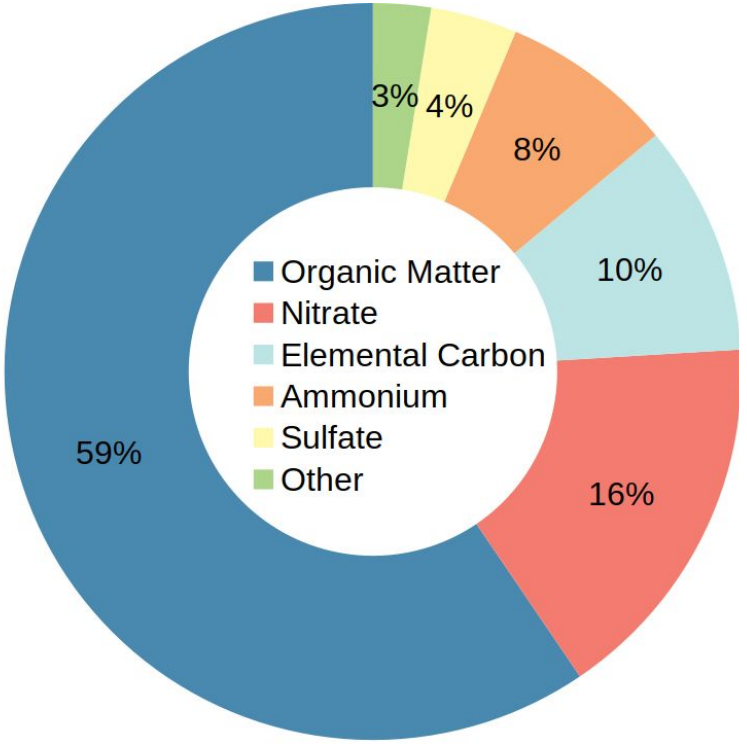
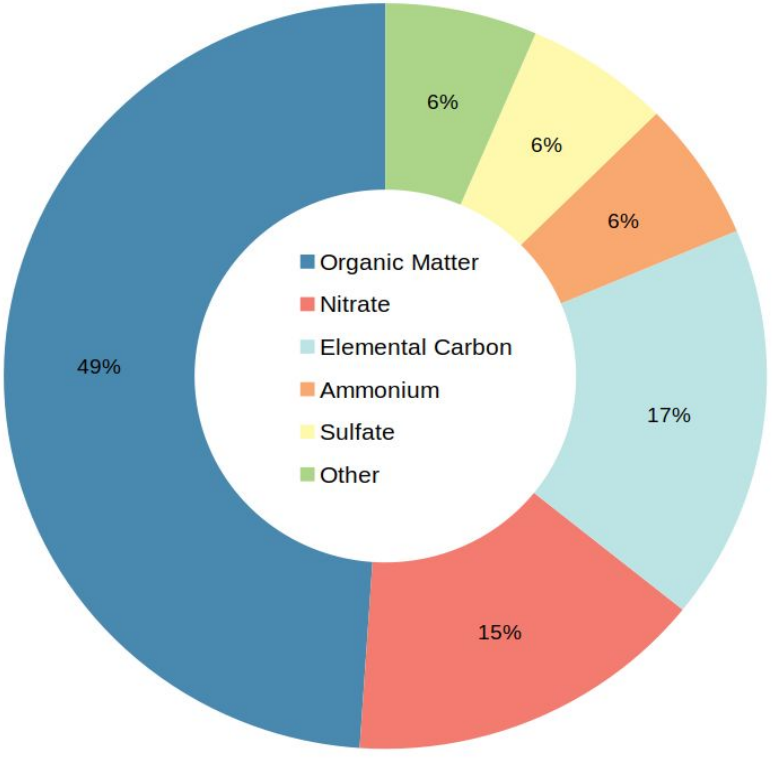


Bulk composition of PM2.5 in Santiago - June 2013

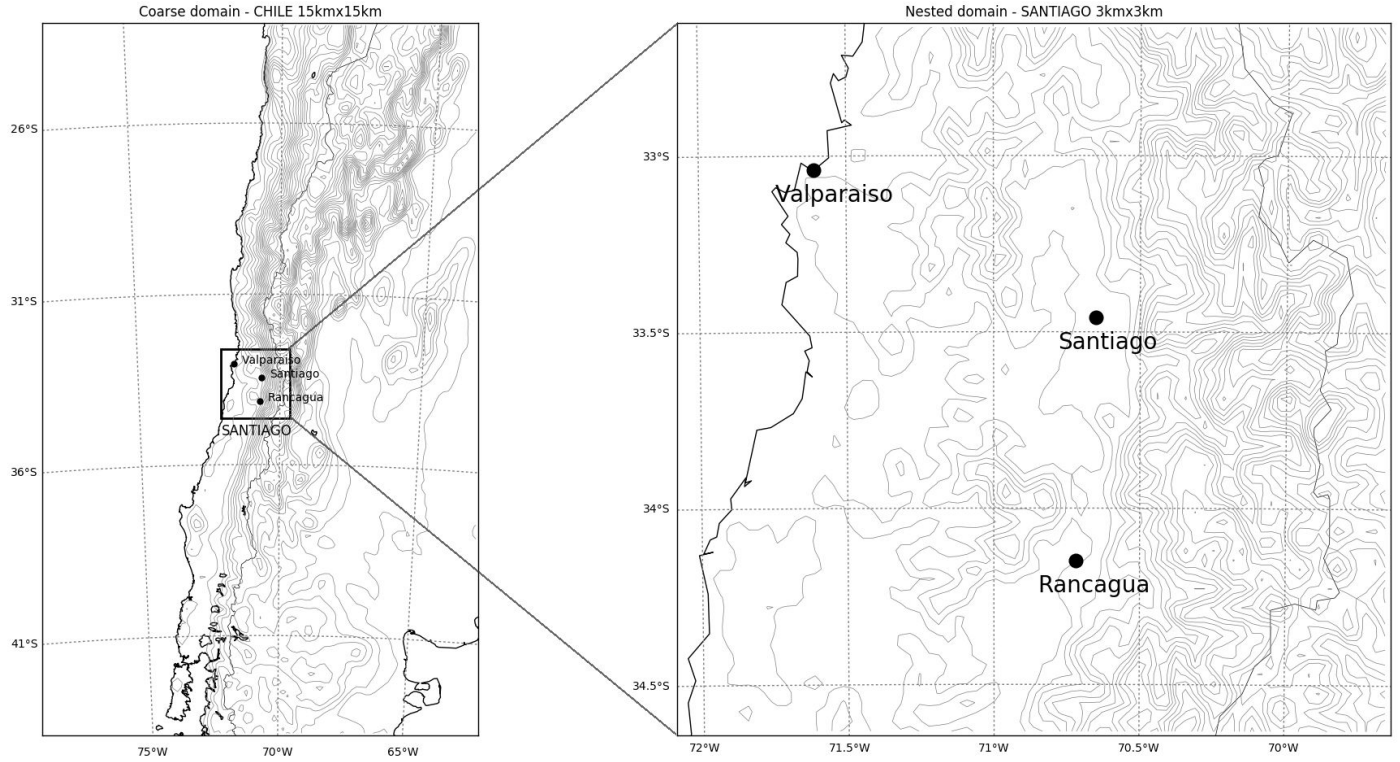
Adapted from *Villalobos et al., 2015*



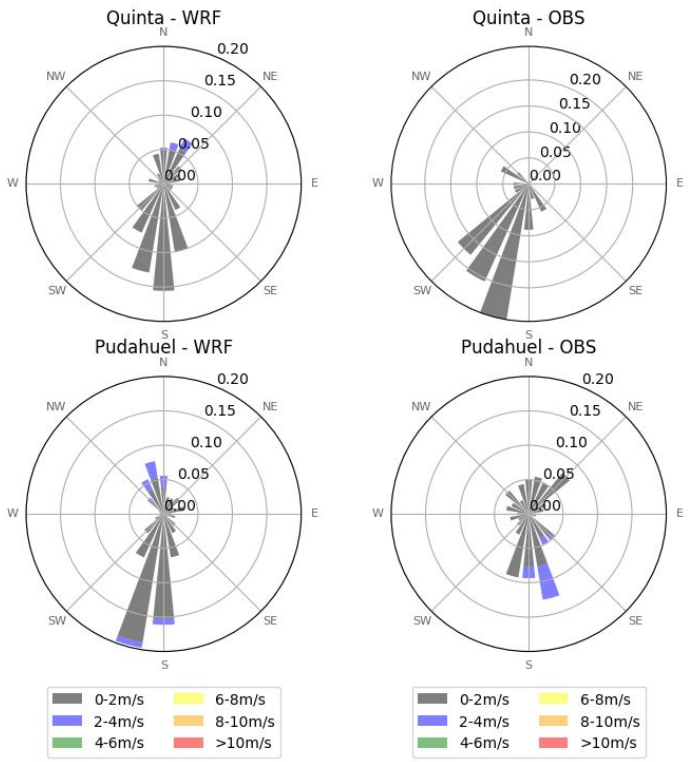
Composition of surface PM2.5 in Santiago simulated by CHIMERE - July 2015

2016 barbecues

2016 barbecues – Simulation design



2016 barbecues – Simulation validation



Station:	El Bosque				Independencia		
	MB	NRMSE	R	:	MB	NRMSE	R
TEMP	1.3	0.34	0.84	:	-1.06	0.2	0.86
RH	-12.7	0.30	0.62	:	-16.6	0.31	0.7
WS	-0.08	0.42	0.56	:	0.23	1.29	0.7

Table 4.2: Simulation scores for low-level meteorology. June 15th to July 15th 2016

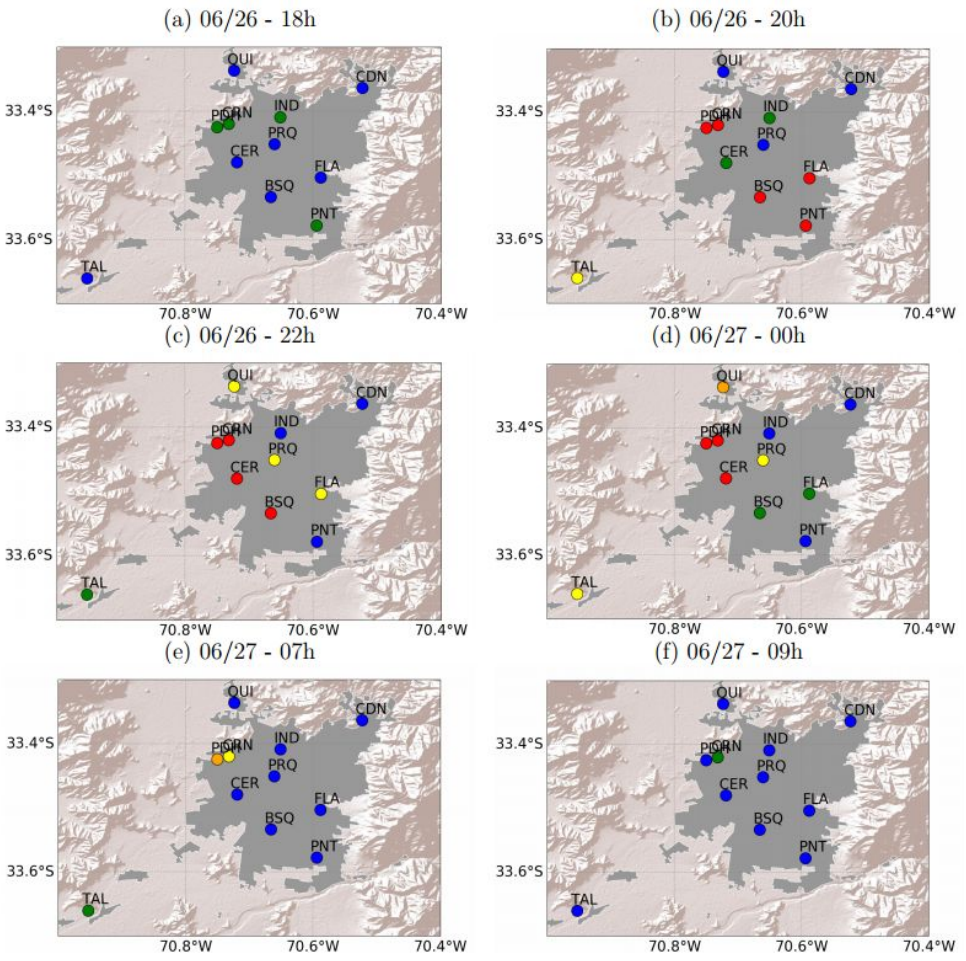
Day:	June 27th 2016				June 28th 2016				June 29th 2016		
	MB	RMSE	R	:	MB	RMSE	R	:	MB	RMSE	R
TEMP	-0.79	2.01	0.99	:	0.51	1.1	0.99	:	0.83	2.4	0.95
RH	7.64	14.3	0.80	:	-1.04	5.88	0.67	:	-7.47	22.2	0.82
WS	-1.92	3.54	0.39	:	1.06	2.71	0.84	:	-3.63	4.38	0.99
WD	-2	4	0.34	:	-4	82	0.71	:	19	85	0.48

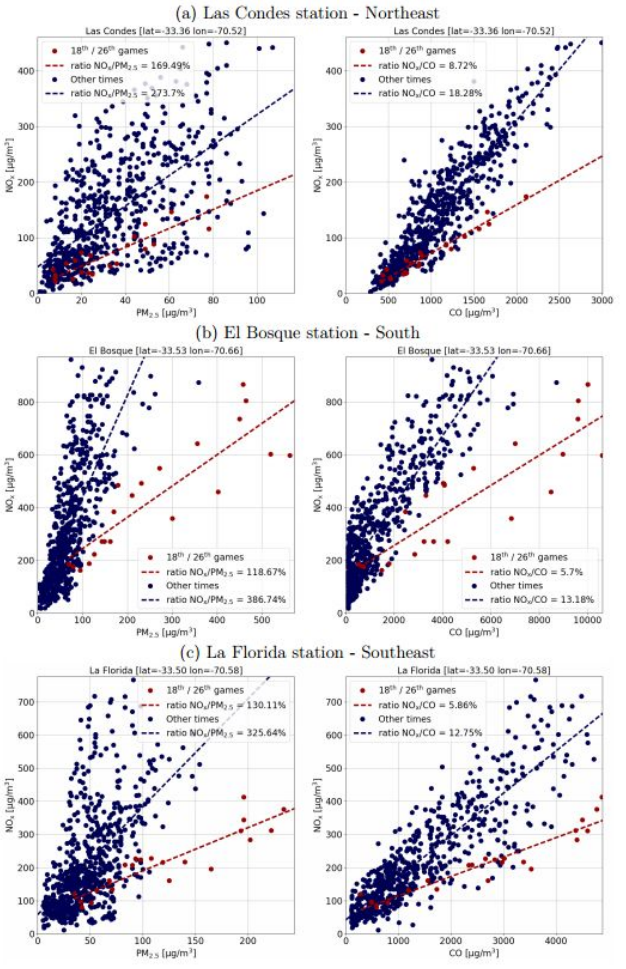
Table 4.3: Simulation scores for meteorological vertical profiles for 3 days at DMC station in Santiago.

