BIOMASS BURNING AND NATURAL EMISSIONS IN THE BRAZILIAN AMAZON RAINFOREST: IMPACT ON THE OXIDATIVE CAPACITY OF THE ATMOSPHERE

Fernando Santos
Instituto Nacional de Pesquisas Espaciais – INPE/Brasil

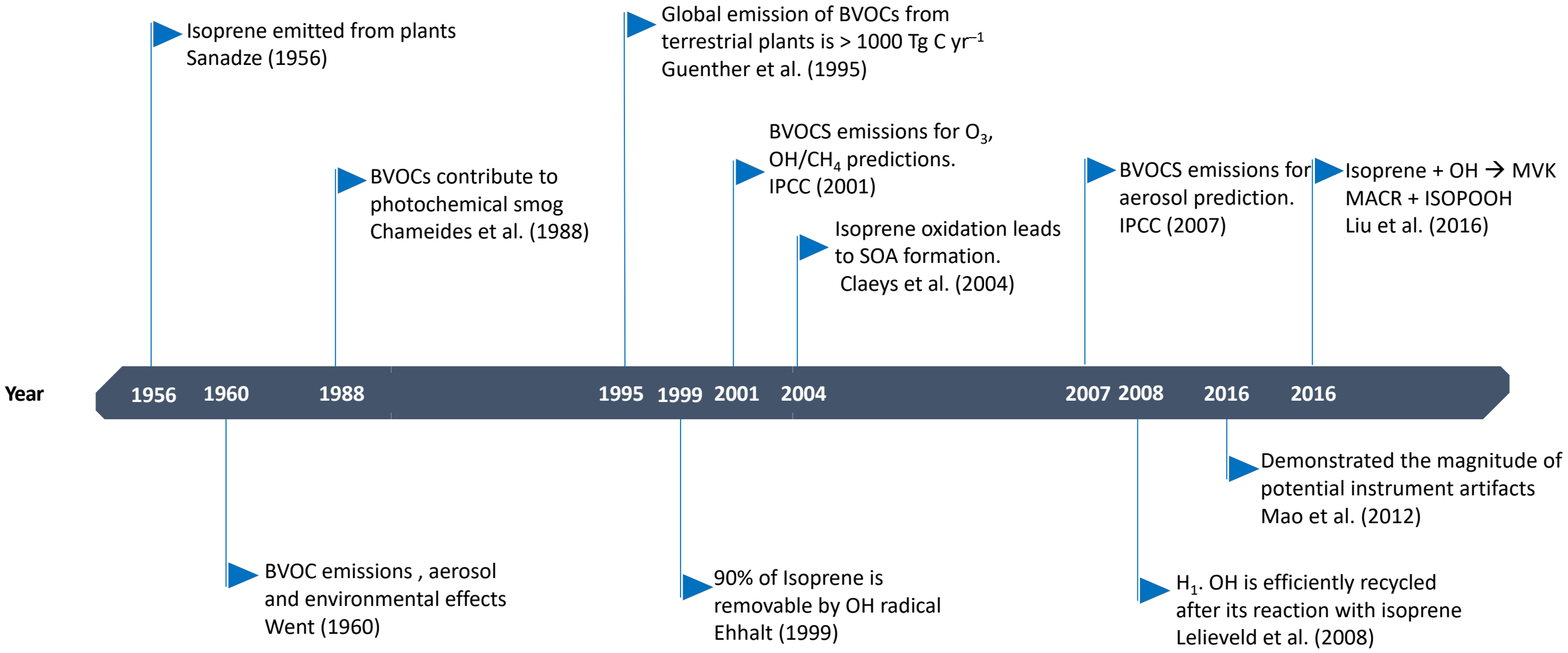
Supervisors: Dr. Karla Longo
Dr. Alex Guenther

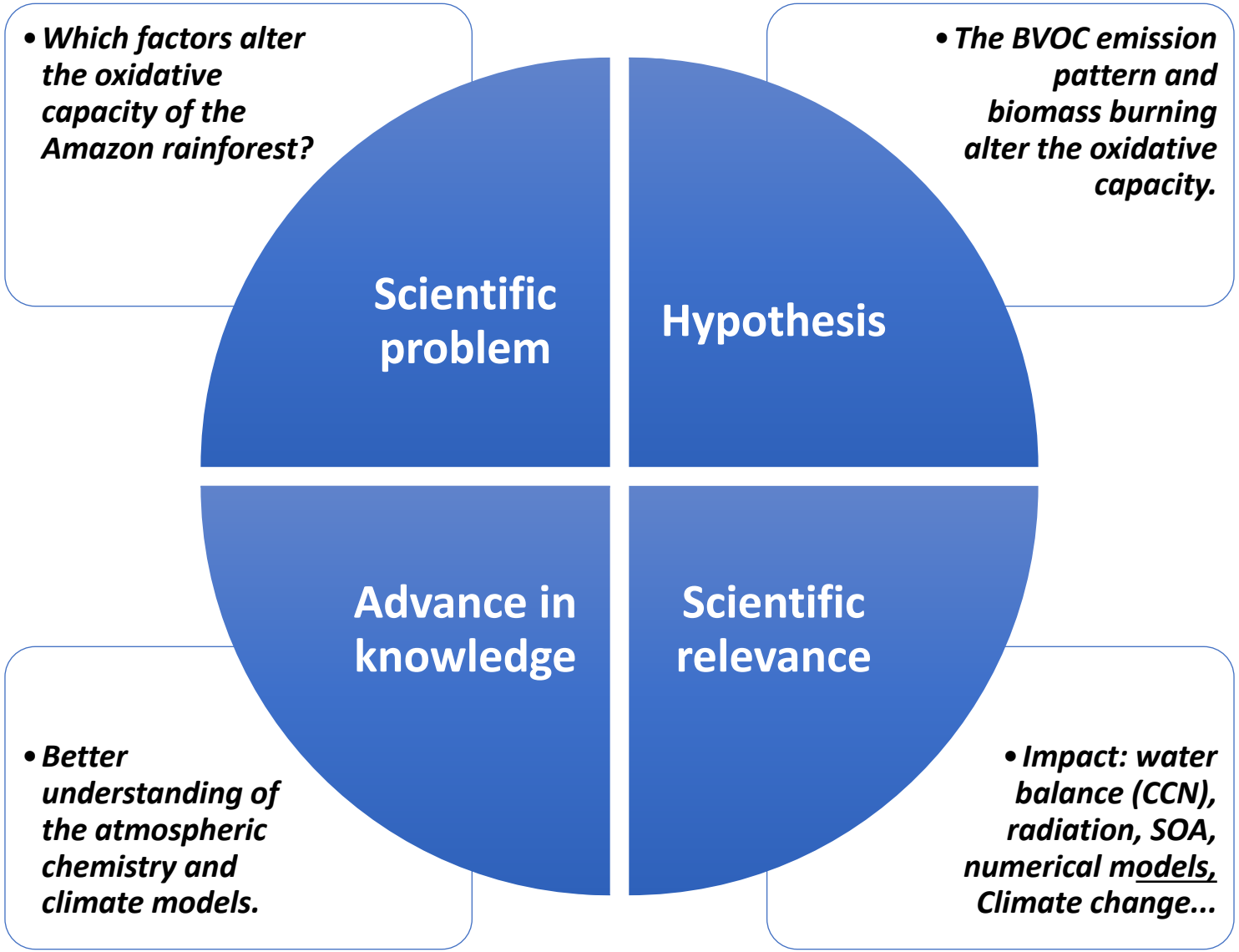
Ciência para sustentabilidade

Presentation Framework

- Introduction
- Objectives
- SAMBBA field campaign
- Modeling system
- Results
- Conclusions
- Future work

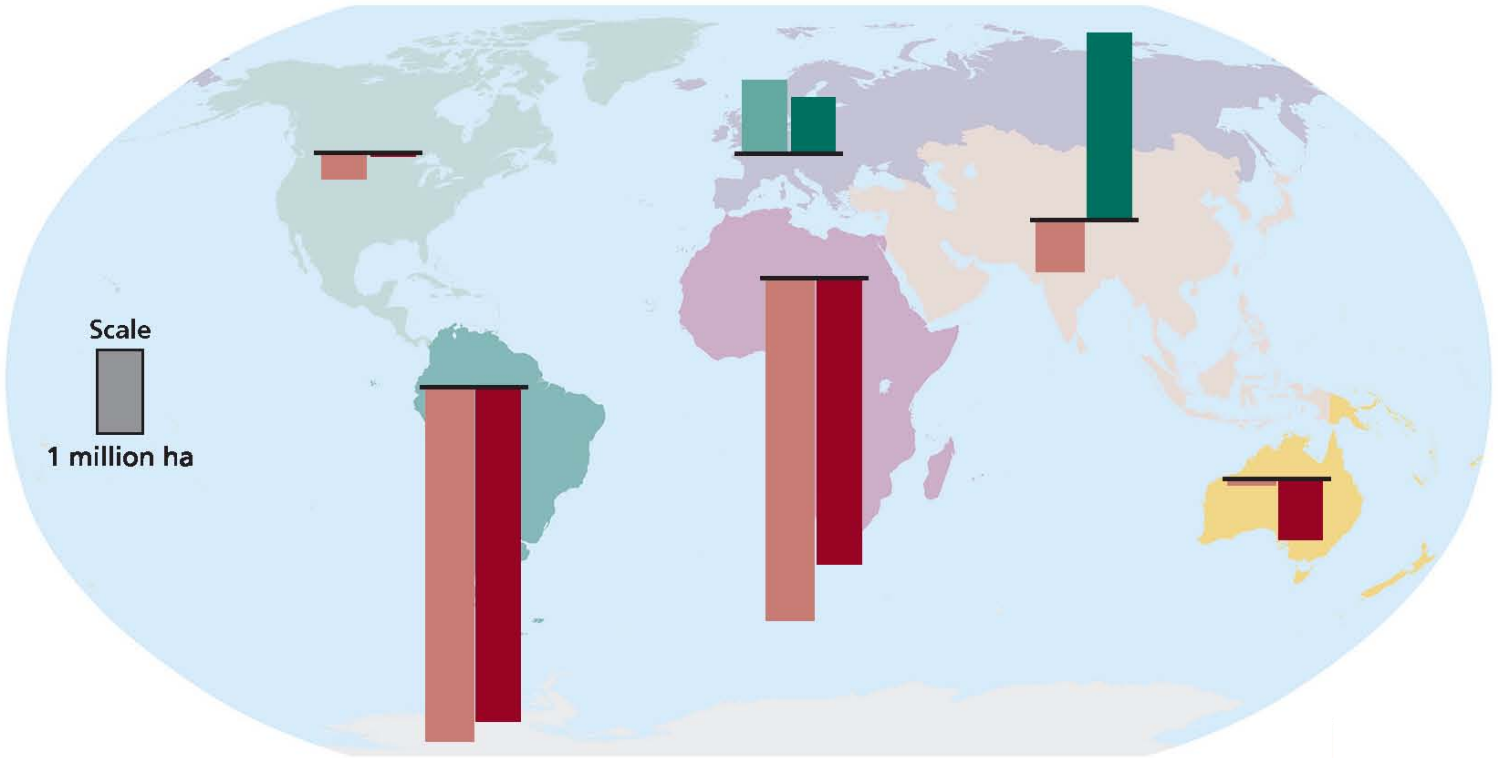
INTRODUCTION


Year
1956
1960
1988
1995
1999
2001
2004
2007
2008
2016
2016
Isoprene emitted from plants
Sanadze (1956)
BVOCs contribute to photochemical smog
Chameides et al. (1988)
Global emission of BVOCs from terrestrial plants is > 1000 Tg C yr−1
Guenther et al. (1995)
BVOCS emissions for O3, OH/CH4 predictions.
IPCC (2001)
Isoprene oxidation leads to SOA formation.
Claeys et al. (2004)
BVOCS emissions for aerosol prediction.
IPCC (2007)
Isoprene + OH → MVK MACR + ISOPOOH
Liu et al. (2016)
BVOC emissions , aerosol and environmental effects
Went (1960)
90% of Isoprene is removable by OH radical
Ehhalt (1999)
H1. OH is efficiently recycled after its reaction with isoprene
Lelieveld et al. (2008)
Demonstrated the magnitude of potential instrument artifacts
Mao et al. (2012)


• Which factors alter the oxidative capacity of the Amazon rainforest?
Scientific problem
• The BVOC emission pattern and biomass burning alter the oxidative capacity.
Hypothesis
Advance in knowledge
• Better understanding of the atmospheric chemistry and climate models.
Scientific relevance
• Impact: water balance (CCN), radiation, SOA, numerical models, Climate change...