Network	$PM_{2.5}$		
	MB	NRMSE	R
La Florida	-0.45	0.55	0.63
Las Condes	3.31	0.62	0.62
Puente Alto	-3.26	0.72	0.45

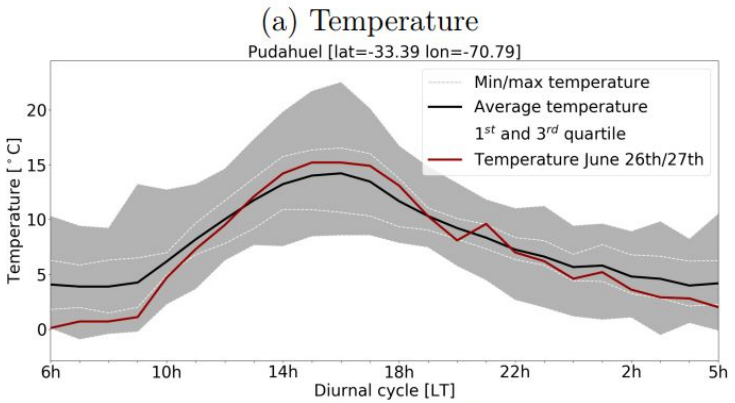
Table 4.4: Simulation scores for low-level $PM_{2.5}$ concentrations. June 28th to July 15th 2016

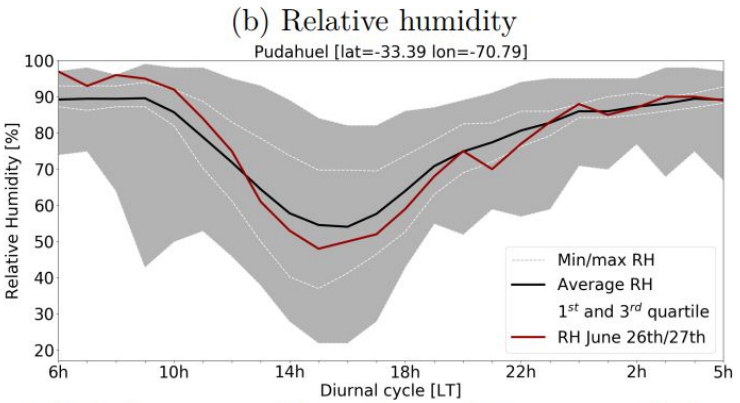
2016 barbecues – Additional material

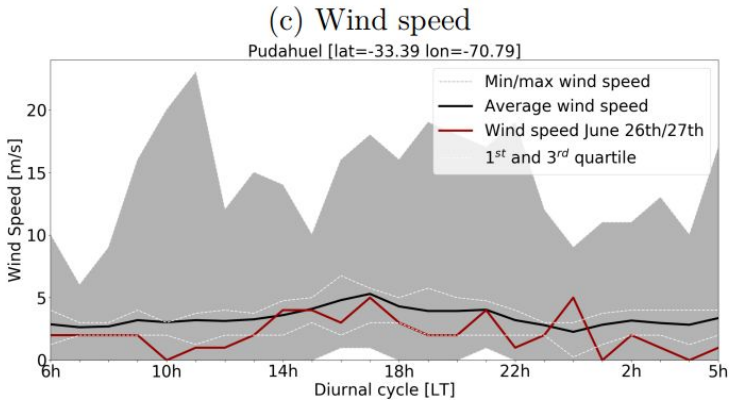


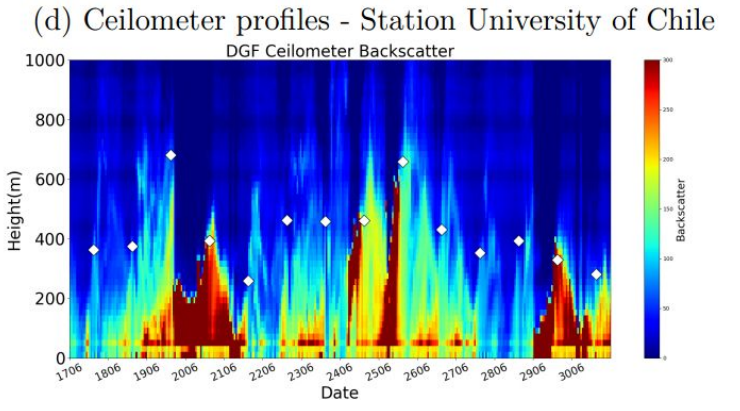


2016 barbecues – Additional material



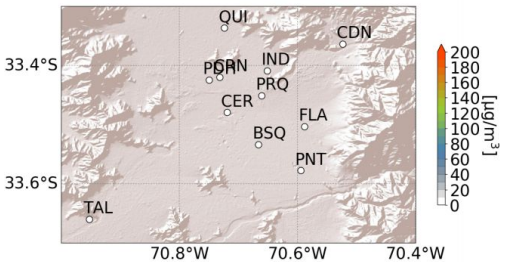




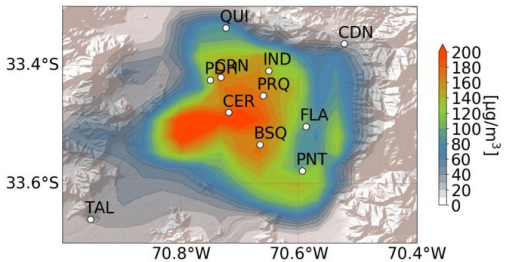


2016 barbecues – Additional material

(a) 18/06 - 20h LT



(b) 18/06 - 22h LT



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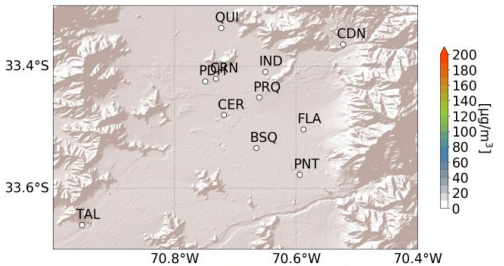

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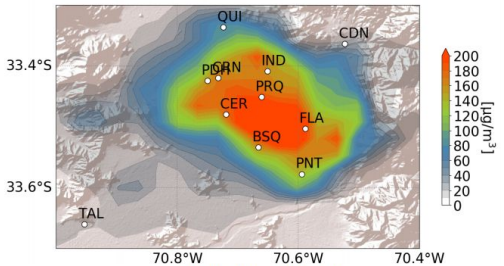
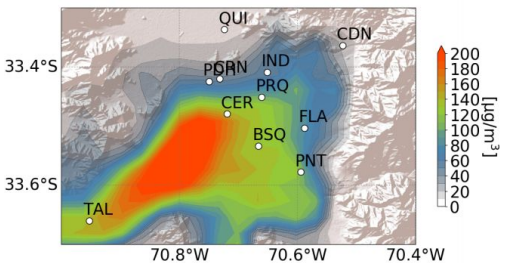


(c) 19/06 - 02h LT



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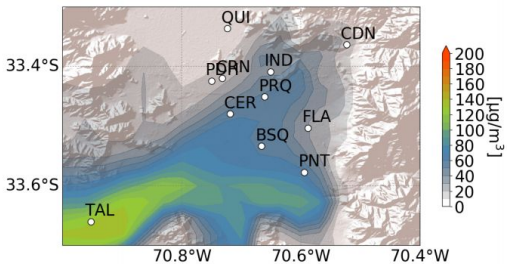

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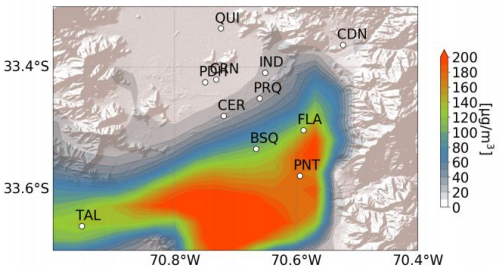

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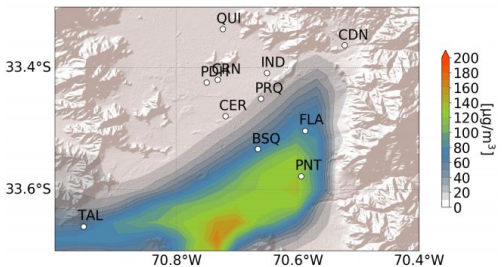
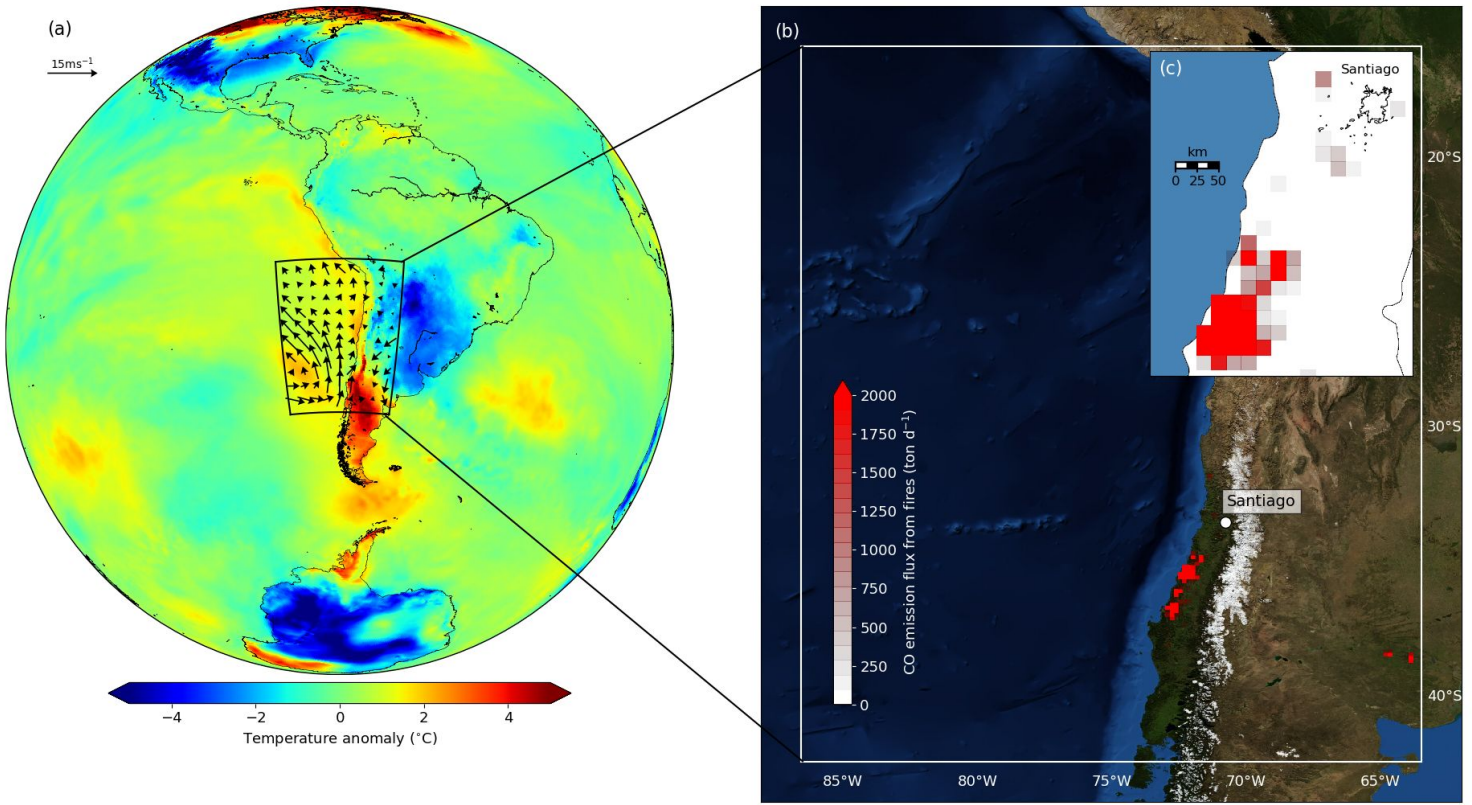


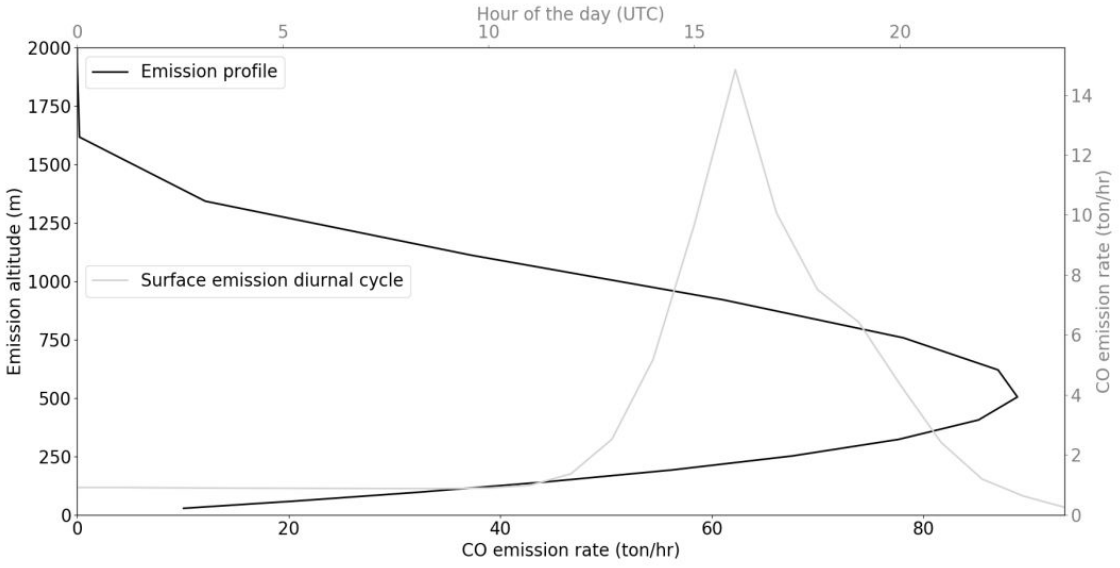
2017 fires

2017 fires – Simulation design



2017 fires – Simulation design

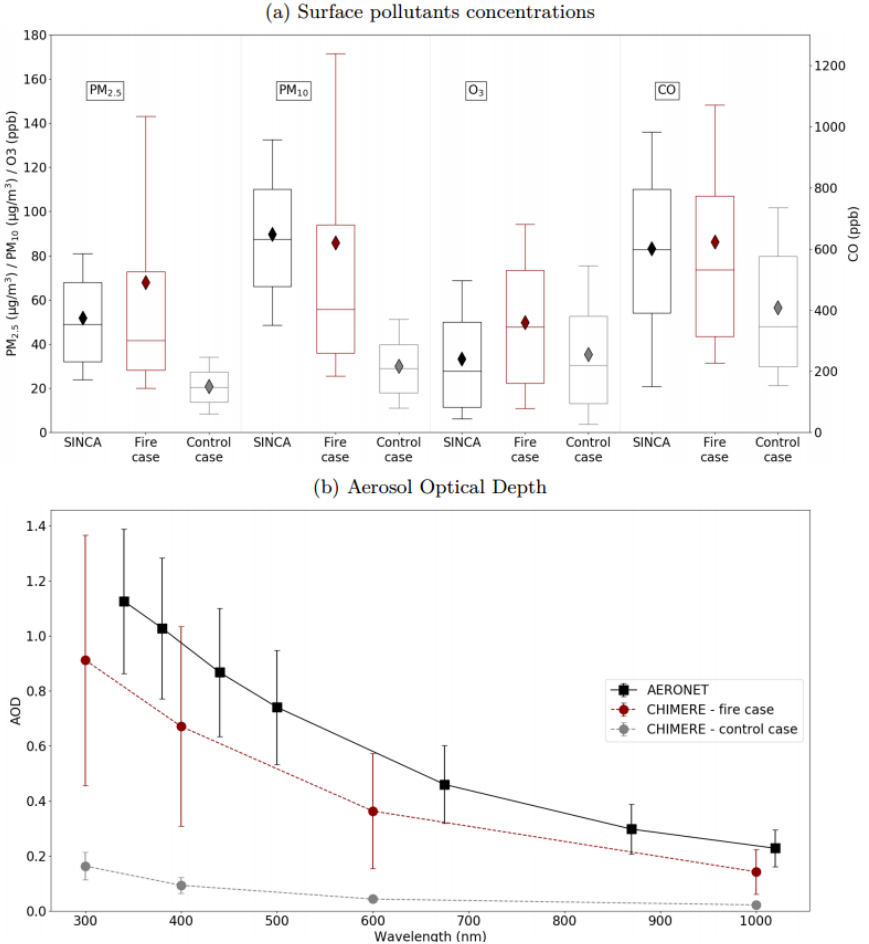
(a) CO emissions diurnal and vertical profiles on January 29