Tropical Forest

Annual change in forest area by region, 1990-2010



Million ha/yr

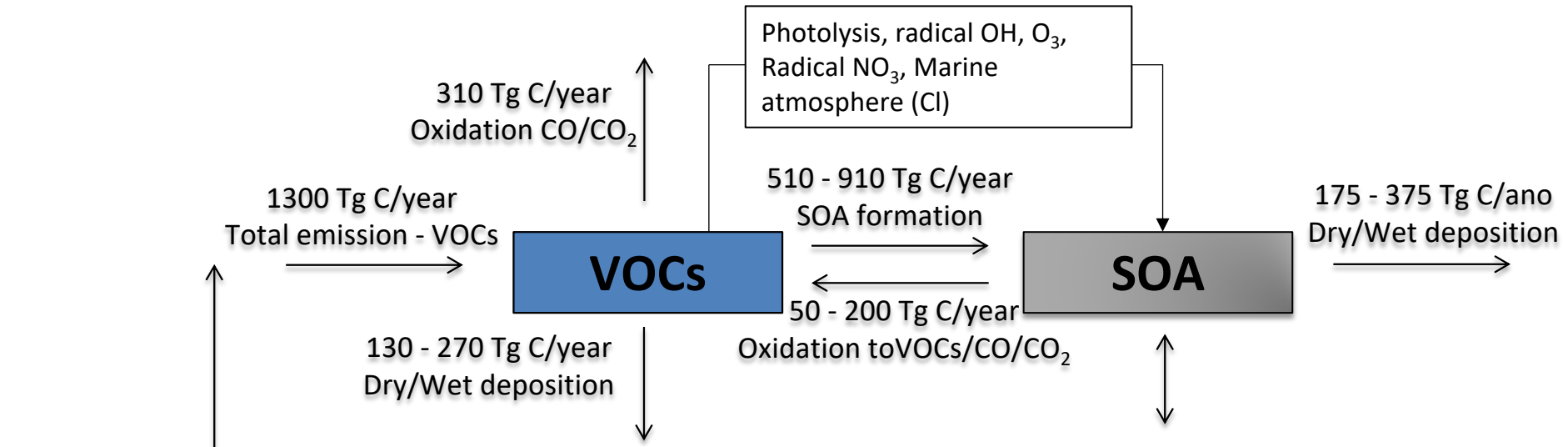
Characteristics of world's forests, 2010

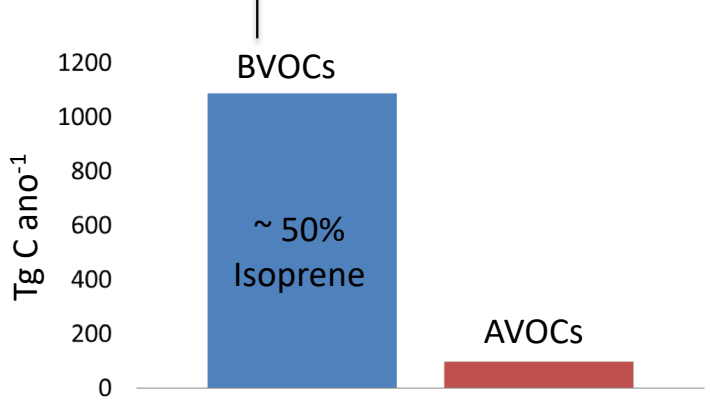
	Primary forest (%)
Africa	10
Asia	18
Europe	25
North and Central America	40
Oceania	18
South America	**75**
World	36

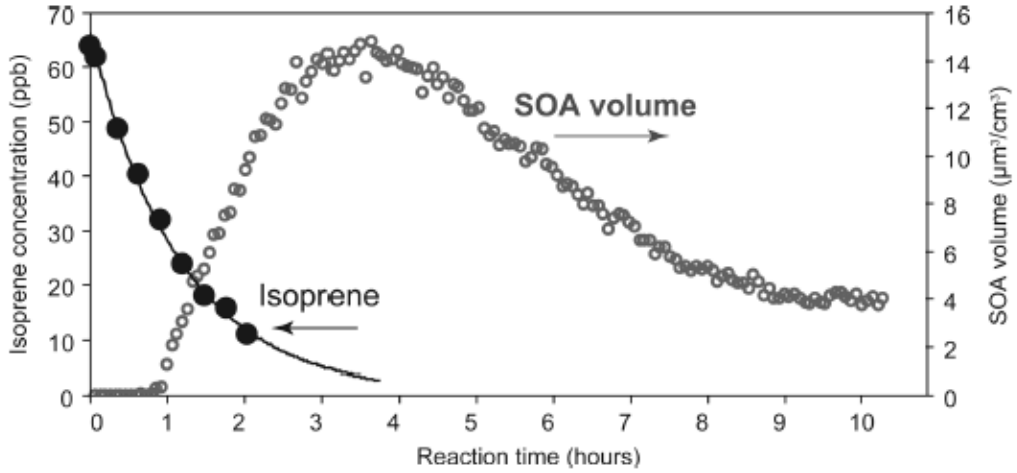
- Large area
- Primary forest → PFT
- Deforestation dynamic

http://neo.sci.gsfc.nasa.gov/

ESTIMATED GLOBAL VOC MASS BALANCE







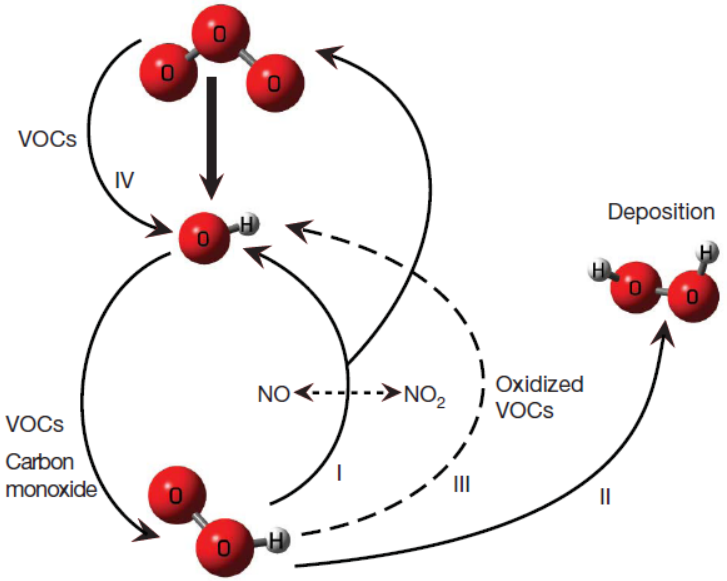
GOLDSTEIN; GALBALLY, 2007; SEINFELD et al., 2006; SCOTT et al., 2014 .

Oxidative capacity

Definition

Atmospheric global concentration OH (hydroxyl radical) → Oxidative capacity

1. Diurnal capacity to oxidize compounds
2. Tropical regions (radiation and humidity)



LELIEVELD *et al.*, 2008; GOLDSTEIN; GALBALLY, 2007

Oxidative capacity

- OH (hydroxyl radical) production
- $H_2O + O_3$ (*uv*)→ $2OH + O_2$

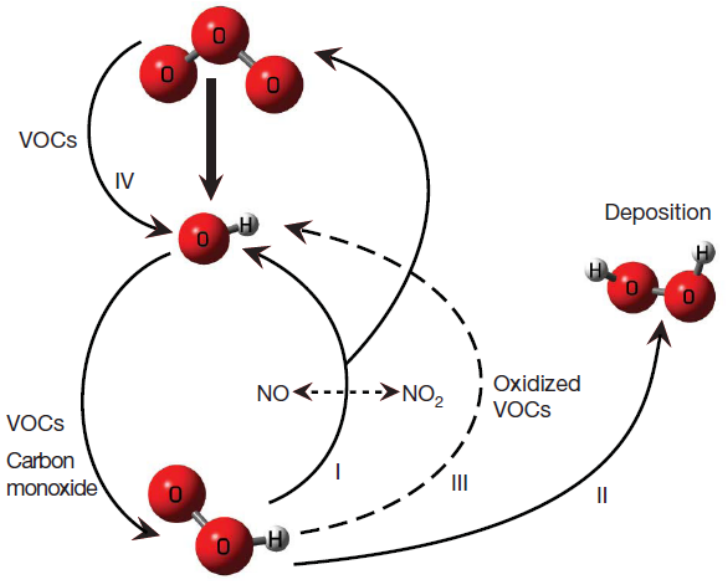
O_3

- OH reaction with VOCs, yelding OOH (peroxide radicals)

OH

- OOH reaction: steps I, II e III

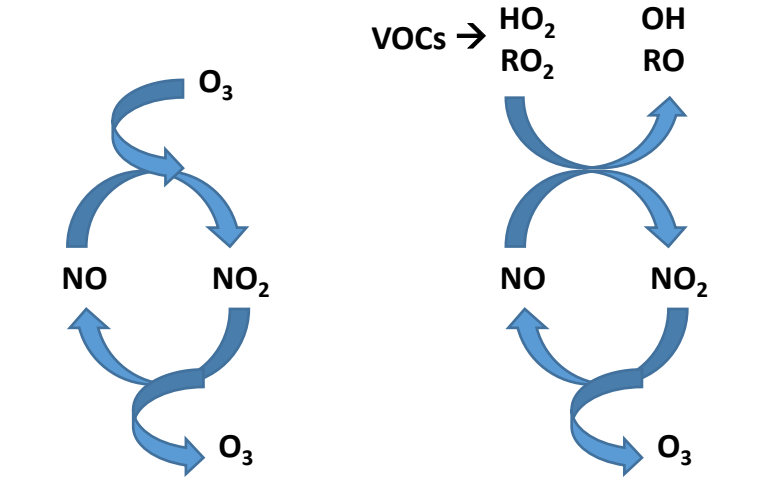
OOH



Step I

↑NO yelding OH directly and net production of O_3 (catalytic conversion NO ↔ NO_2)