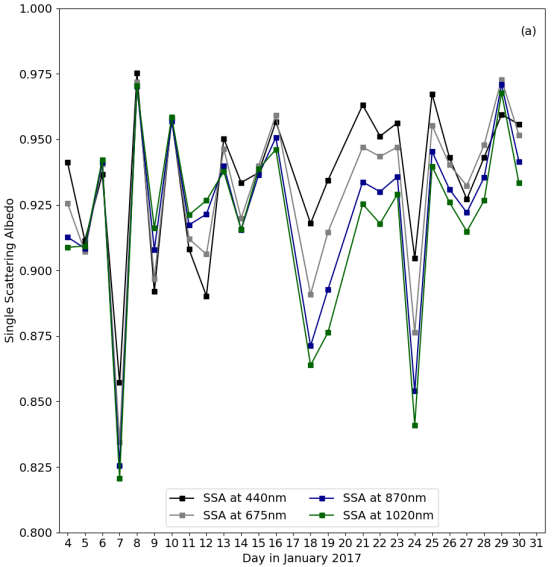


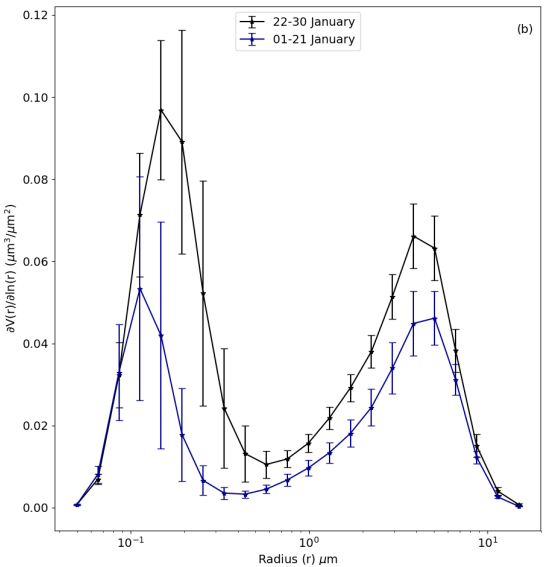
(b) Chemical composition at emission

Species		Ratio at emission
C2H4	Ethene	1.1%
C2H6	Ethane	0.4%
C3H6	Propene	0.9%
CH3CHO	Acetaldehyde	0.4%
CH3COE	Methyl Ethyl Ketone	0.1%
CH4	Methane	5.2%
CO	Carbon monoxide	82%
HCHO	Formaldehyde	0.9%
NC4H10	n-Butane	0.3%
NH3	Ammoniac	1.6%
NO	Nitrogen monoxide	1.5%
NO2	Nitrogen dioxide	0.2%
SO2	Sulphur dioxide	0.2%
TOL	benzene, toluene and other mono-substituted aromatics	0.1%
OC	Organic carbon	2.1%
BC	Black carbon	0.2%
PPM_fin	fine PPM	1.8%
PPM_coa	coarse PPM	1.3%

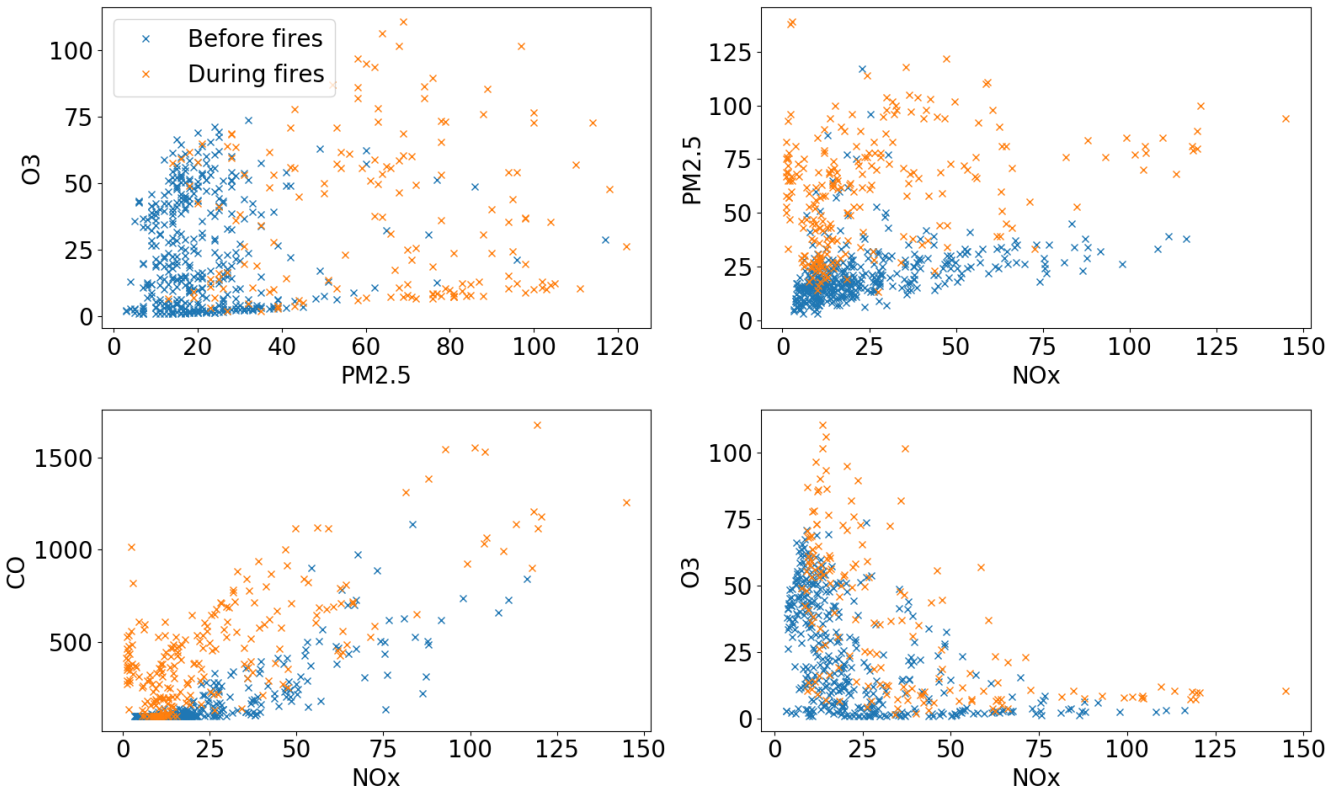
2017 fires – Simulation validation







2017 fires – Additional material



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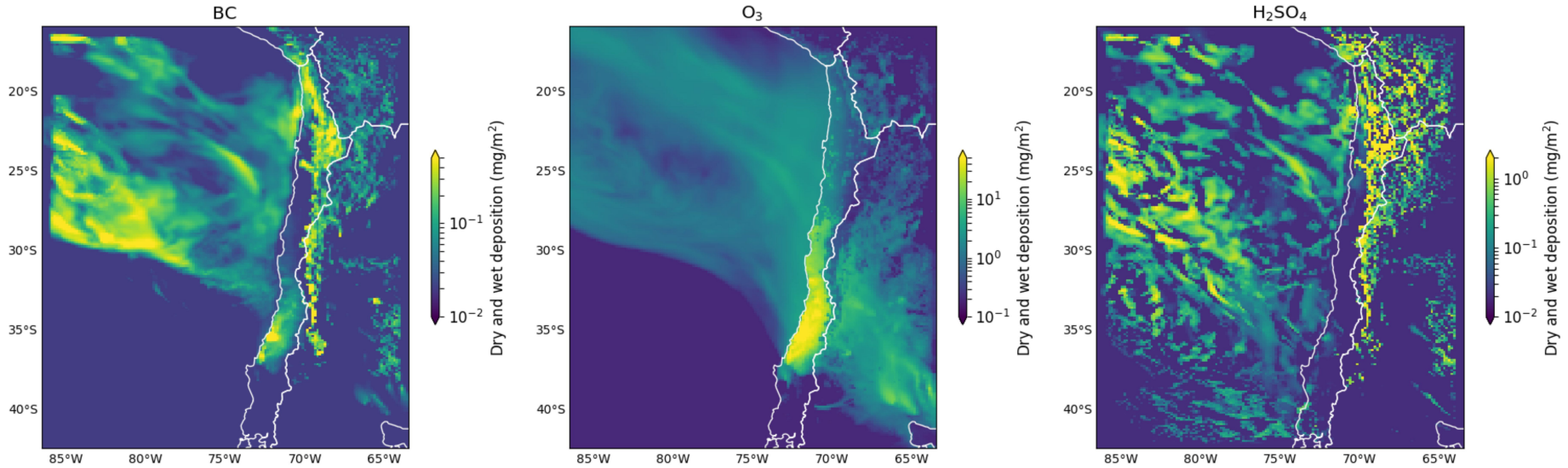

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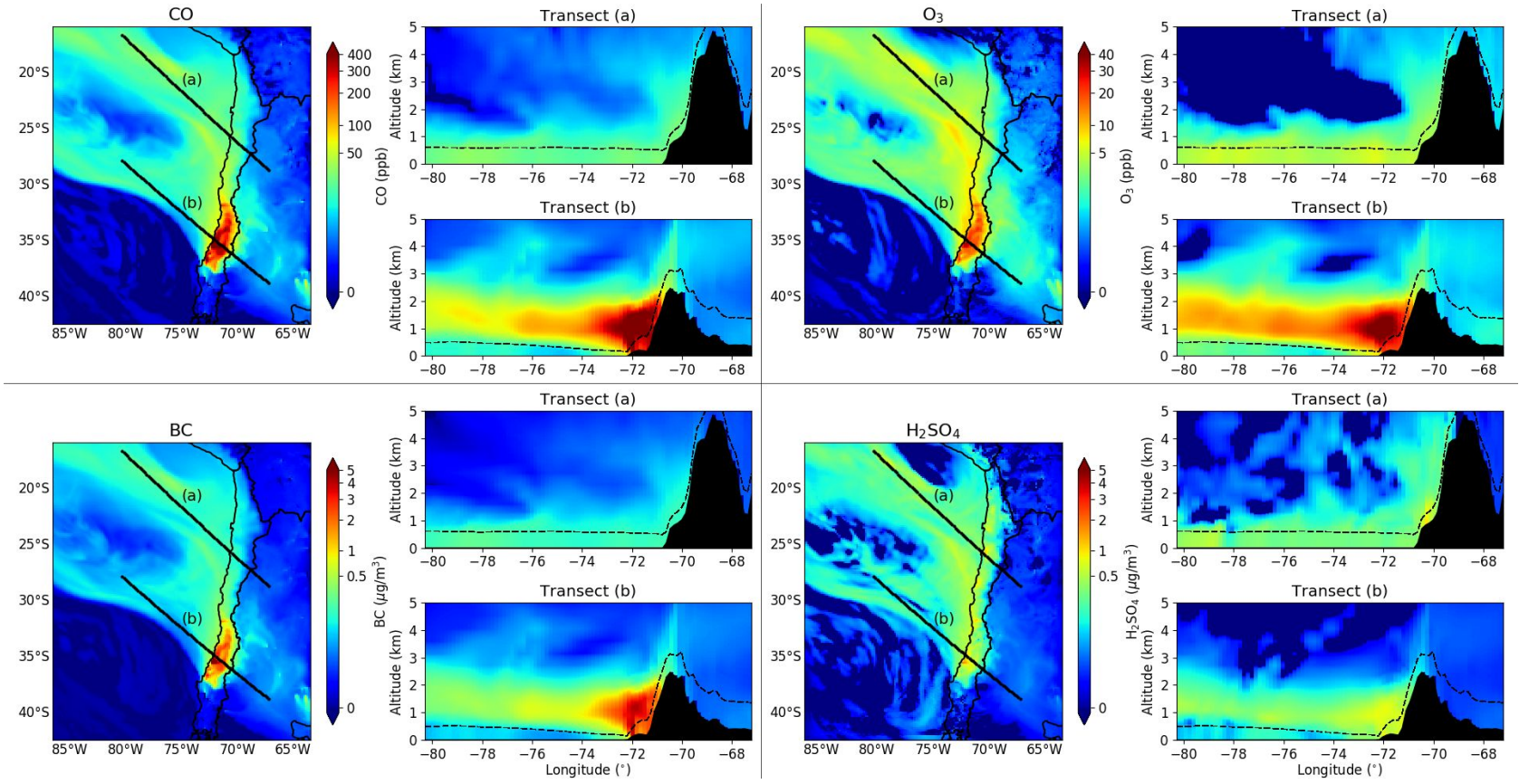

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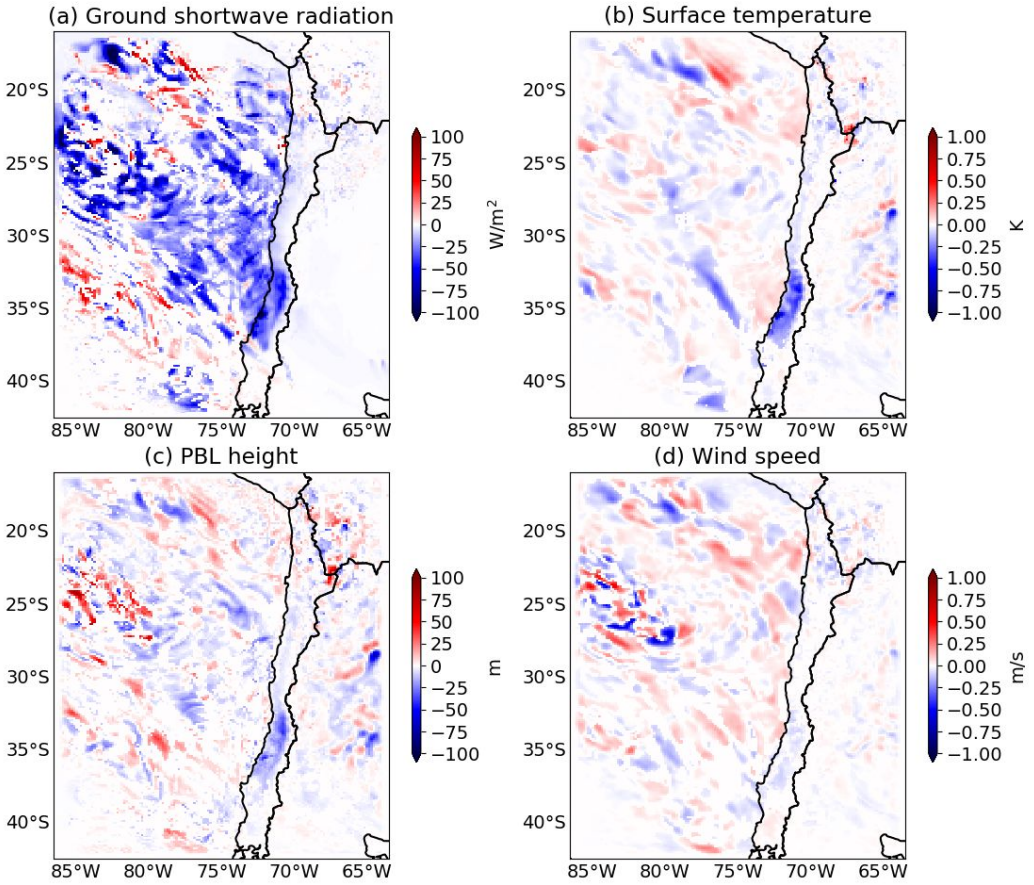

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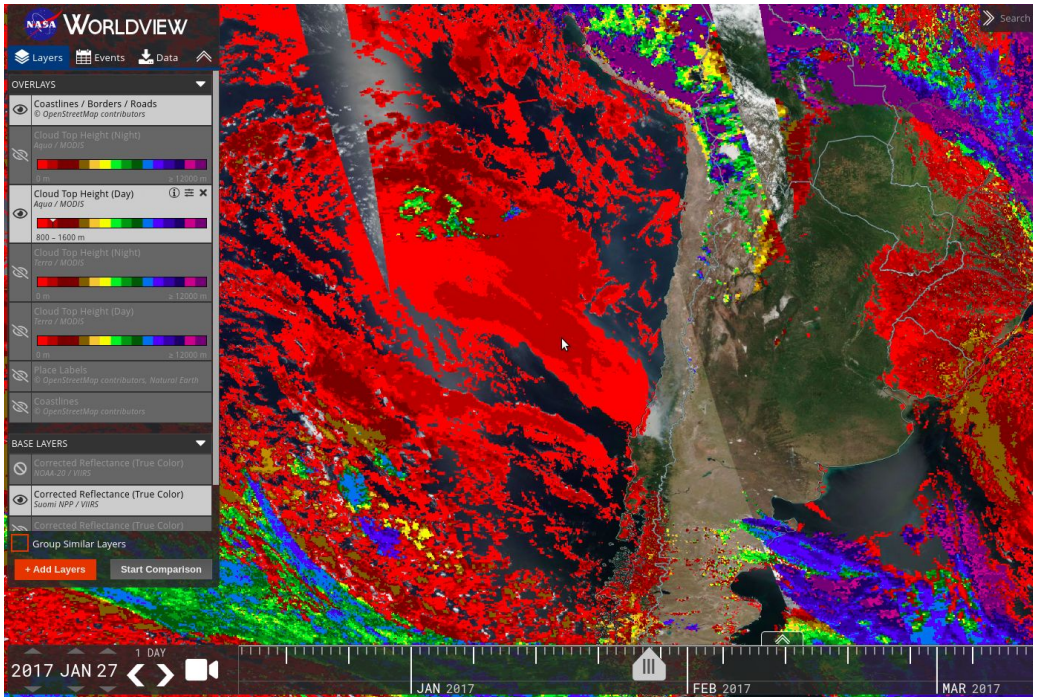