$HO_2 + NO$ → $OH + NO_2$



LELIEVELD *et al.*, 2008

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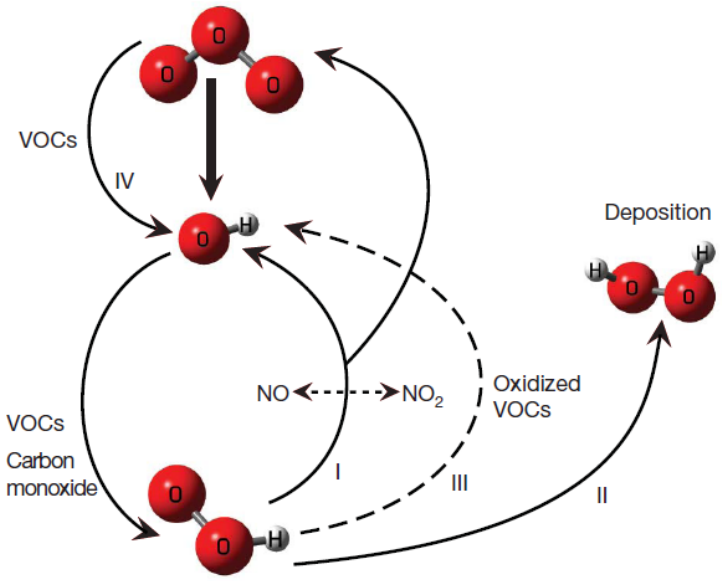

Step II

↓NO contributing to remove OH radicals (indirectly)

$HO_2 + RO_2$ → $ROOH + O_2$ (Deposition)

LELIEVELD *et al.*, 2008

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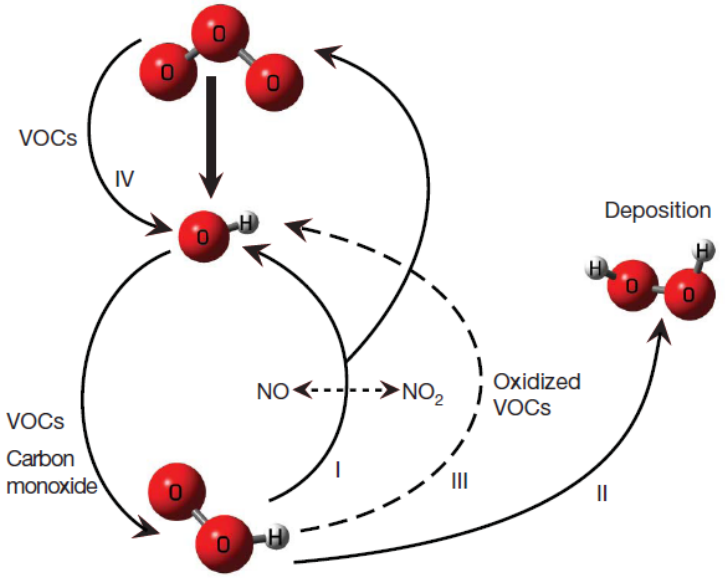


Step III

VOCs oxidized (isoprene) with ↓ NO can yeld/recycle OH radicals

Despite ↓NO, OH radical is present in the atmosphere at high levels

Isoprene + OH → R

$R + O_2 \rightarrow RO_2$

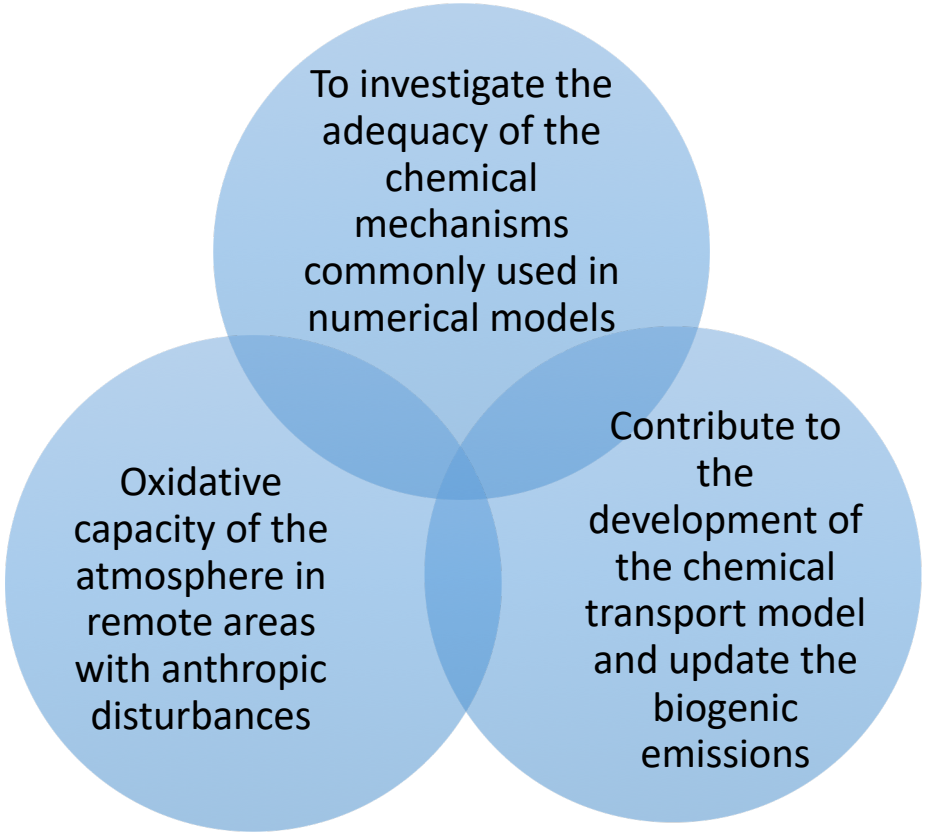
$RO_{2(\text{from isoprene})} + HO_2 \rightarrow OH +$ other product

LELIEVELD *et al.*, 2008; DILLON et al., 2006; ATKINSON et al., 2007; HASSON et al. (2004); JENKIN et al. (2007)

OBJECTIVES

Investigate the oxidative capacity of the atmosphere in the Amazon rainforest

(numerical modeling + direct observations)



SAMBBA FIELD CAMPAIGN

The South American Biomass Burning Analysis

- Sep 14 - Oct 3, 2012
- 67 hours flight
- Flights in burned out regions and remote areas
- Inside and outside boundary layer
- Aerosol Properties
- Gaseous chemistry (O_3, CO, NO/NO_2, CH_4, CO_2, PTRMS, GCMS, WAS bottles for GC analysis, tedlar bag samples)
- Basic meteorology, cloud and precipitation properties

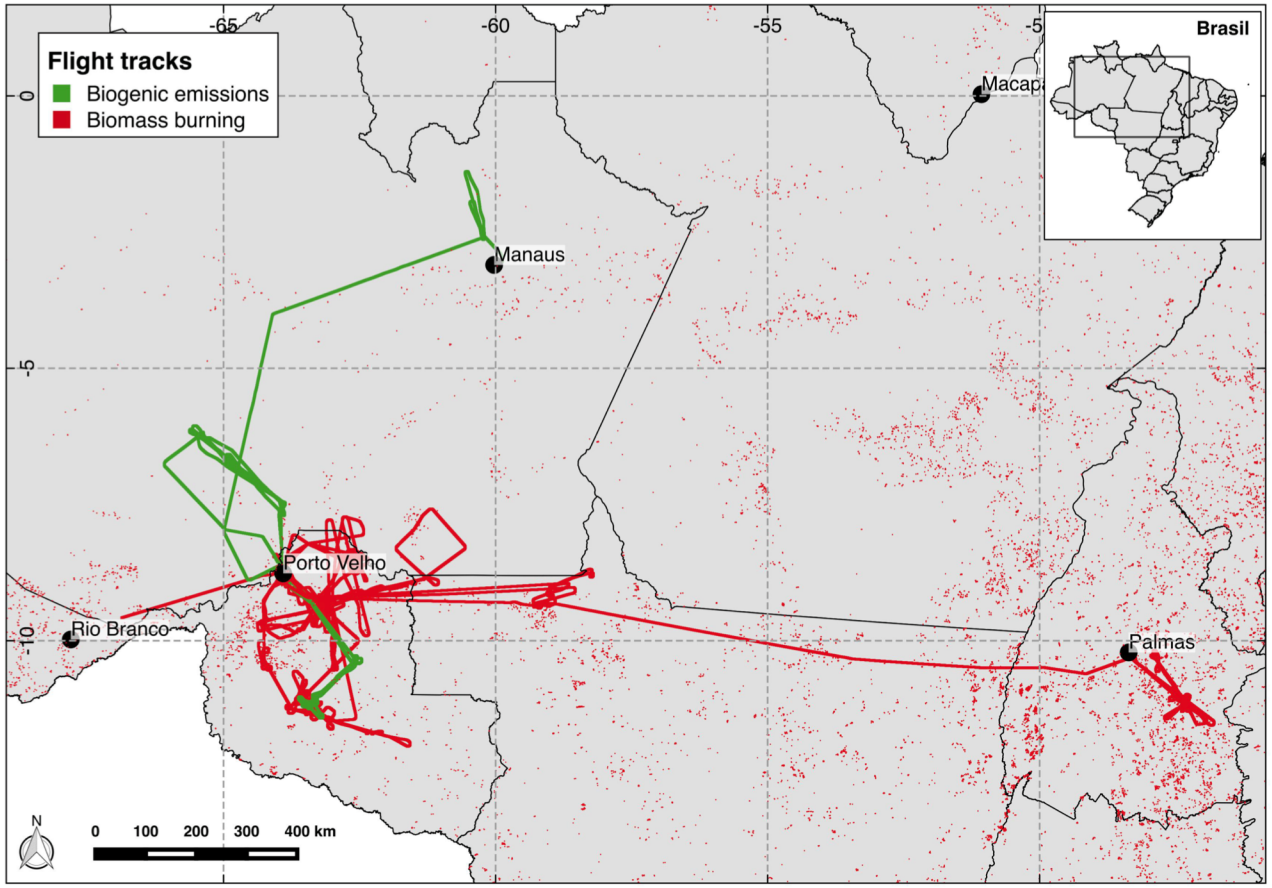
The BAe-146


The Core Console

Cloud Physics Probes

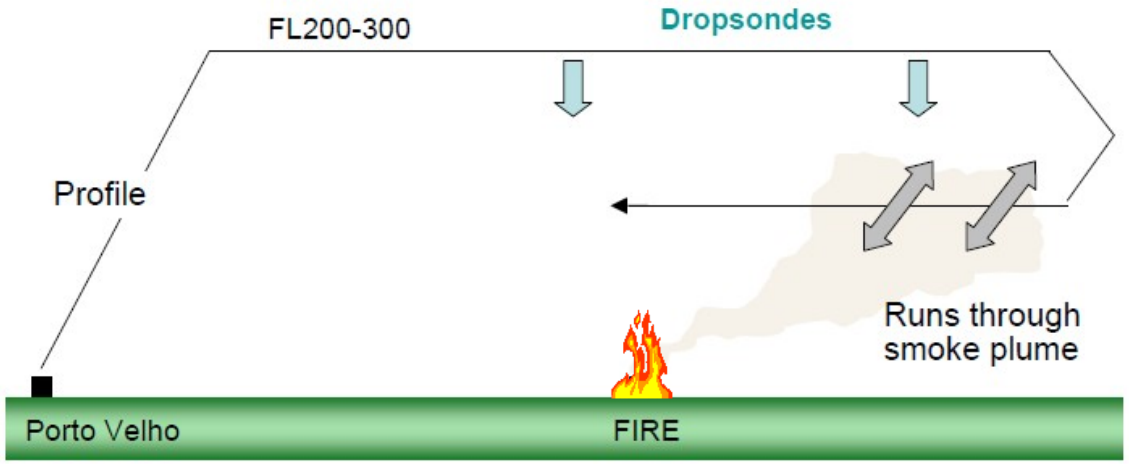

Aerosol Mass Spectrometry Rack

SAMBBA flights tracks according to their original goals as biogenic emissions (green) and biomass burning (red). The red points are depicting the fires detected by MODIS onboard AQUA satellite

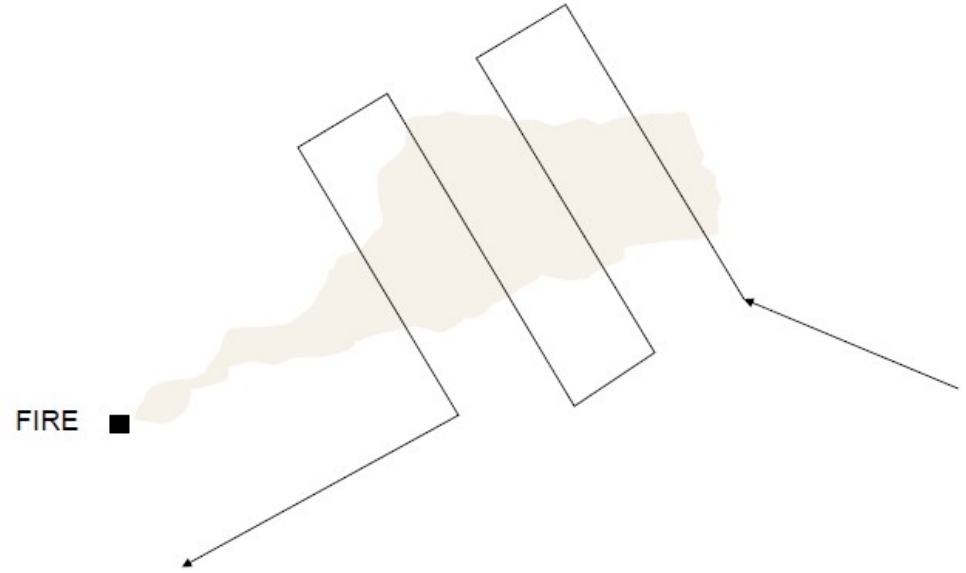


Scientific Cooperation between UK and Brazil

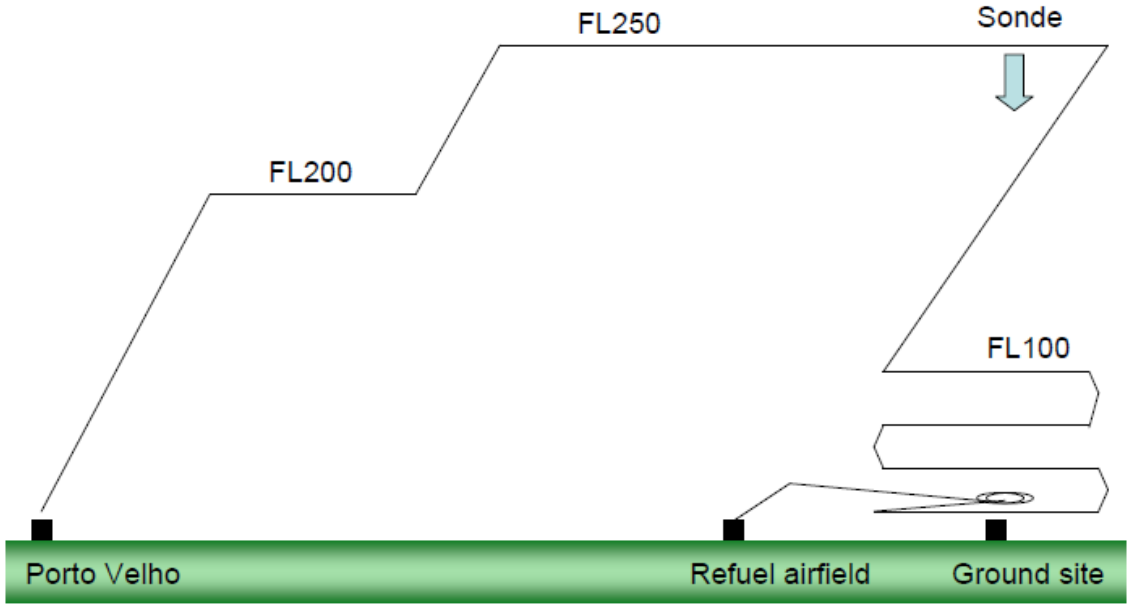
Met Office ⟷ INPE



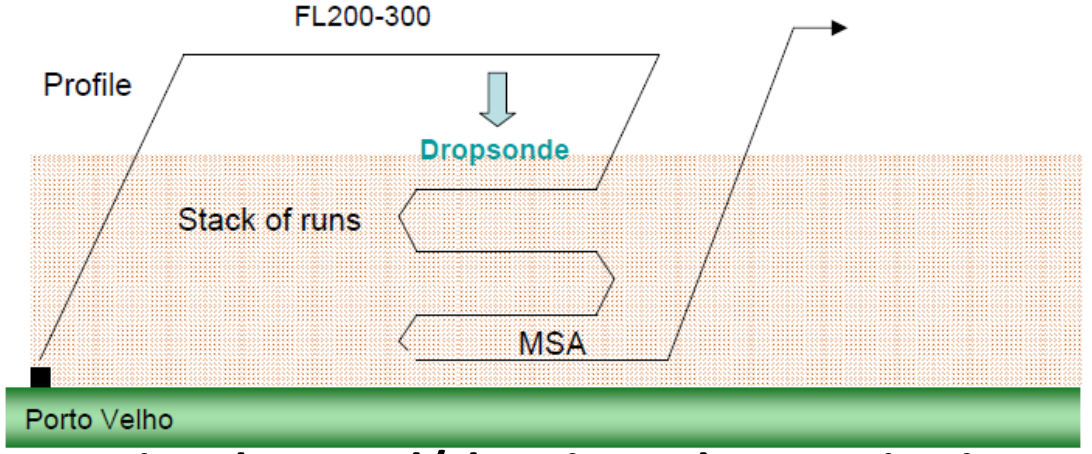
Smoke Plume Study



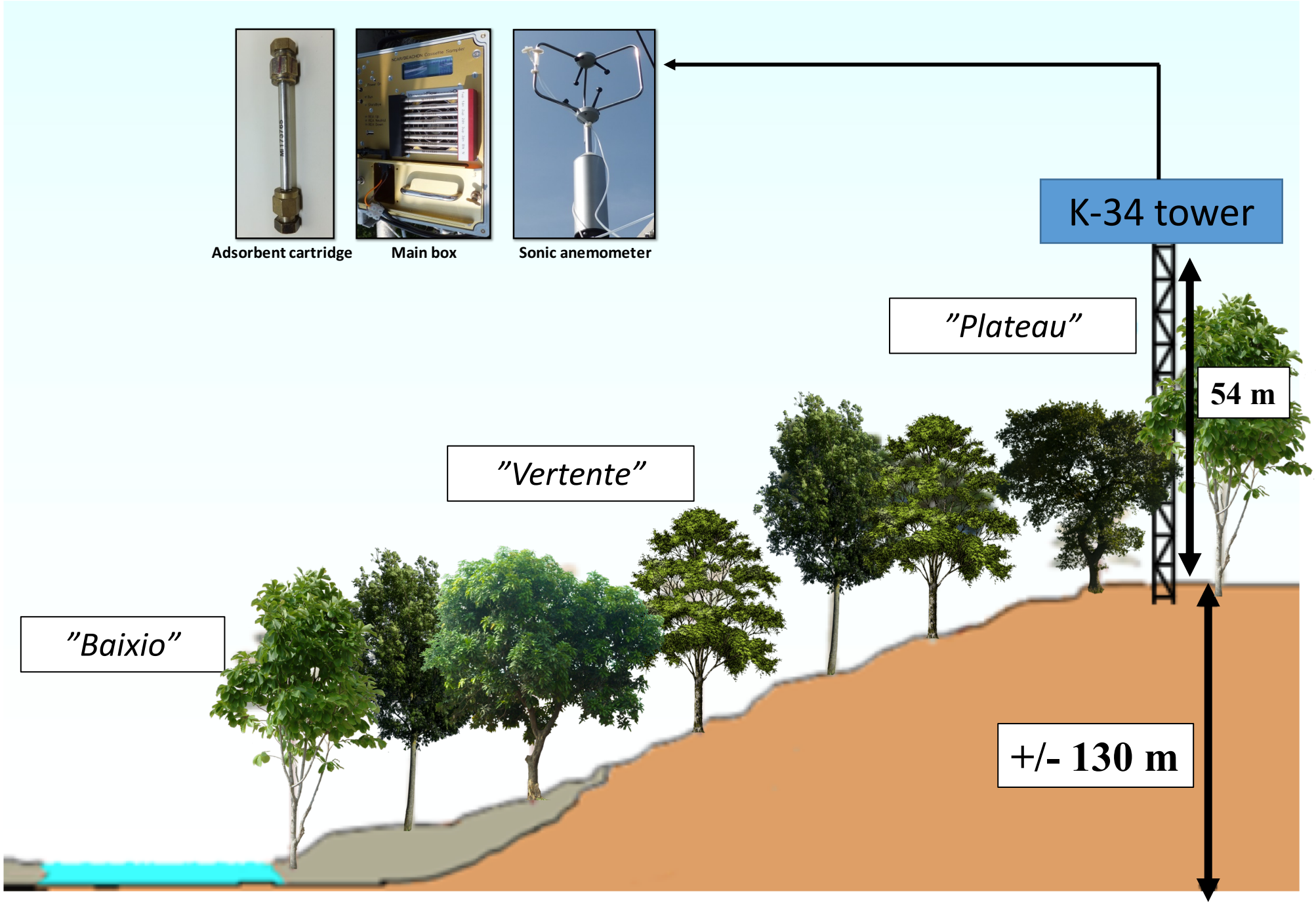
Overhead view of pattern through a smoke plume