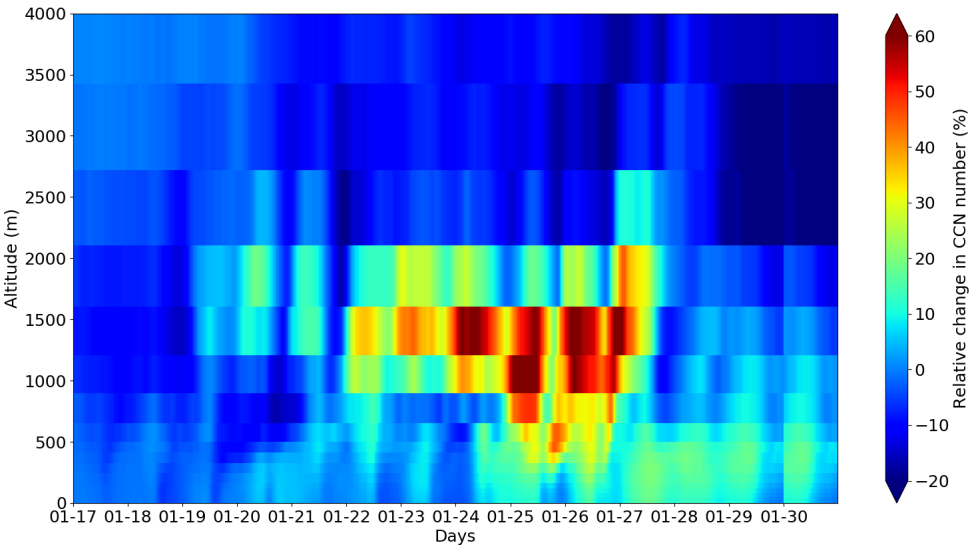

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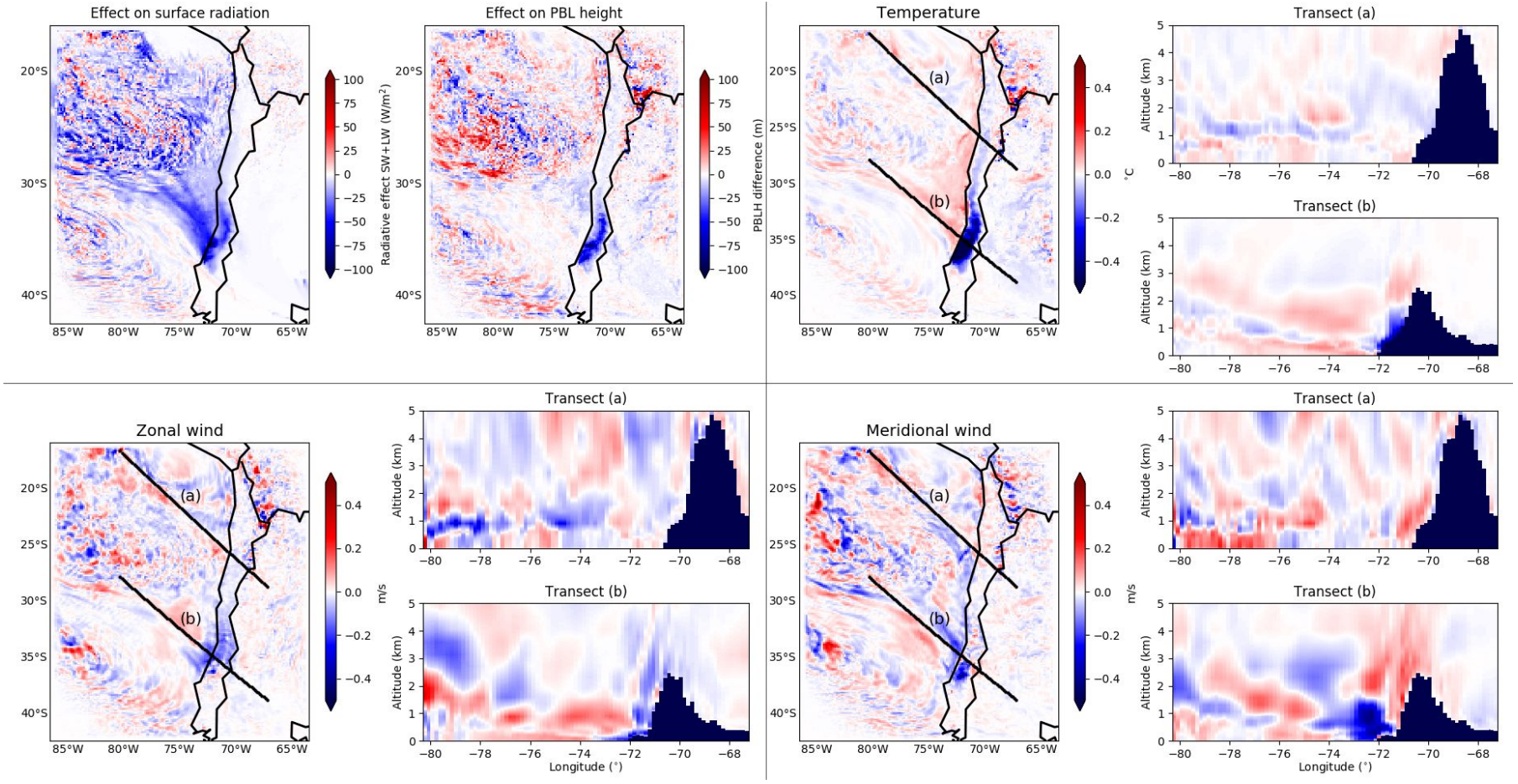

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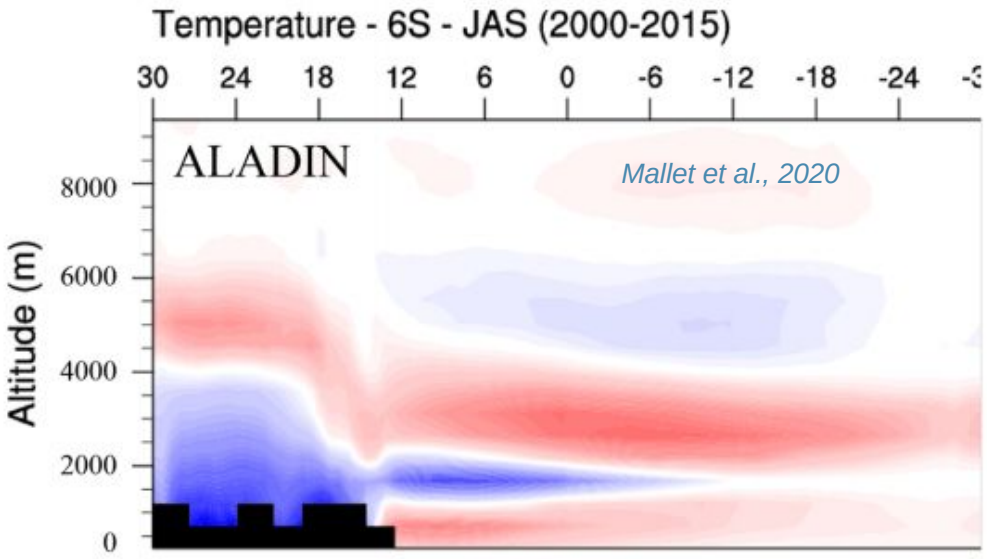
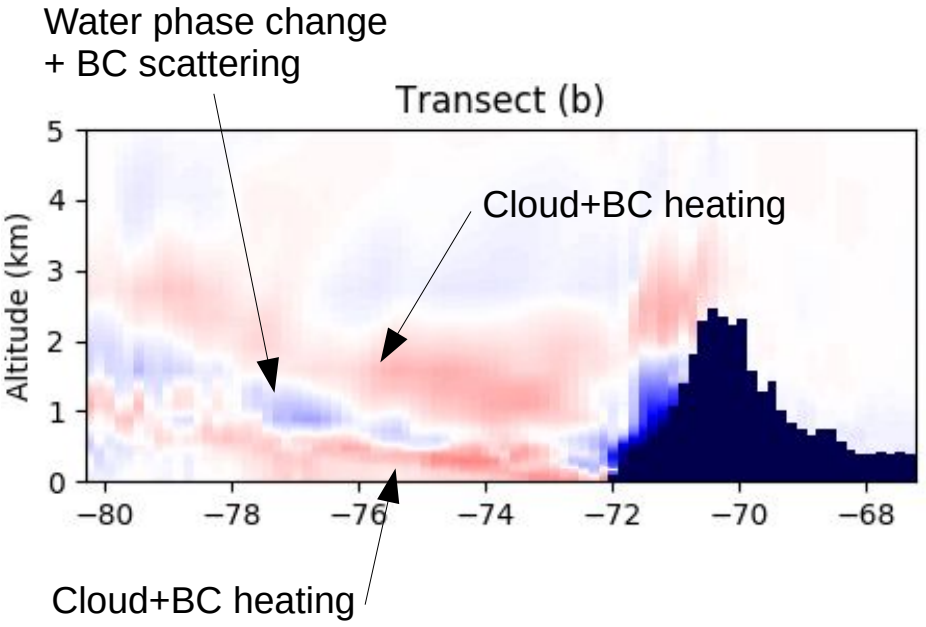
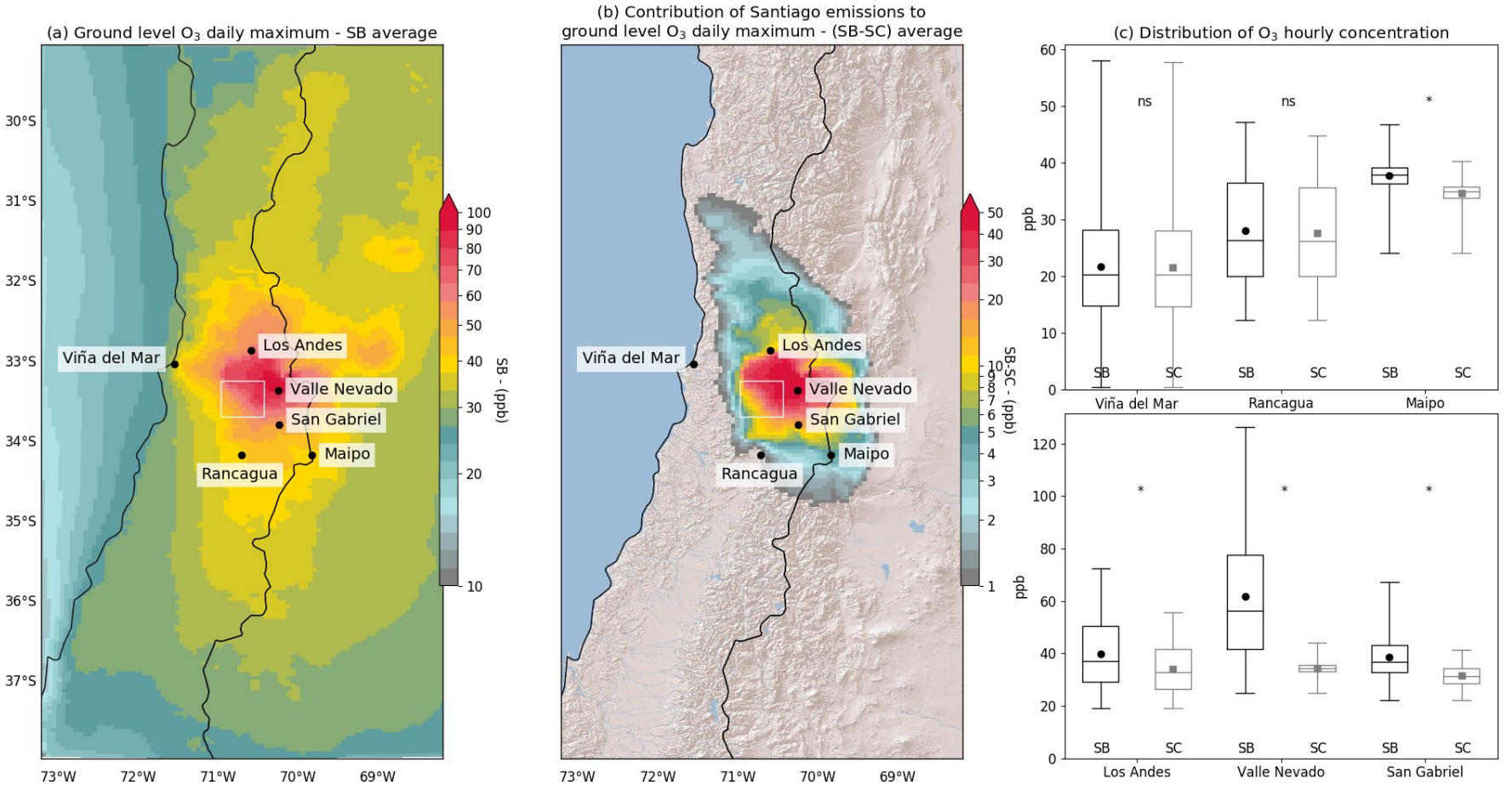




Regional transport

Regional transport – Additional material



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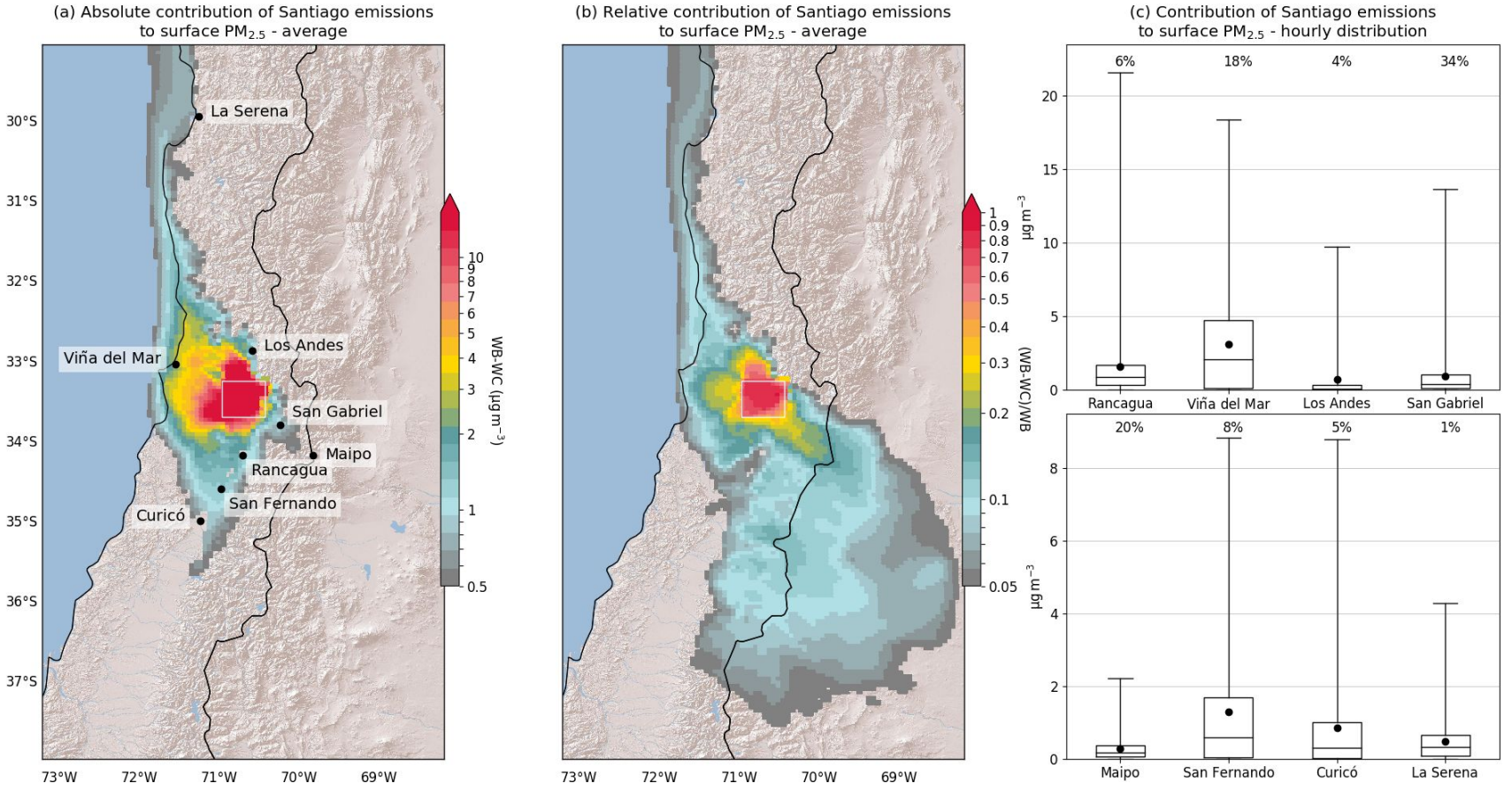

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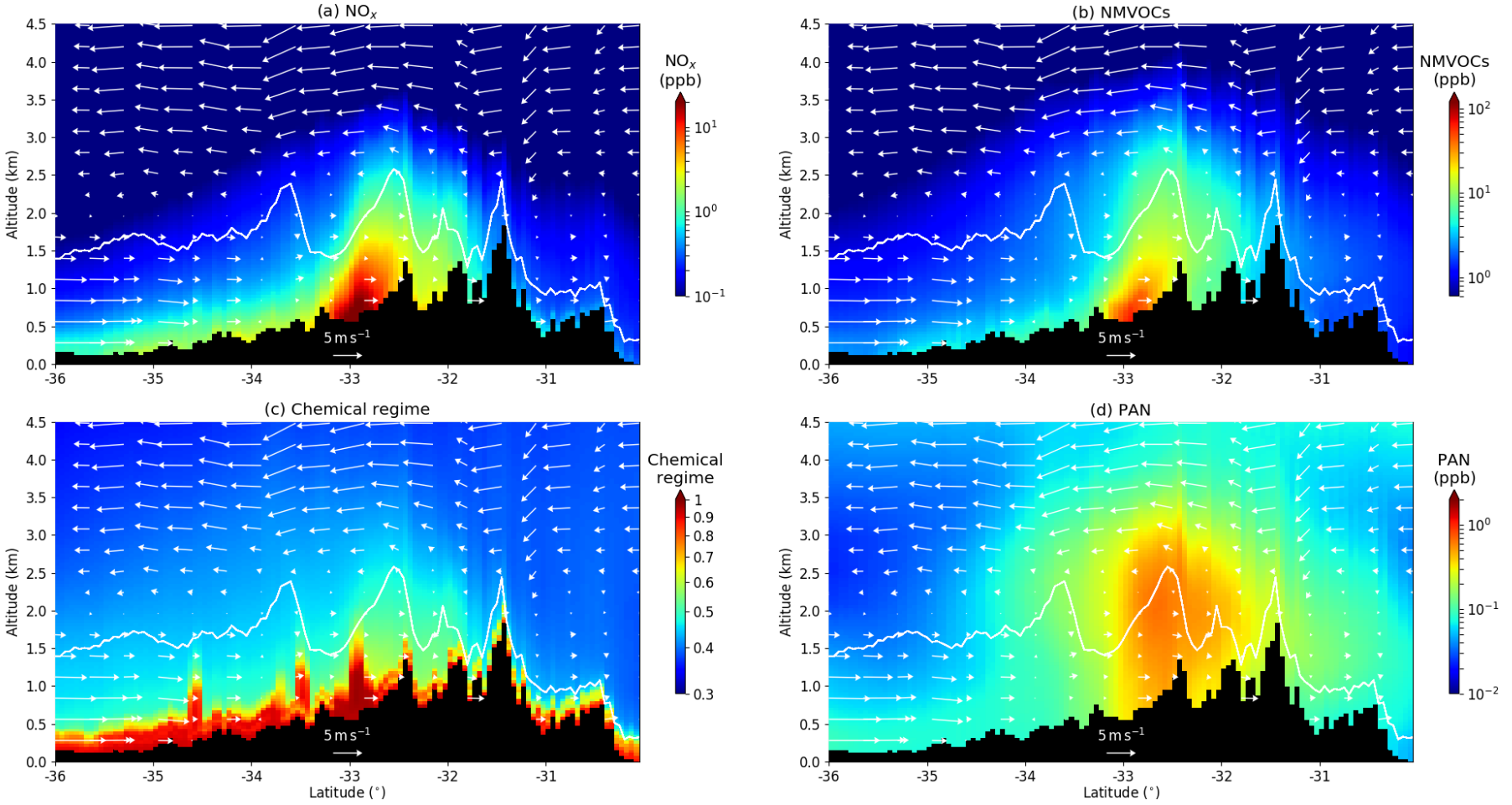

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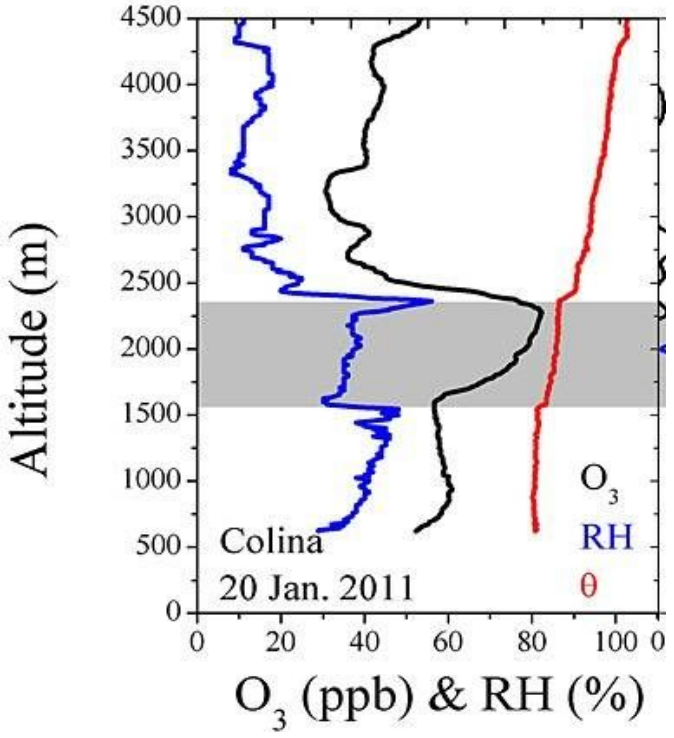
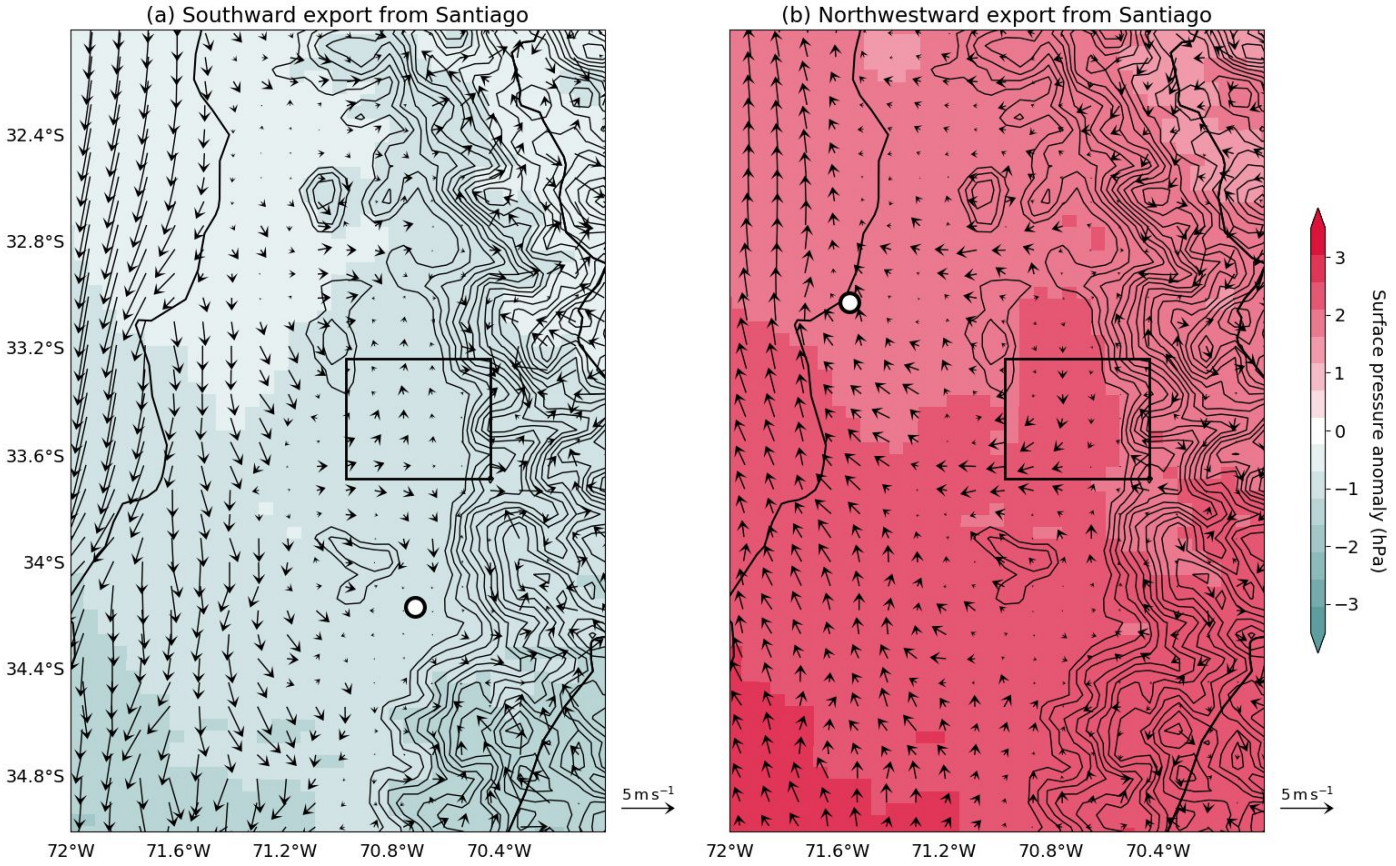


(Seguel et al., 2013)

Regional transport – Additional material



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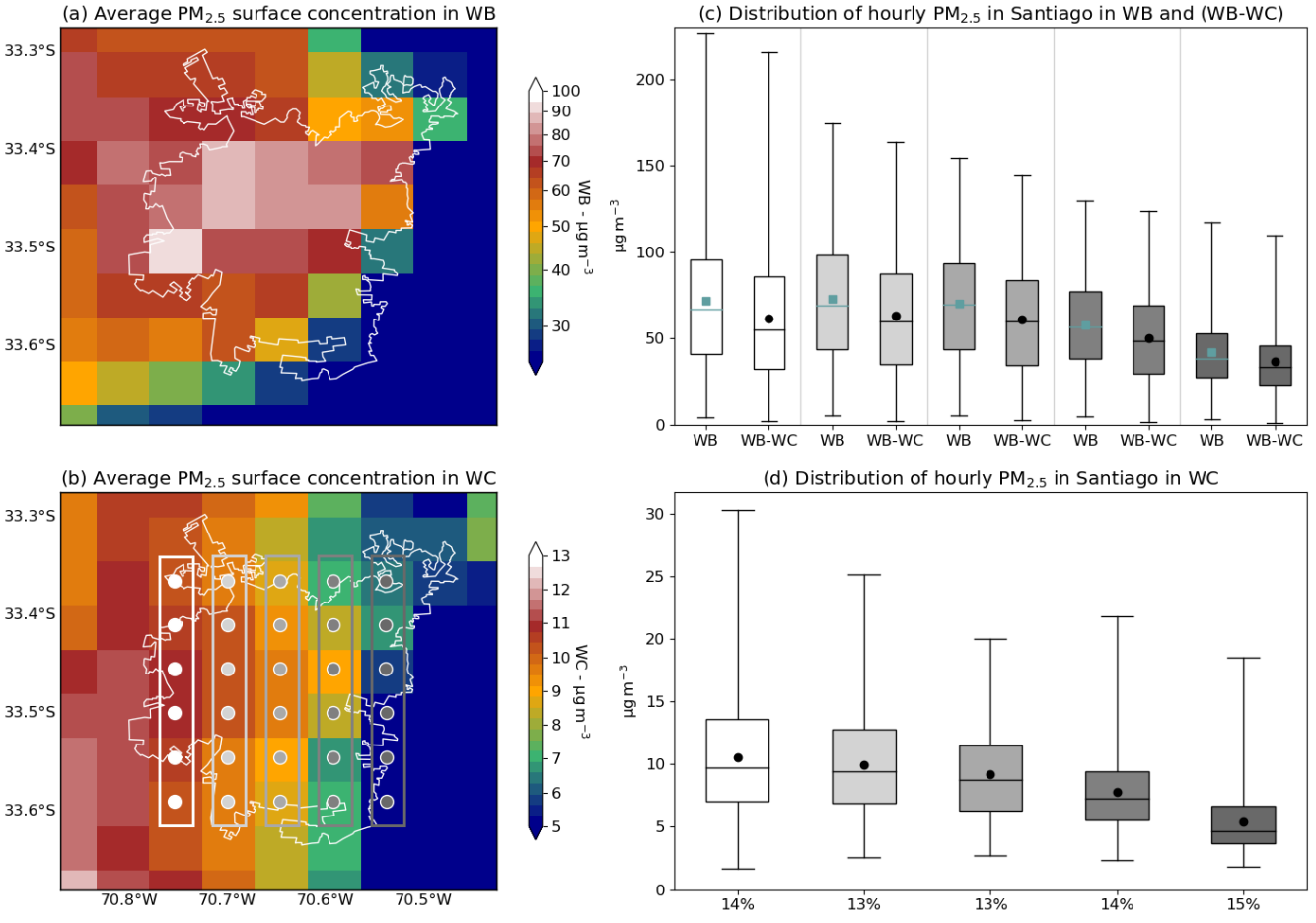

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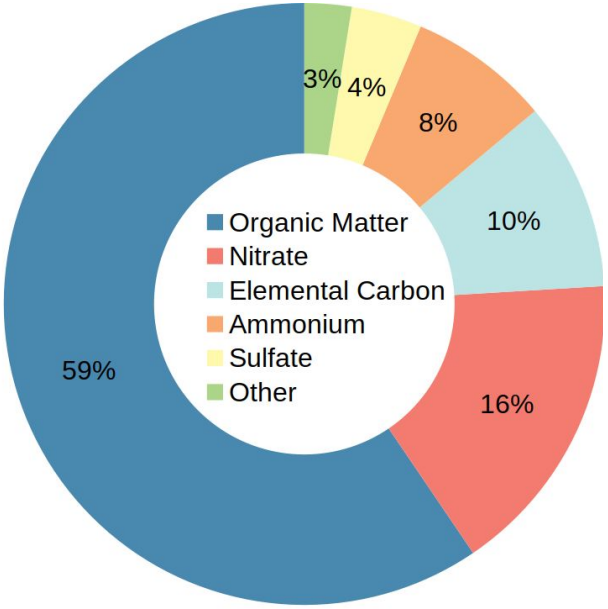
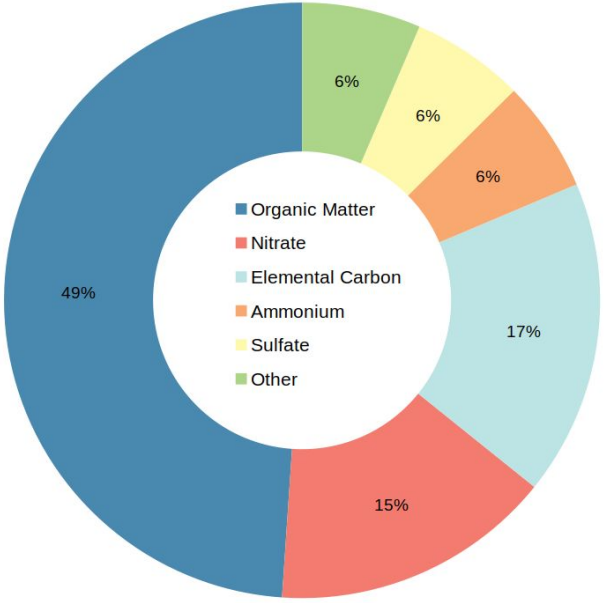
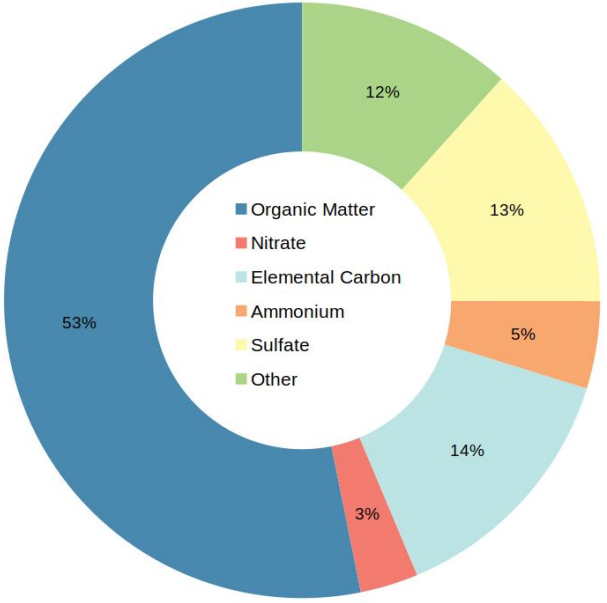


Bulk composition of PM2.5 in Santiago - June 2013.