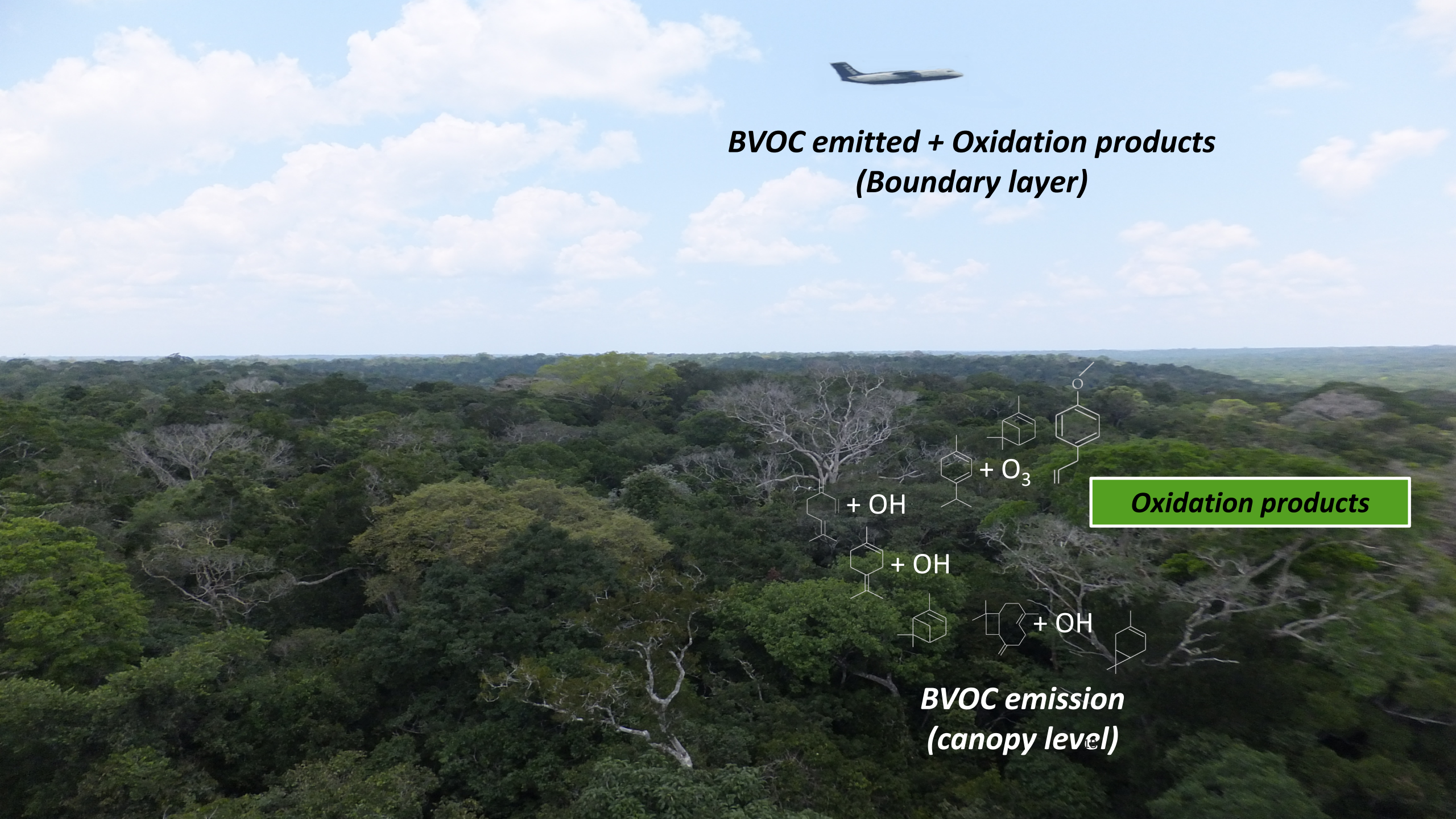


Radiative Closure Study



Regional aerosol/chemistry characterisation


Adsorbent cartridge
Main box
Sonic anemometer
K-34 tower
”Plateau”
54 m
”Vertente”
”Baixio”
+/- 130 m


BVOC emitted + Oxidation products
(Boundary layer)
+ O_3
+ OH
+ OH
+ OH
Oxidation products
BVOC emission
(canopy level)

MODELING SYSTEM

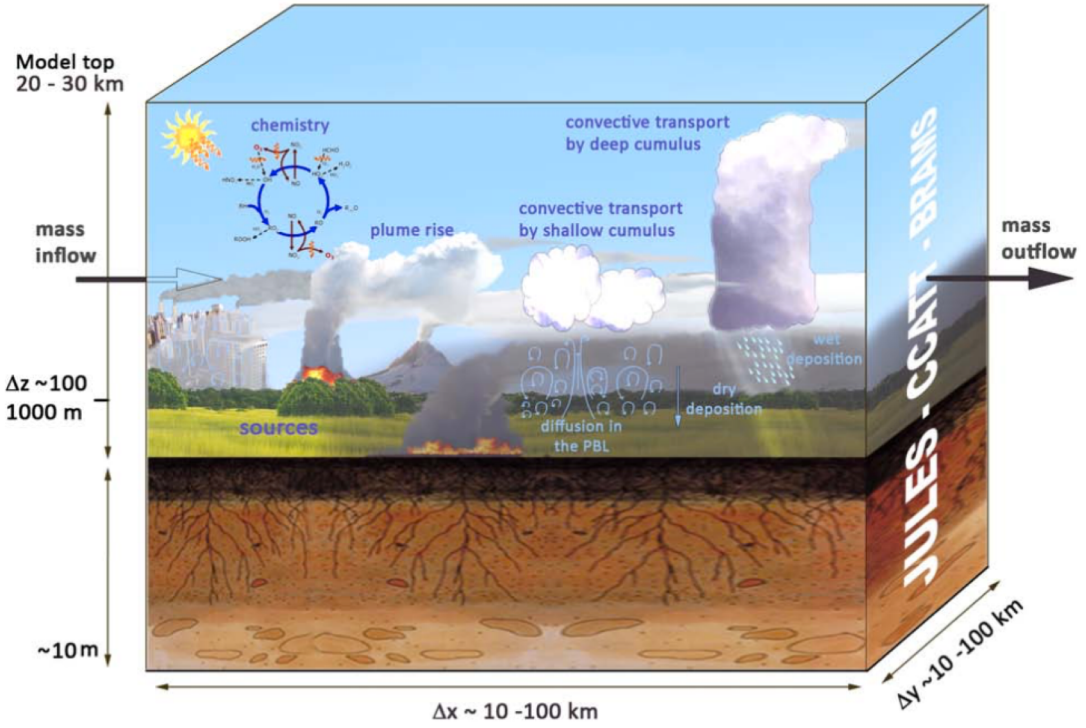
Brazilian developments on the Regional Atmospheric Modeling System – BRAMS v5.2

General mass continuity equation

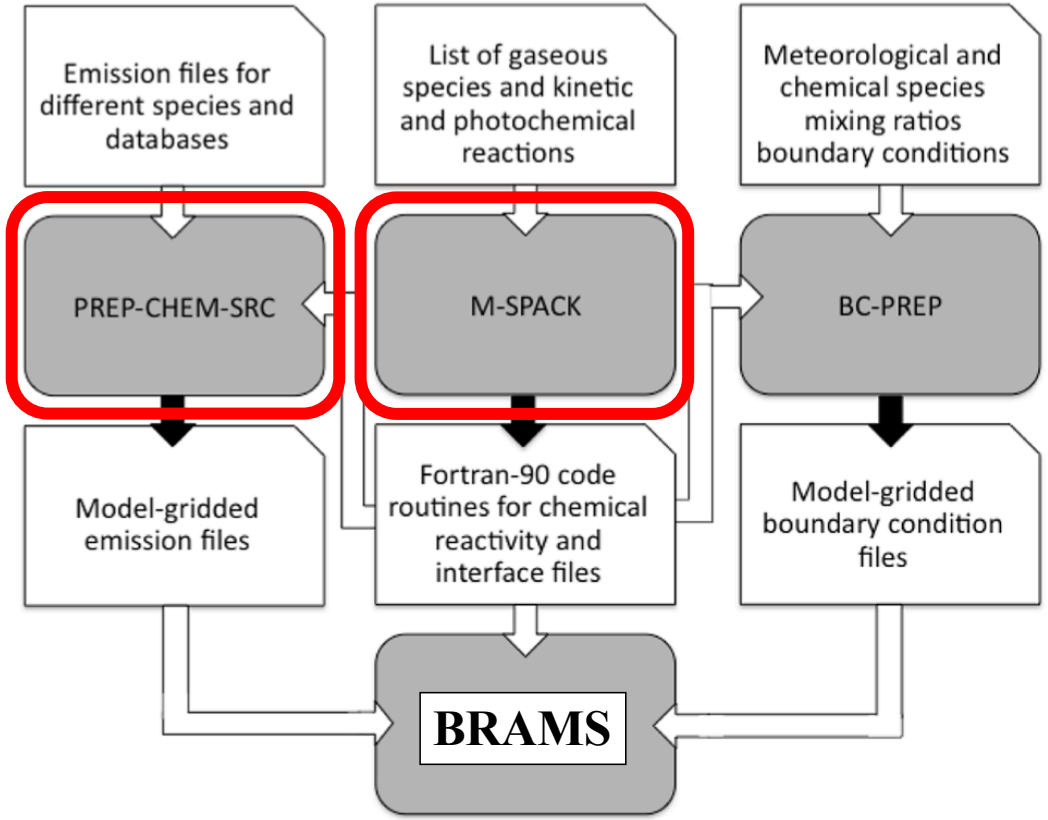
$$\frac{\partial \bar{s}}{\partial t} = \left(\frac{\partial \bar{s}}{\partial t}\right)_{adv} + \left(\frac{\partial \bar{s}}{\partial t}\right)_{\substack{PBL \\ turb}} + \left(\frac{\partial \bar{s}}{\partial t}\right)_{\substack{deep \\ conv}} + \left(\frac{\partial \bar{s}}{\partial t}\right)_{\substack{shallow \\ conv}} + W + R + Q_{\substack{plume \\ rise}} + \left(\frac{\partial \bar{s}}{\partial t}\right)_{\substack{chem \\ react}}$$

(1) (2) (3) (4) (5) (6) (7) (8) (9)

- **(1) grid box mean tracer mixing ratio**
- **(2) 3-D resolved transport (advection by the mean wind)**
- **(3) Turbulence in the PBL**
- **(4) Deep convection**
- **(5) Shallow convection**
- **(6) Wet deposition (aerosol)**
- **(7) Dry deposition (aerosol + gases)**
- **(8) Plume rise mechanism associated with vegetation fires**
- **(9) Chemical trasnformations (passive tracers lifetime)**



Source: Adapted from Moreira et al. (2013)



Source: Freitas et al. (2017)

Biogenic emissions

GEIA/ACCENT - *Global Emissions Inventory Activity of the Atmospheric Composition Change: the European Network*

MEGAN - *Model of Emissions of Gases and Aerosols from Nature*, (GUENTHER *et al.*, 2012).