Adapted from *Villalobos et al., 2015*

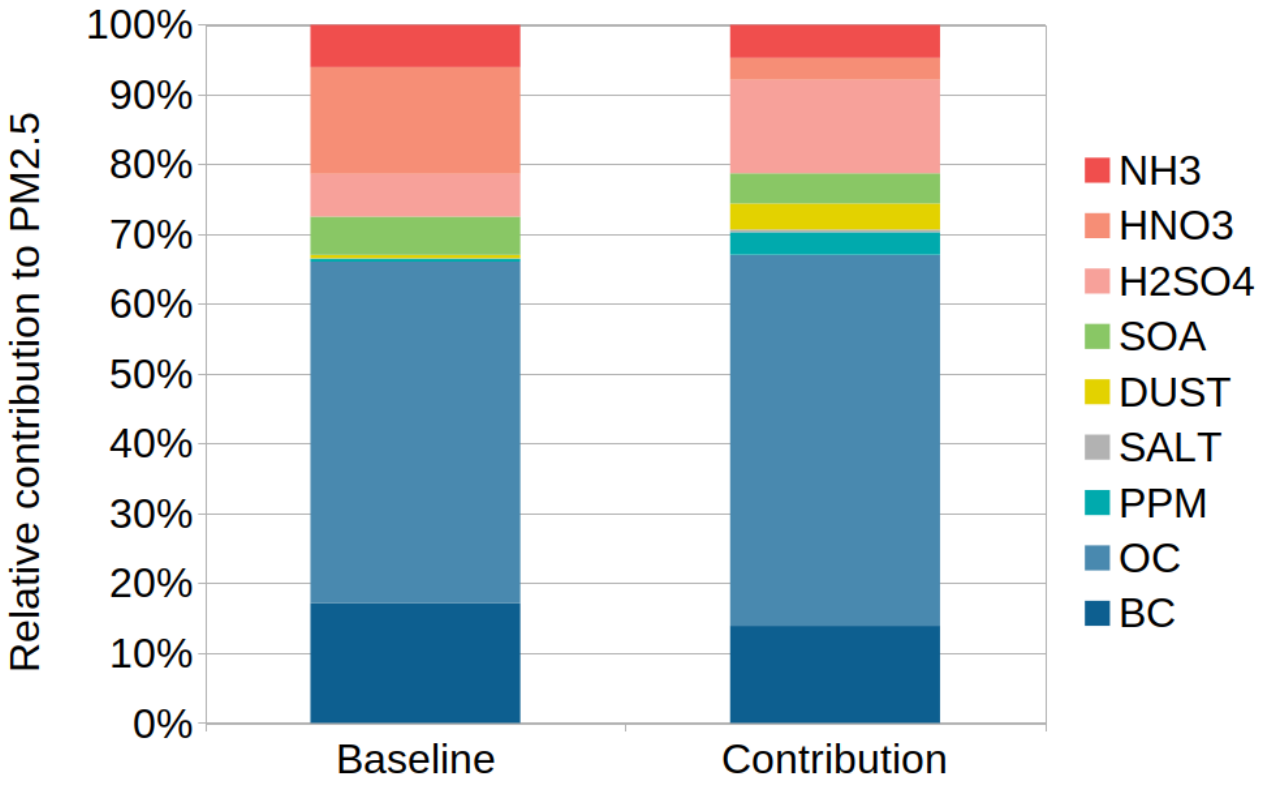


Composition of PM2.5 in Santiago – CHIMERE simulation July 2015



Composition of PM2.5 **transported to** Santiago – CHIMERE simulation July 2015

Regional transport – Additional material



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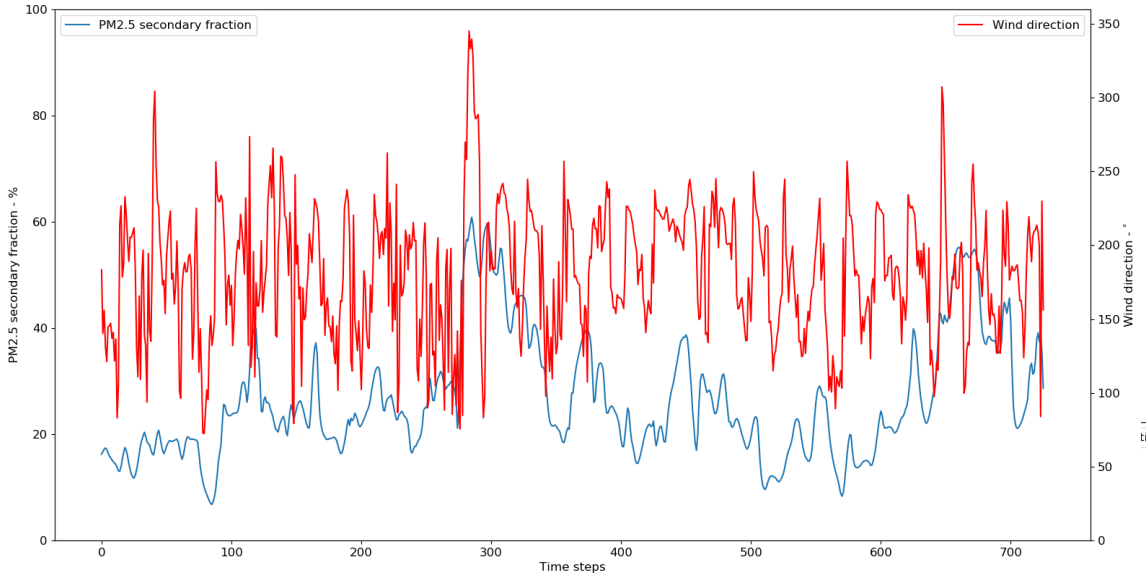
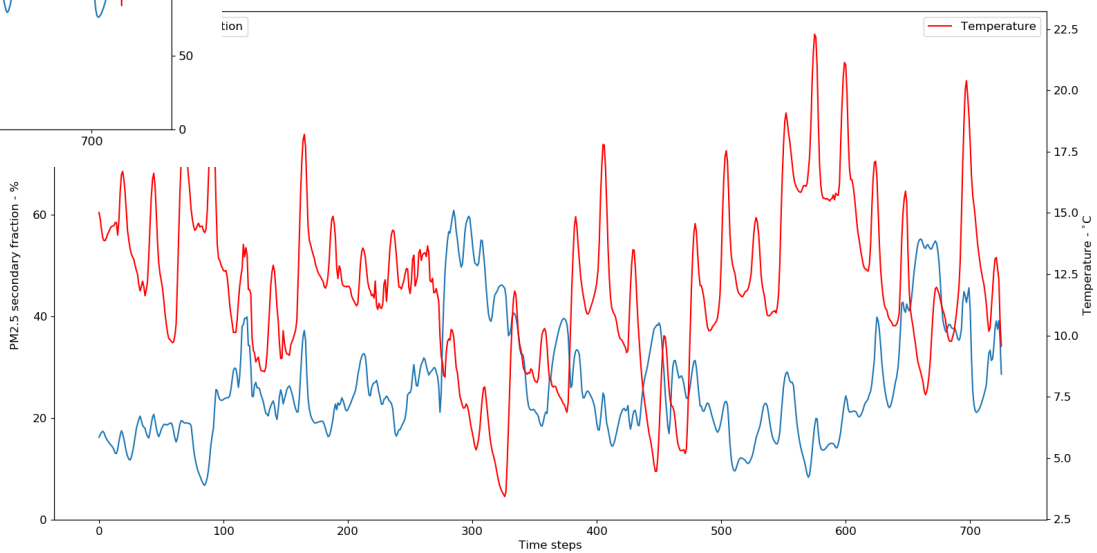


WB: 33% secondary
WC: 25% secondary



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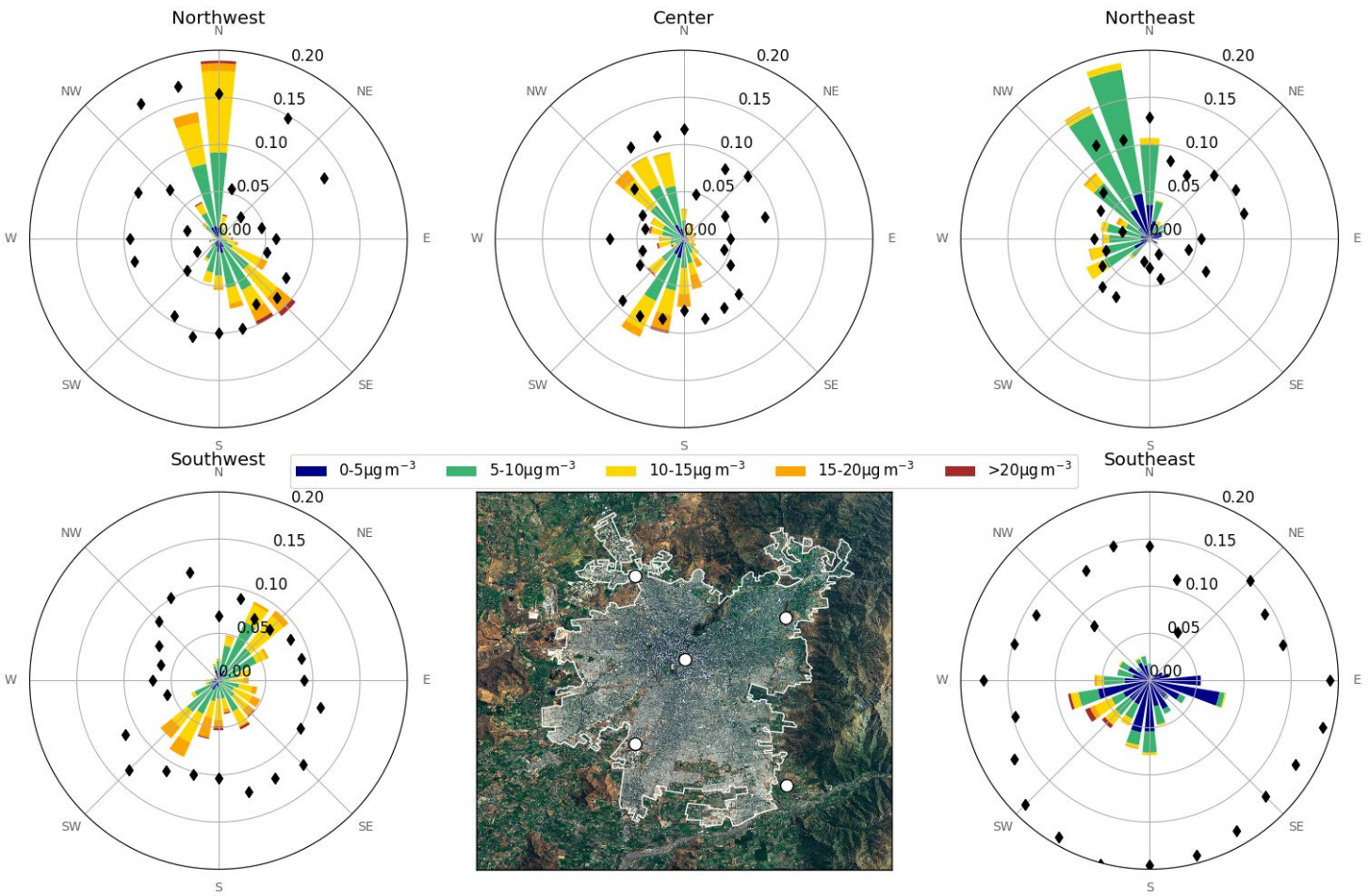

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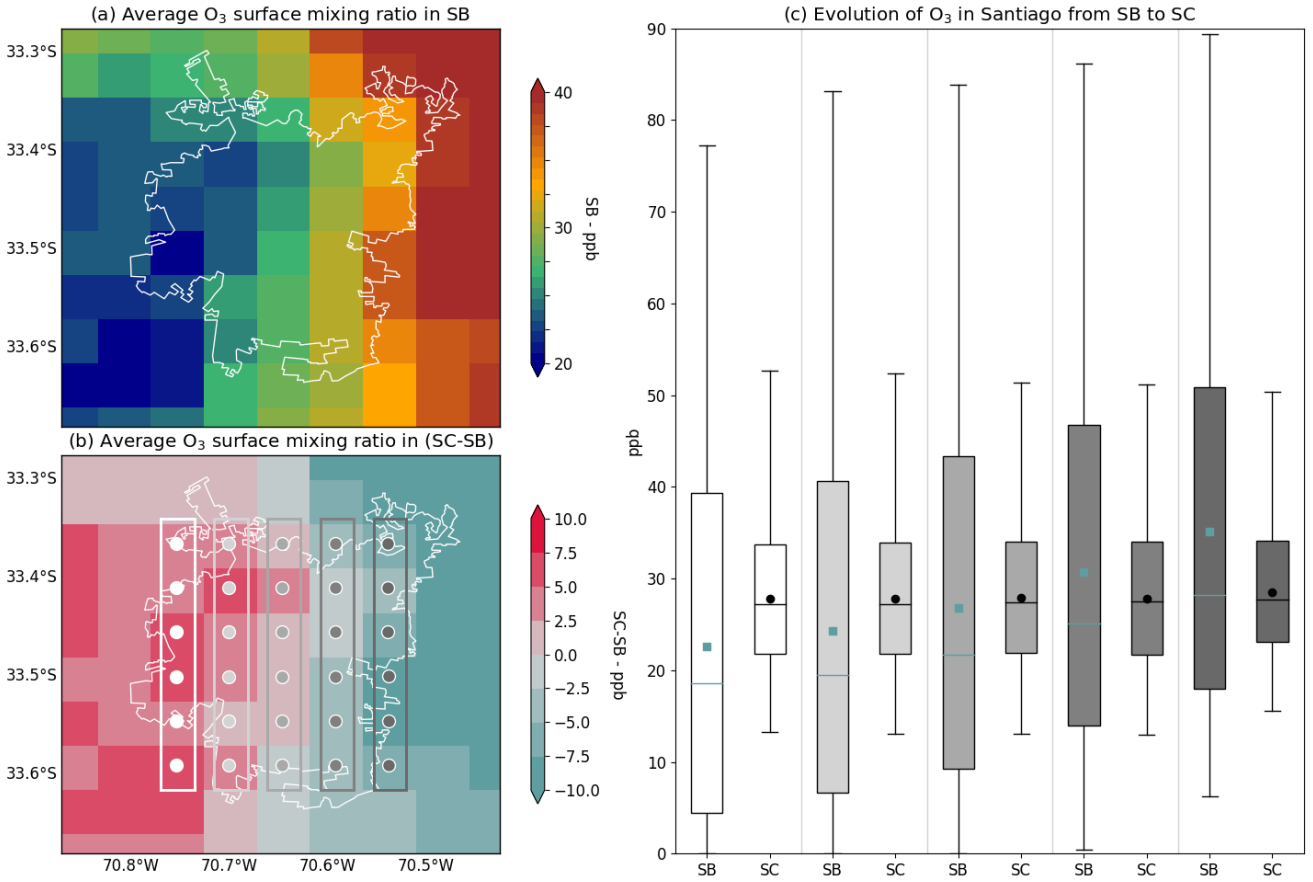
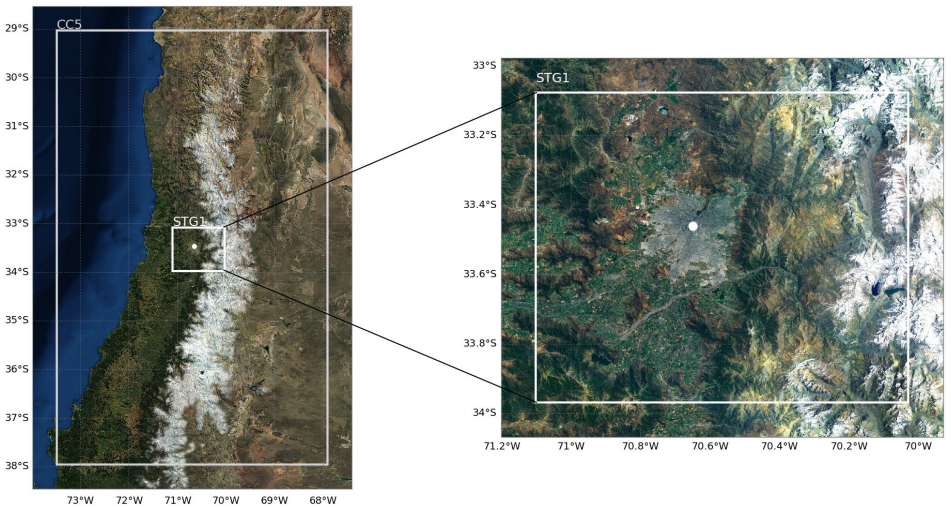


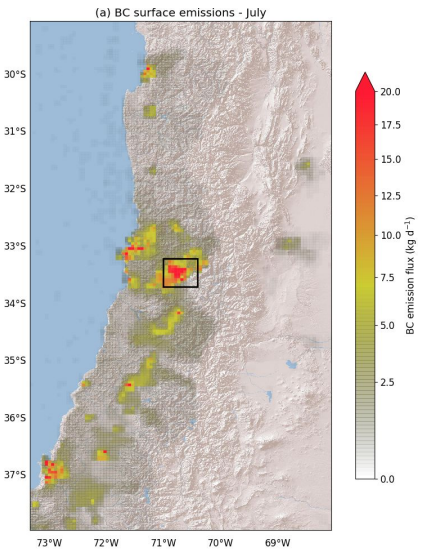
Black carbon and the Andes

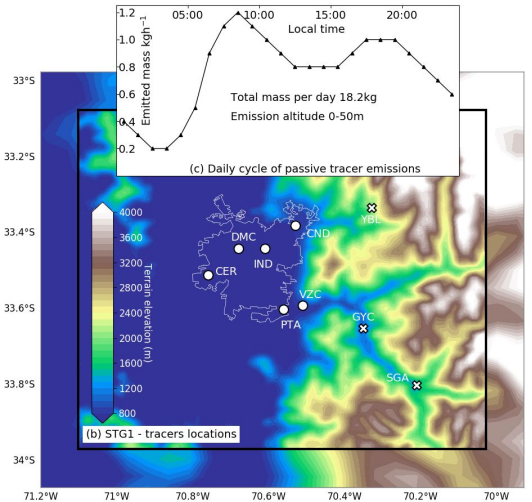
Simulation design

2 domains / 2 objectives

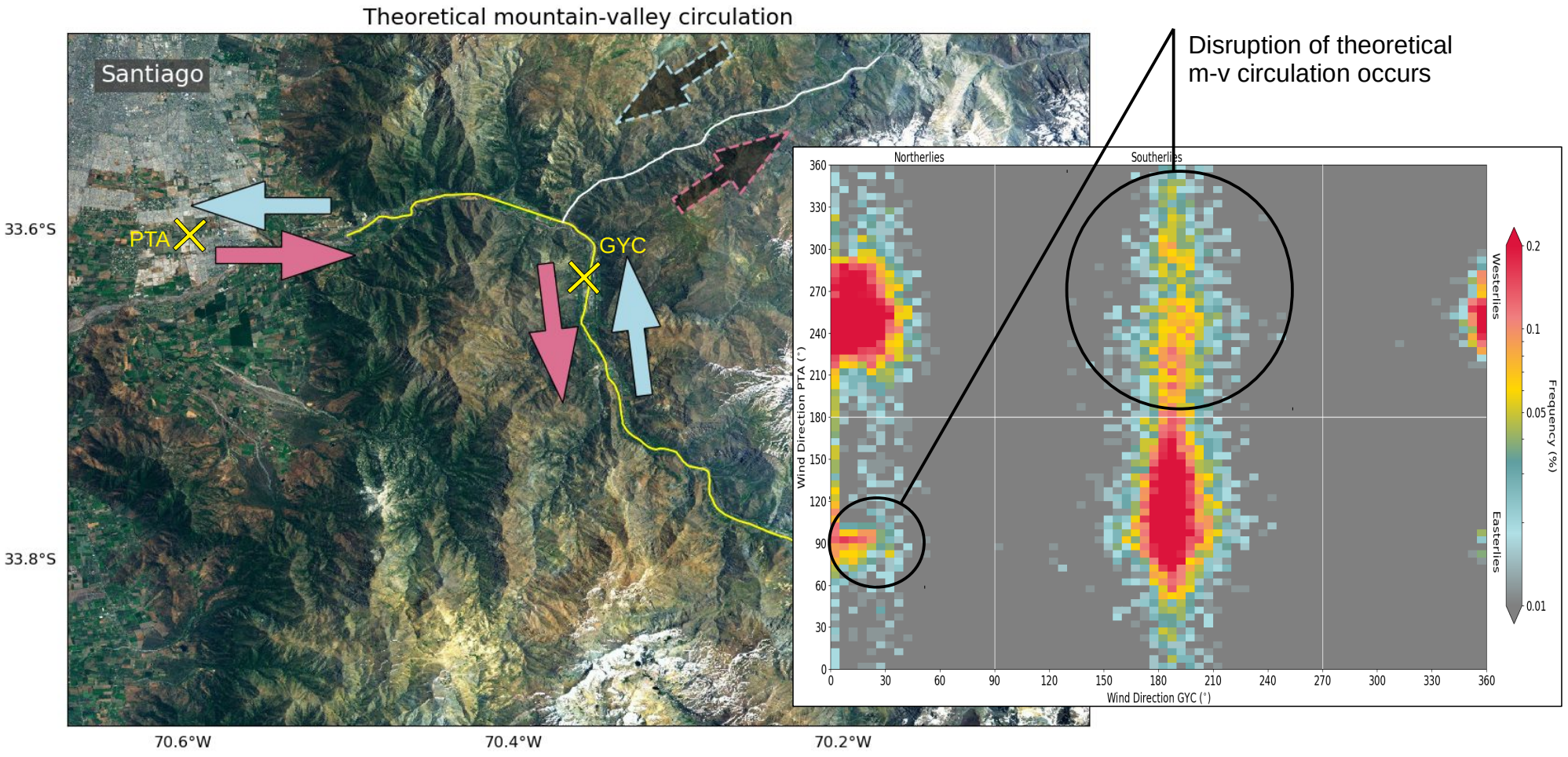
Domain	Horizontal resolution	Purpose	Key output
CC5	5km (100x200 grid points)	Scenario on BC urban emissions	Contribution of BC from Santiago to deposition on glaciers
STG1	1km (100x100 grid points)	High resolution transport of tracers without chemistry	Pathways of aerosols from Santiago to the glaciers