Climatology data (for 2000)
Monthly average for emissions
Resolution 0.5° x 0.5°

Chemistry mechanism

RACM (*Regional Atmospheric Chemistry Mechanism* - STOCKWELL *et al.*, 1997): 77 species
- more complete than Carbon Bond and RELACS
- based on the lumped molecule approach (species grouped according to their chemical nature)
- originally designed to simulate troposphere chemistry from urban to remote conditions
- 17 stable inorganic species, 4 inorganic intermediates, 32 stable organic species and 24 organic intermediates
- total of 237 reactions, including 23 photolytic reactions

CB05 (*Carbon Bond* - YARWOOD *et al.*, 2005): 36 species

RELACS (*Regional Lumped Atmospheric Chemical Scheme* - CRASSIER *et al.*, 2000): 37 species

Model of Emissions of Gases and Aerosols from Nature - MEGAN

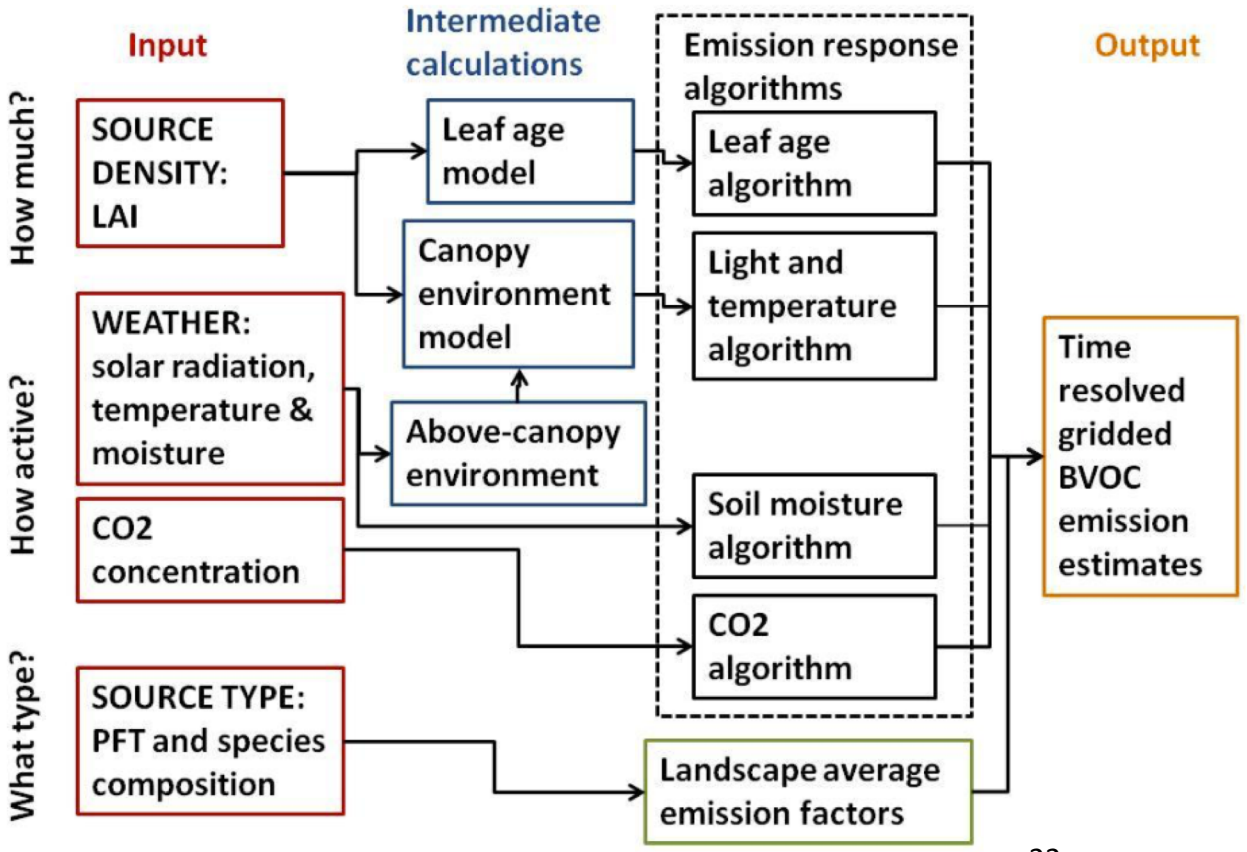
$$Fi = \gamma i \sum Ei,j \ \chi j$$

F_i (µg m^{-2} h^{-1}) → **estimates emissions of chemical species** *i* from terrestrial landscapes.

γ_i emission **activity factor** accounts for the processes controlling emission **responses to environmental and phenological conditions**.

$E_{i,j}$ is the **emission factor at standard conditions** for vegetation type *j* with fractional grid box areal coverage ***Xj***.

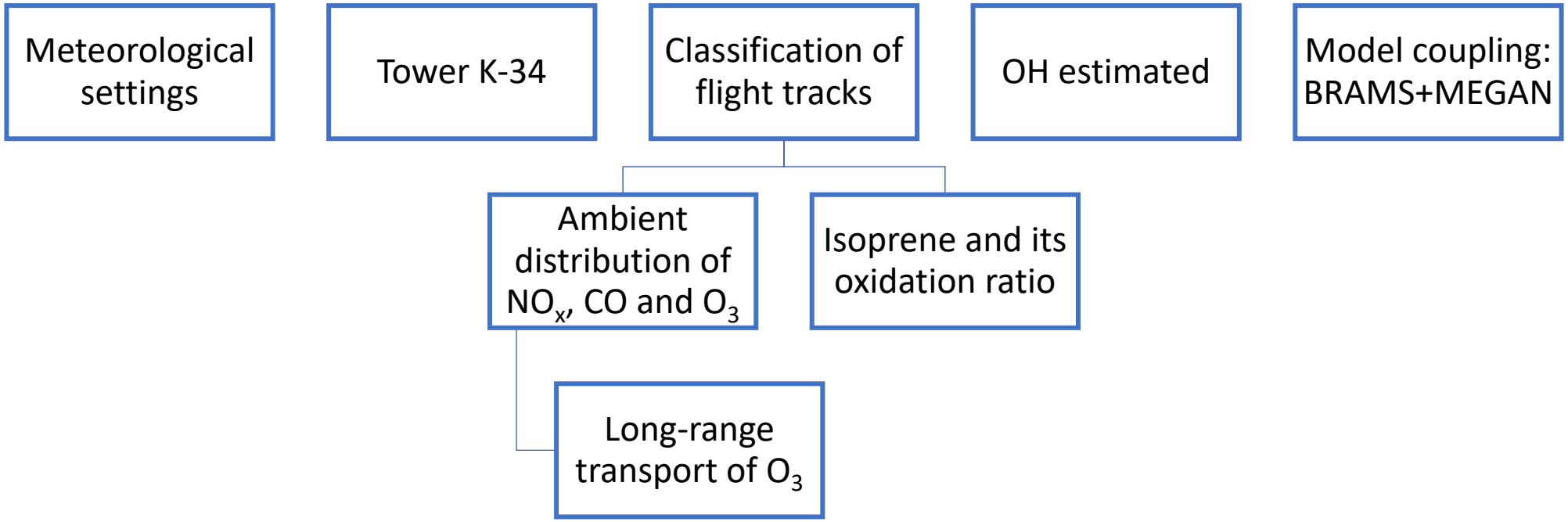
- Spatial Resolution of about 1 km^2
- Hourly emissions of 19 categories of chemical compounds representing 147.
- Compatible with other chemical mechanisms (RACM for example…)



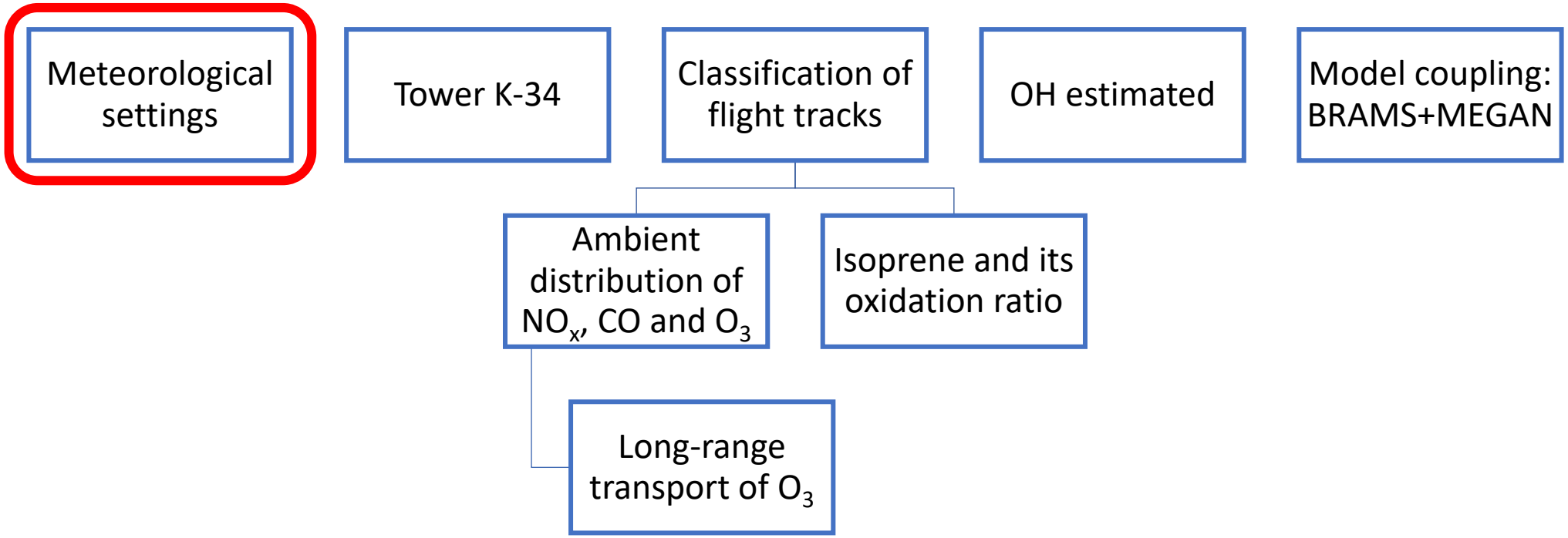
Source: Guenther et al. (2012)

RESULTS

Results


Meteorological settings
Tower K-34
Classification of flight tracks
OH estimated
Model coupling: BRAMS+MEGAN
Ambient distribution of NO_x, CO and O_3
Isoprene and its oxidation ratio
Long-range transport of O_3

Results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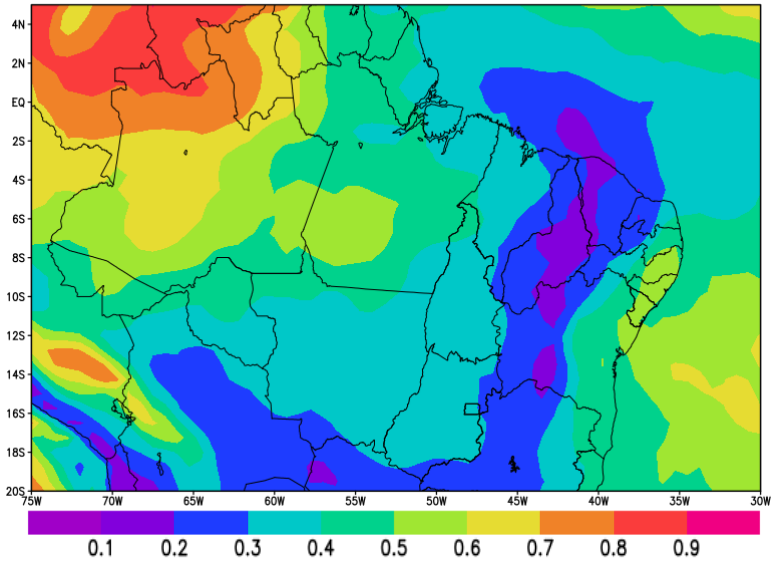
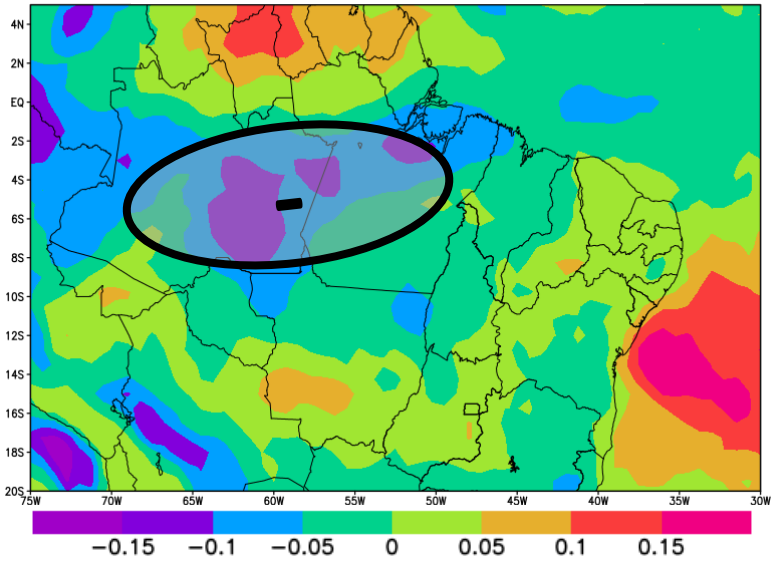
Meteorological settings