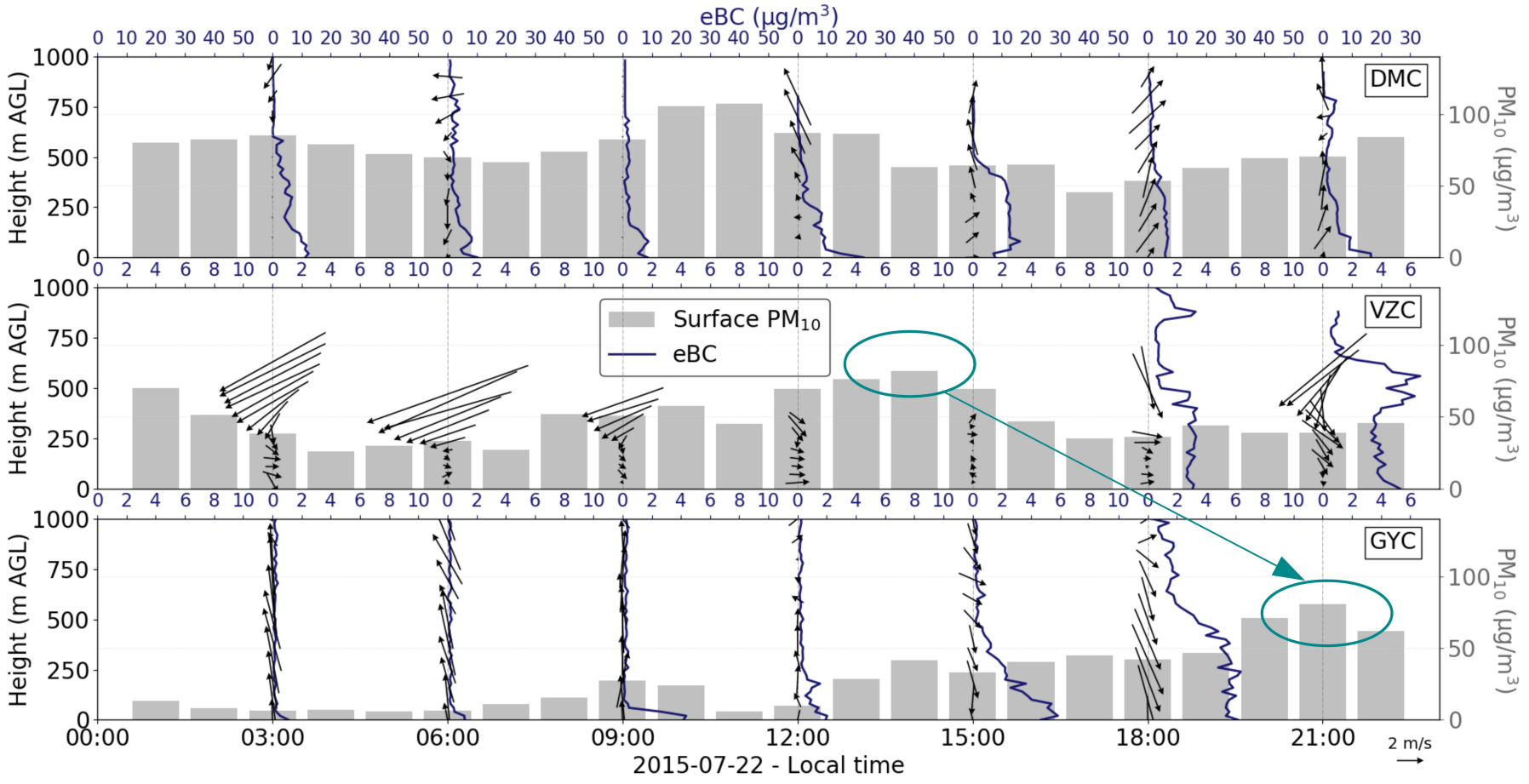




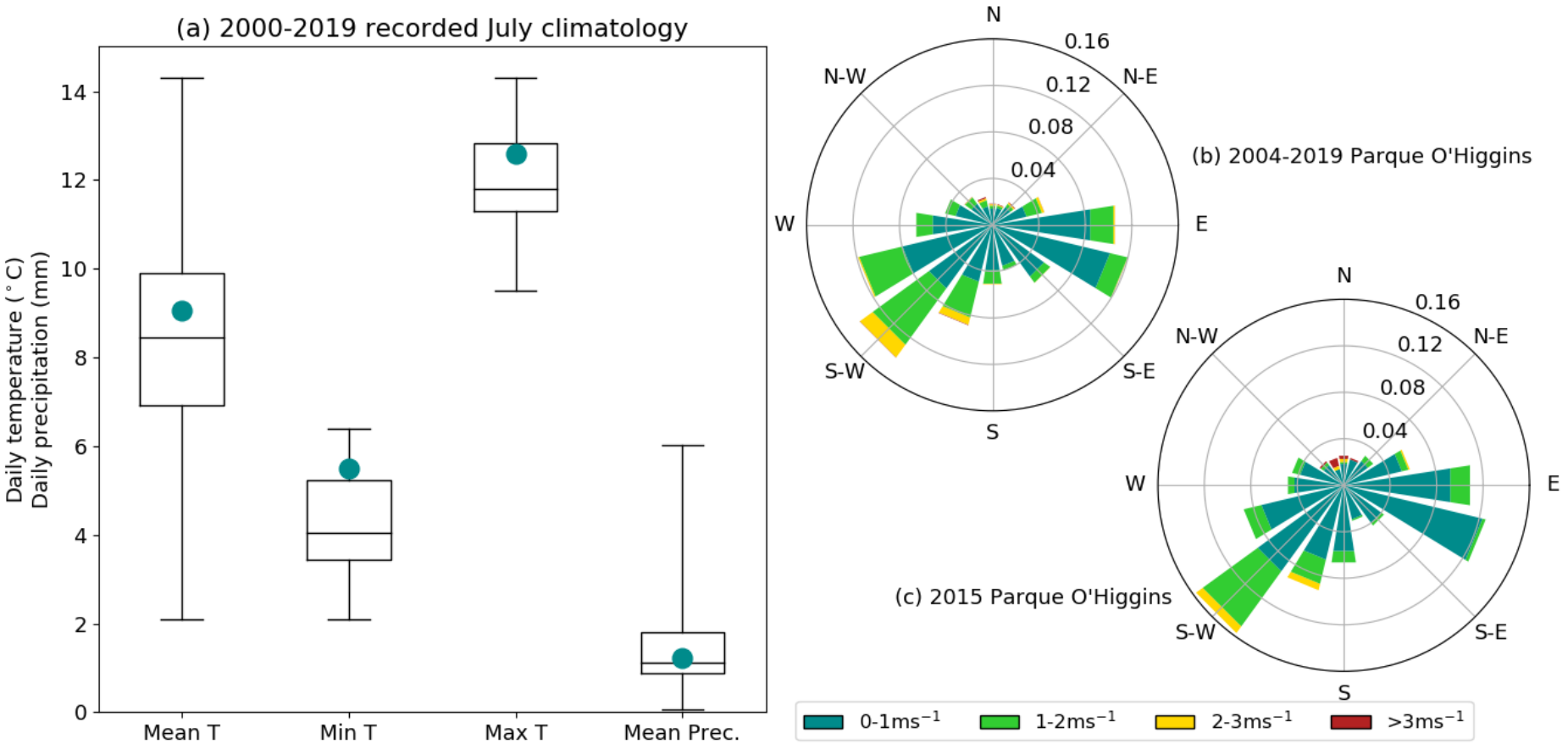
Mountain-valley circulation



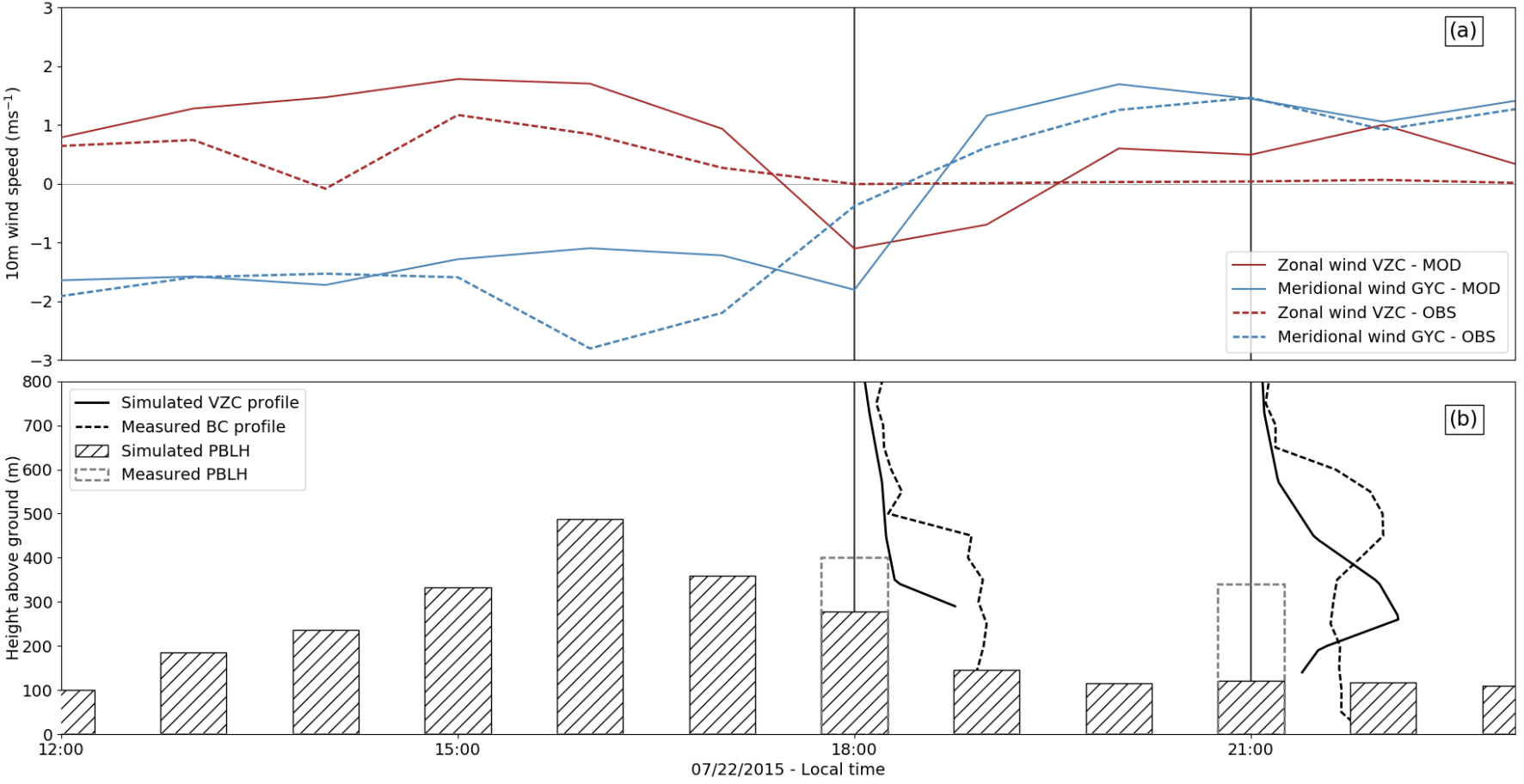
Mountain-valley circulation



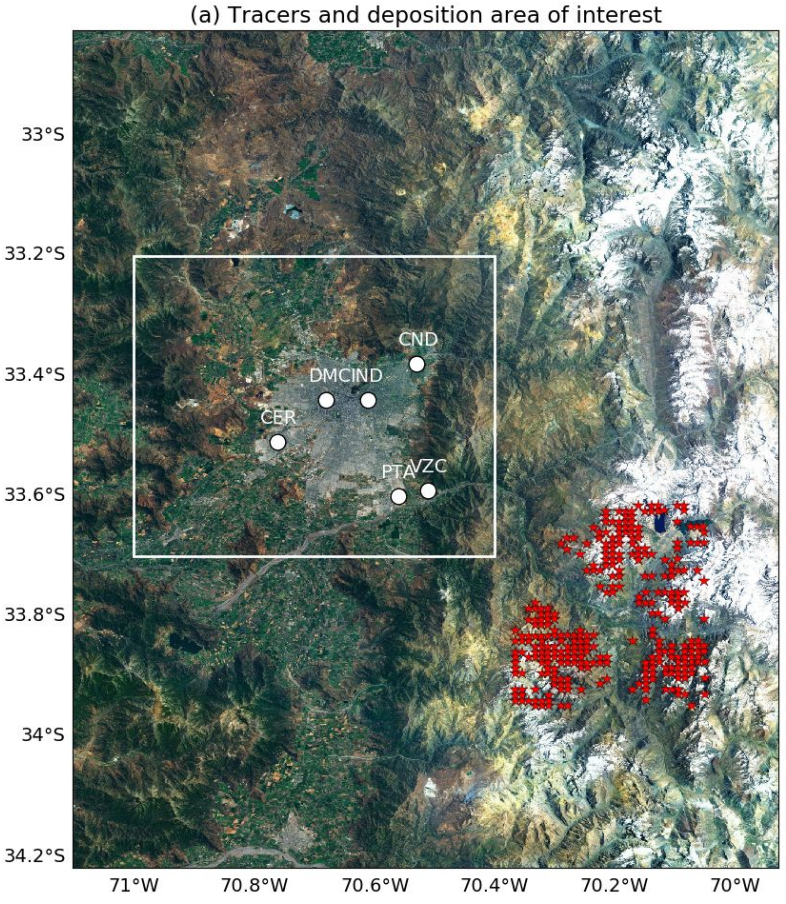
Climatological relevance

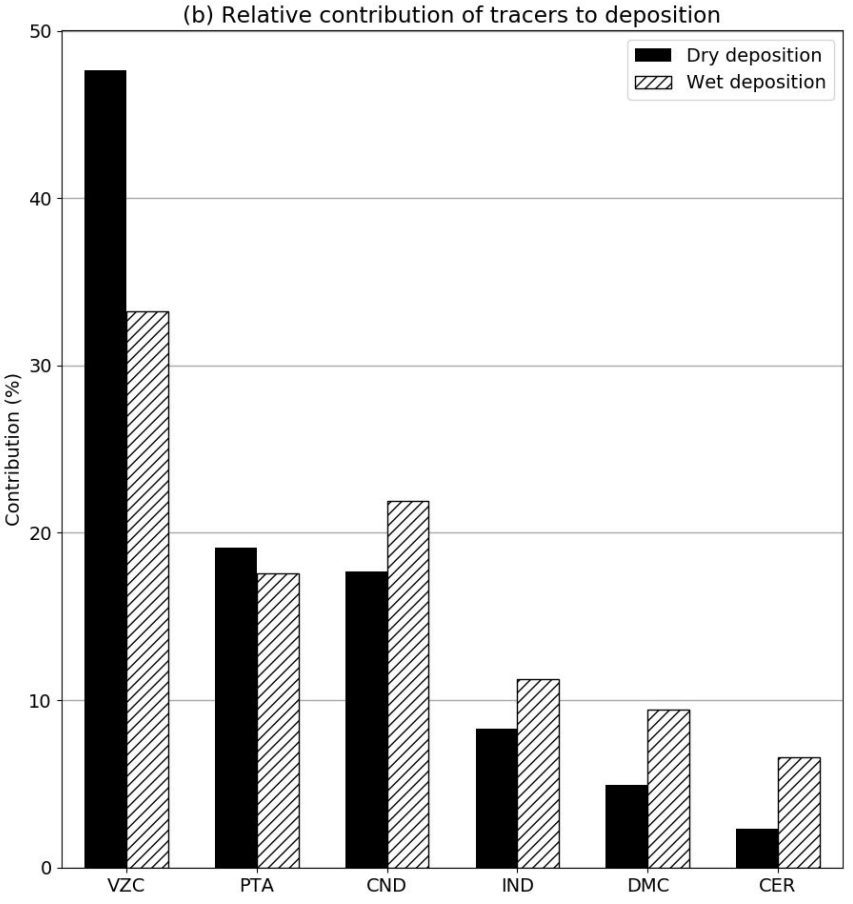


Simulation relevance



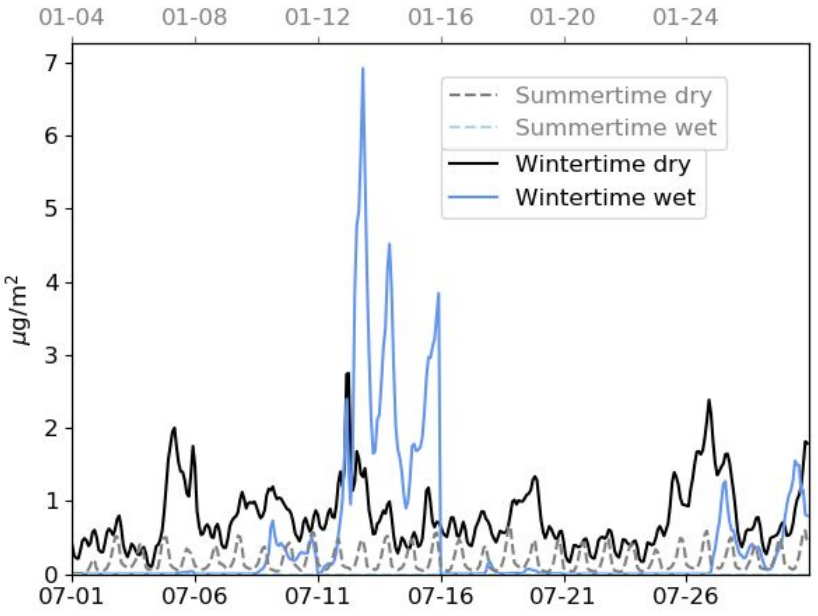
PM origin matters



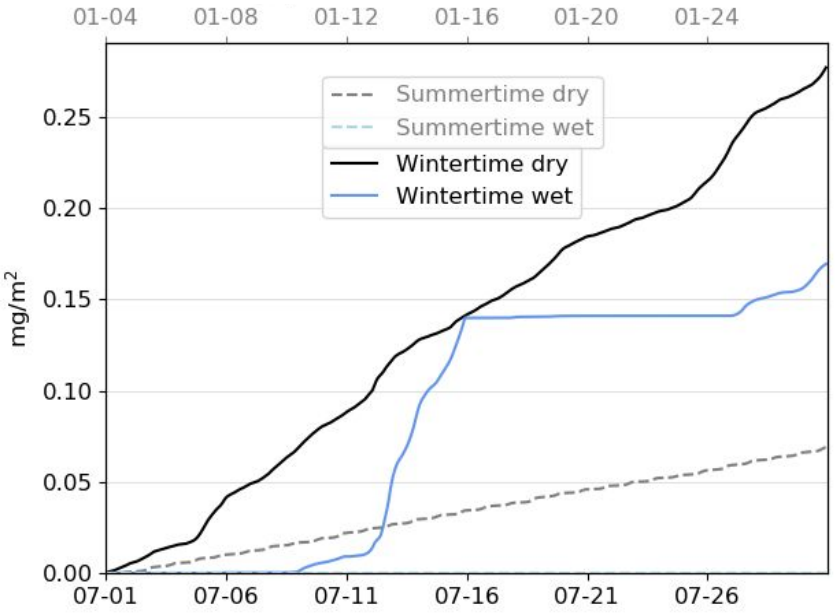


Deposition components and time series

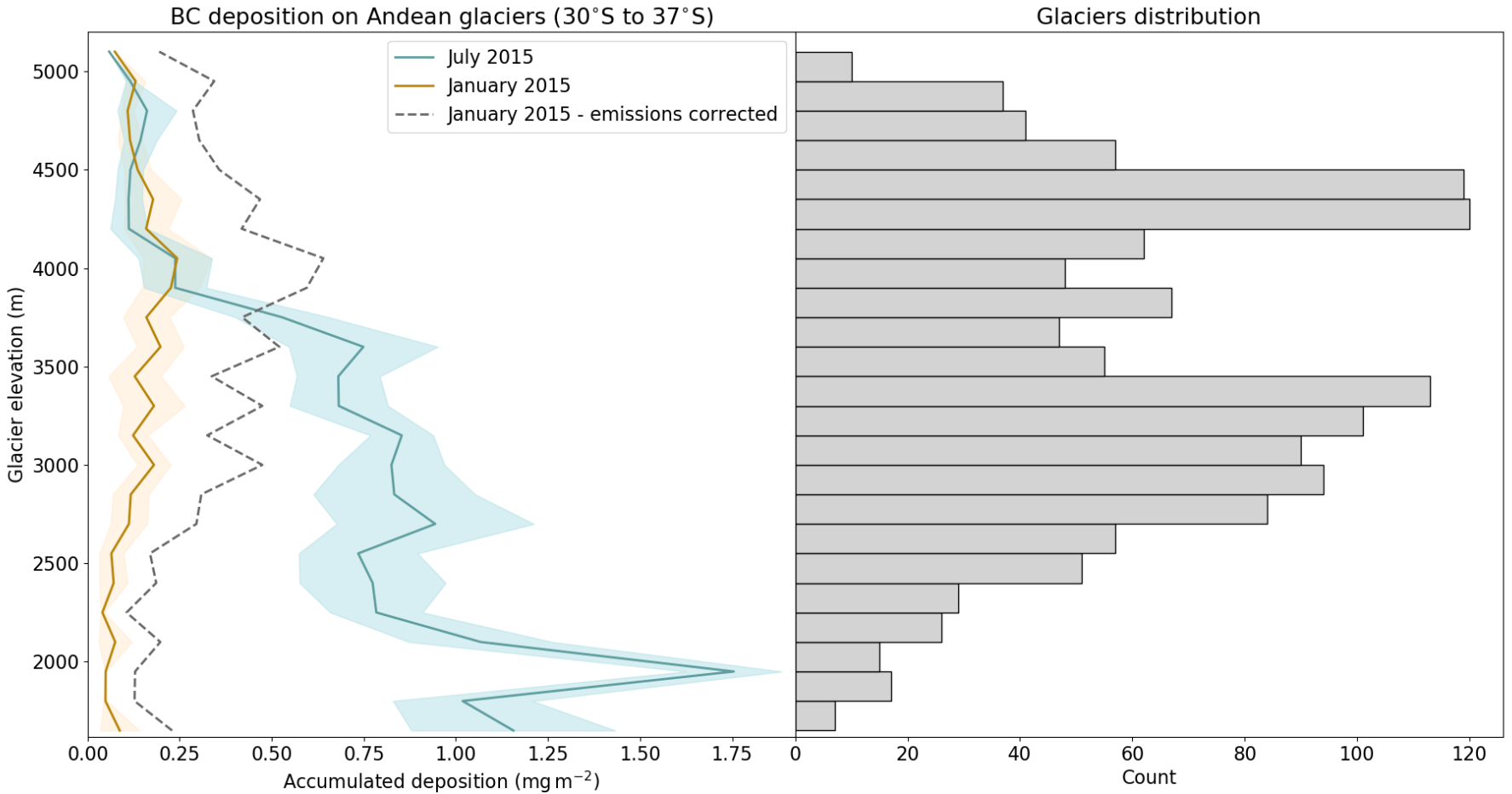
BC deposition flux – average on glaciers



Accumulated BC deposition – average on glaciers



Additional material



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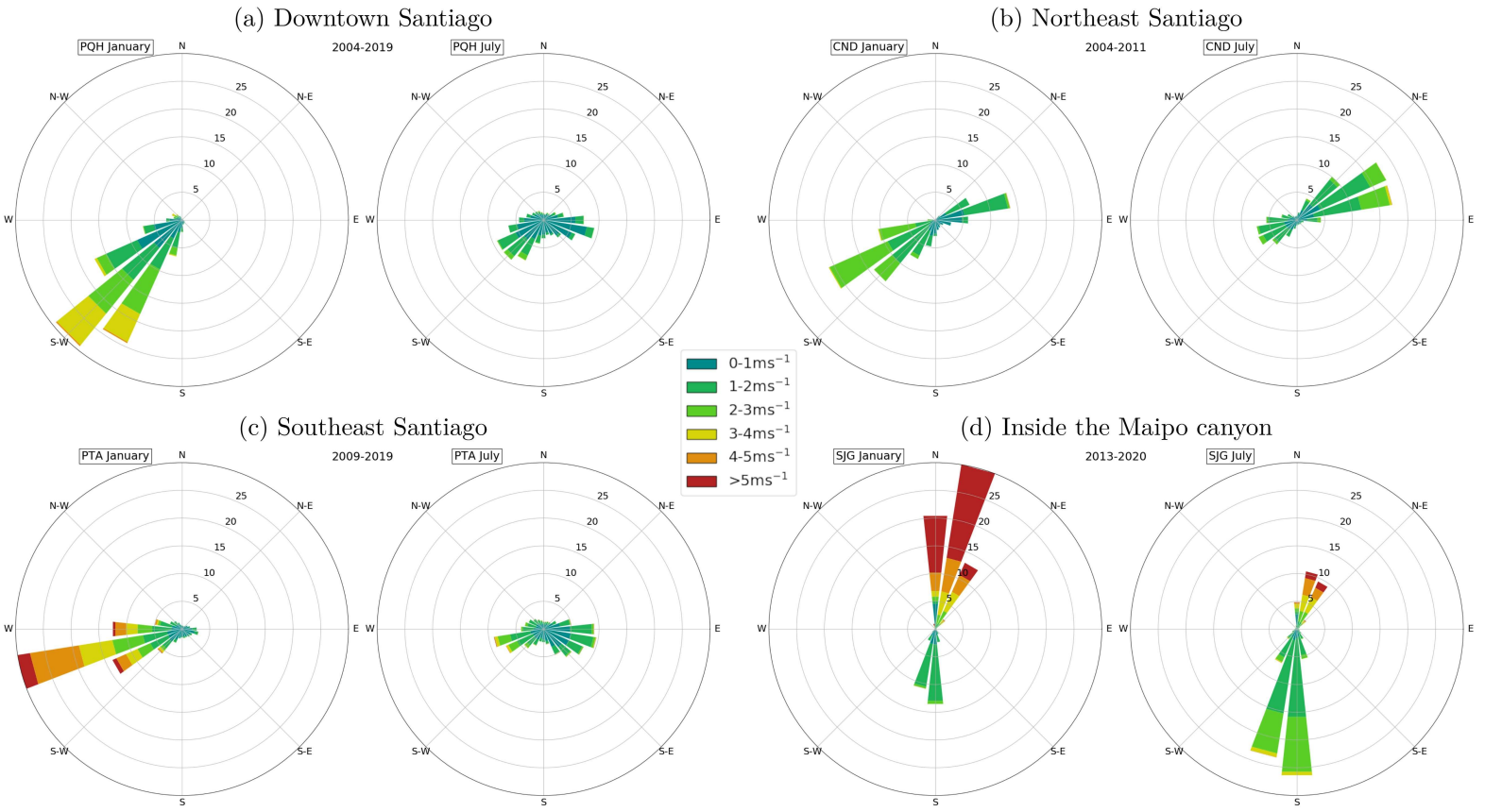

Additional material