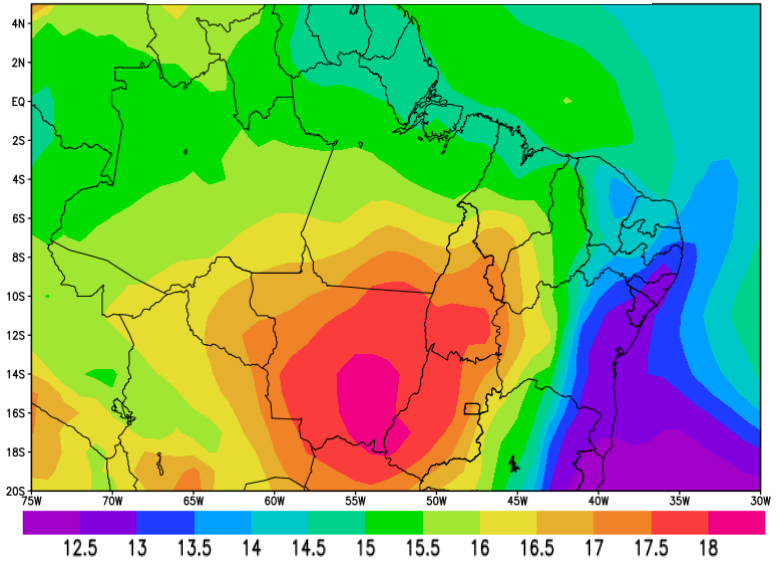
Total Cloud Cover - September



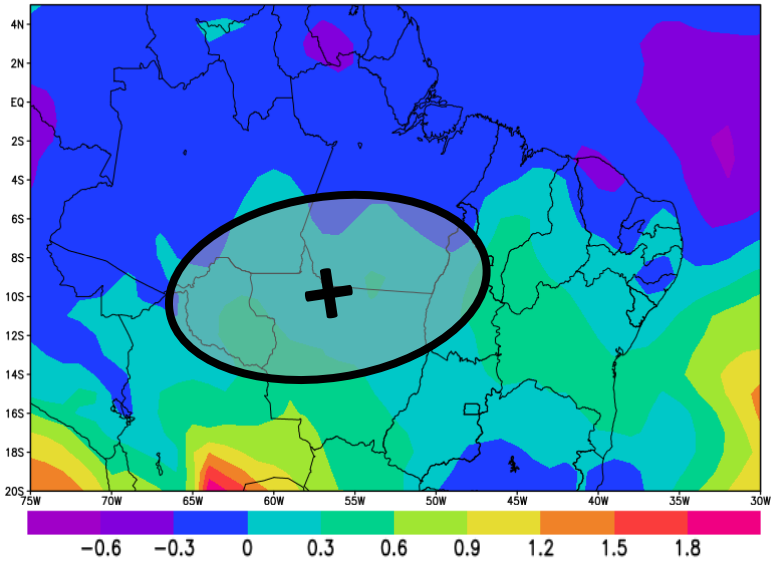
Total Cloud Cover – Anomaly



Temperature [C°] - September



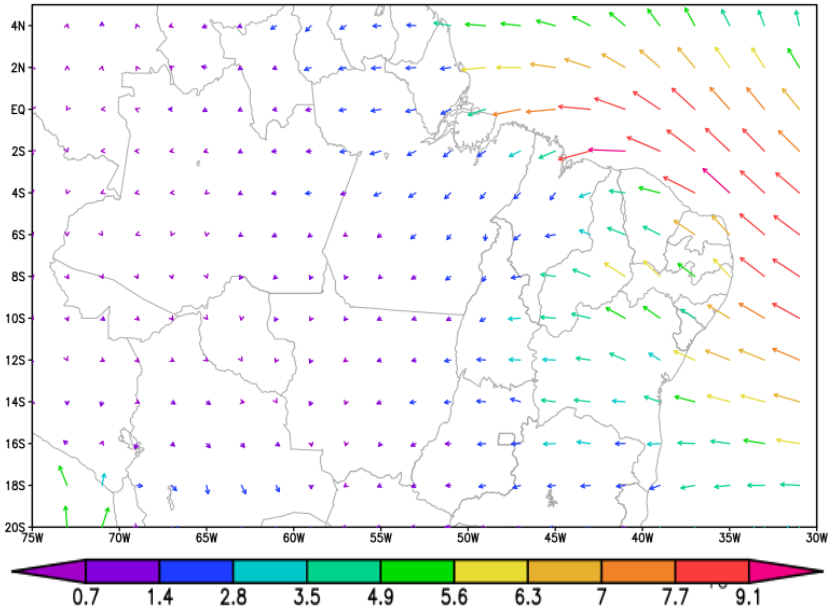
Temperature [C°] - Anomaly



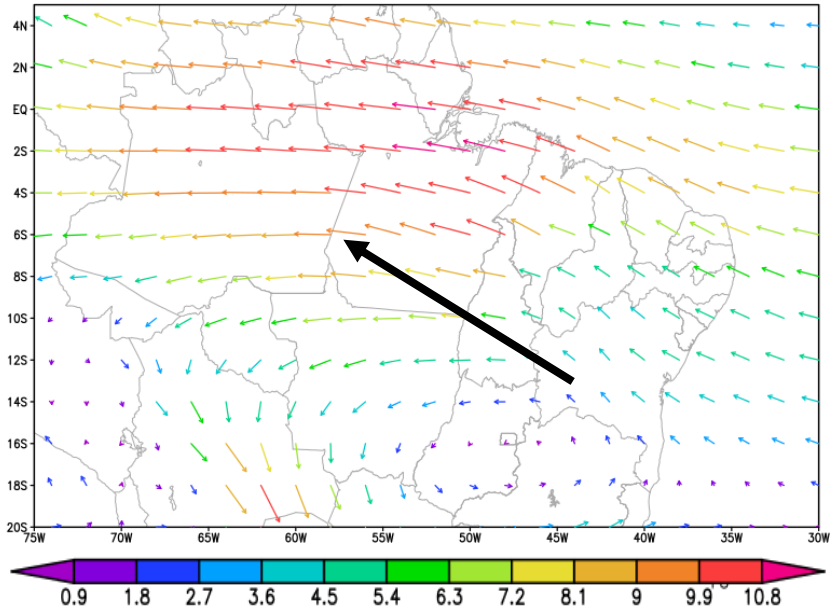
Negative anomalies of total cloud cover were intense mostly in the western and central Amazonia, while in the eastern sector the reduction in the total cloud cover was less intense.

The temperature presented a **positive anomaly in the southeast part of the Amazon forest**, with values achieving up to 18°C, in contrast, negative anomalies were found in the north/northwest part of the forest.

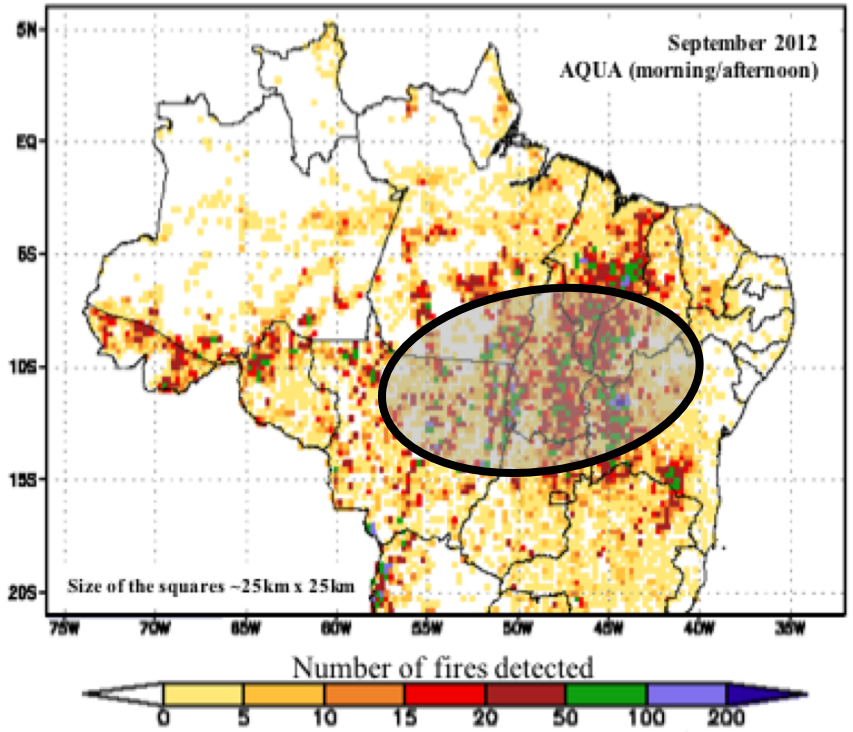
Wind (m/s) – September 10m

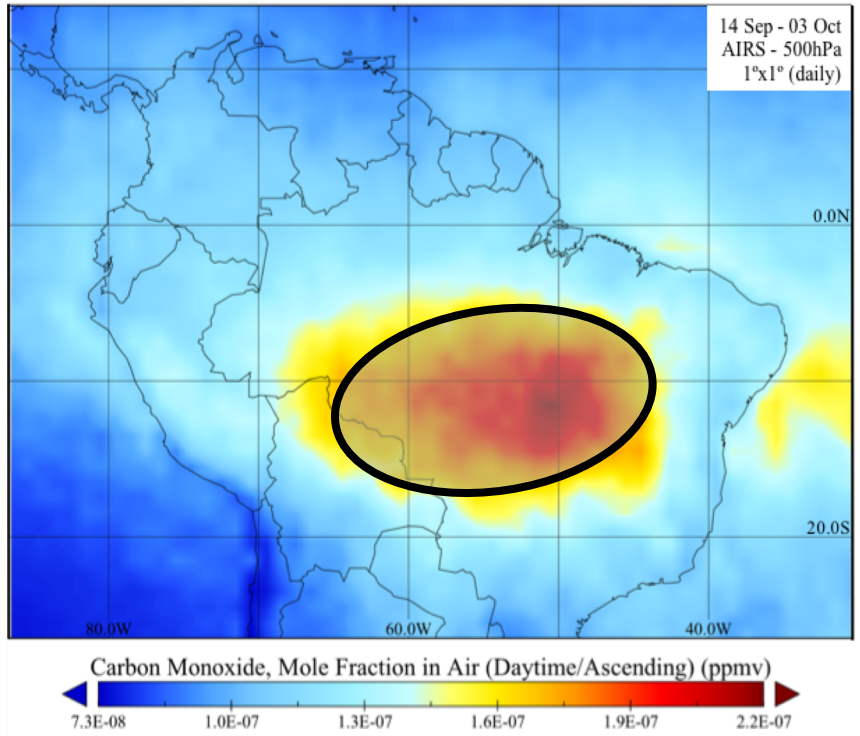


Wind (m/s) – September 700 mb



- Through the air masses transport in high altitudes, the **recirculation of the air from eastern to western part** of Amazon rainforest can influence in the atmospheric composition of the northwest part, previously defined as a potential clean area.



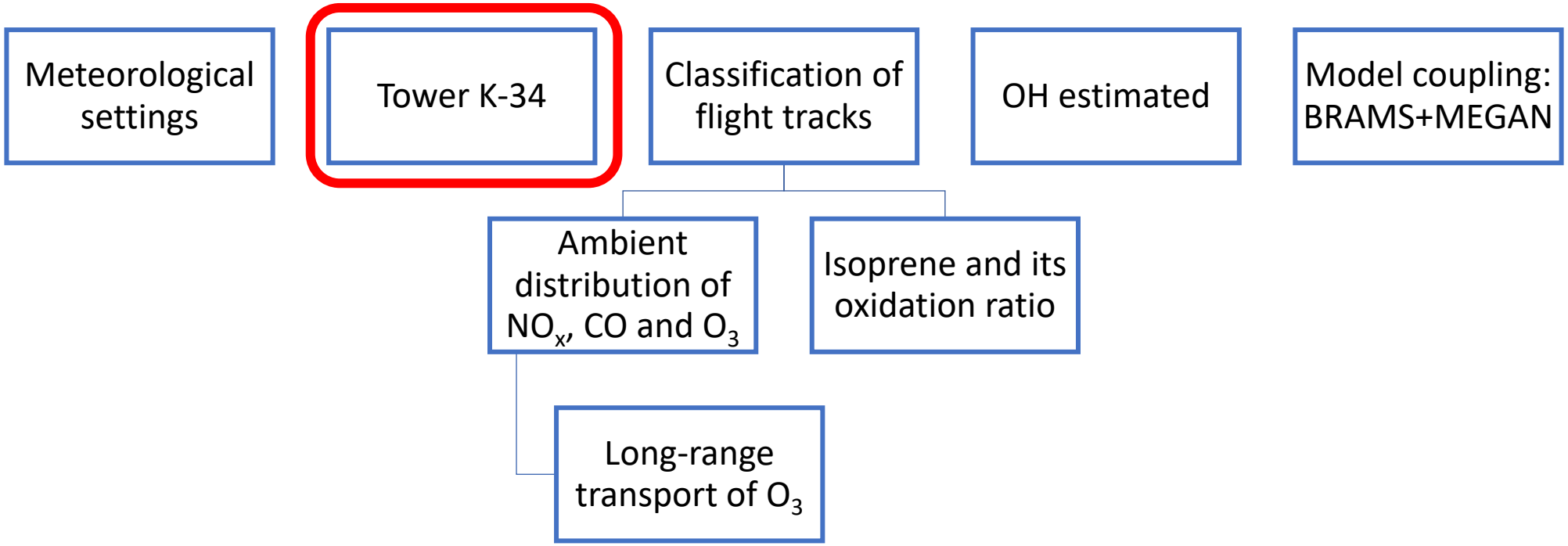


- **Same pattern for number of fires and CO mixing ratio.**
- Intense and persistent fire activity occurring in the eastern part.

  CO: ~200 – 220 ppbv
- The **northwest sector was representative of a clean area**.

  CO: ~130 ppbv
- Most SAMBBA flights with a biogenic emission objective were carried out in the northwest part, with some of them further south of Amazon rainforest.

Results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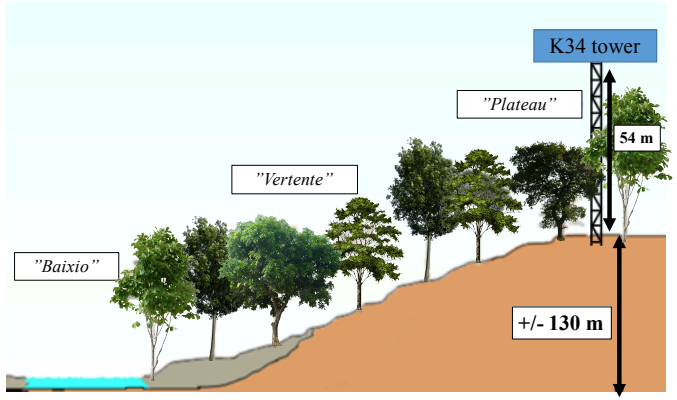
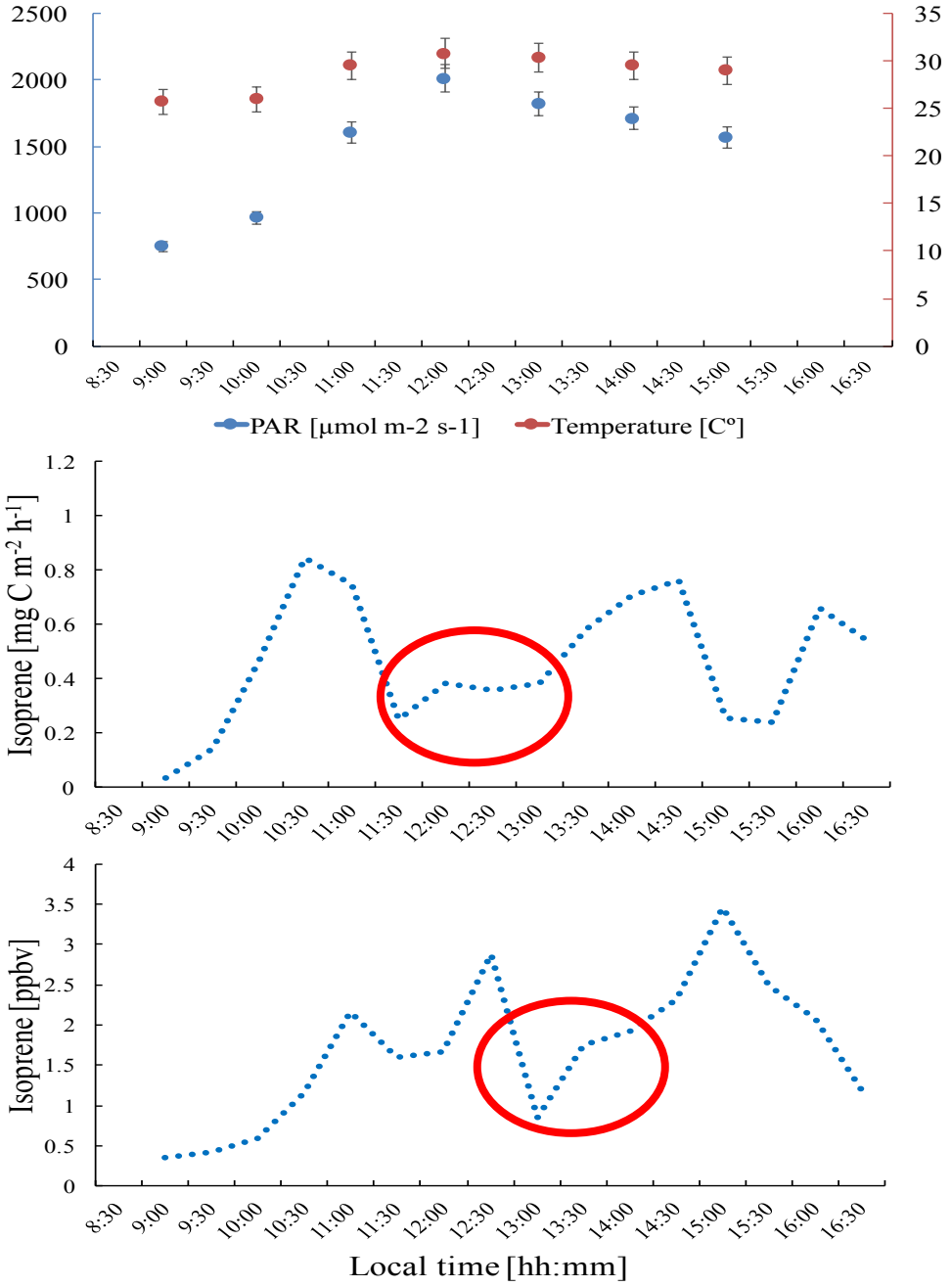
Tower K-34

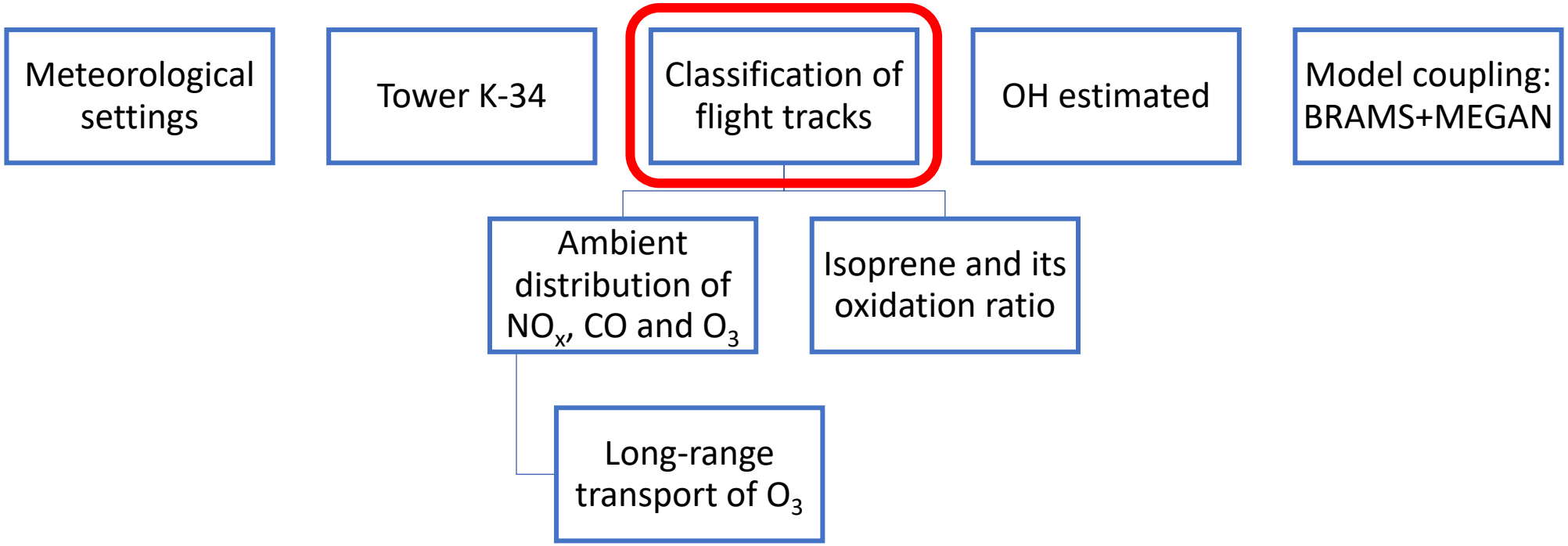
DATE	Nº of experiment	Nº of cycles	Hour (local time)
19 Sep 2012	1	3 (30 min./each)	12:30 13:30 ←
20 Sep	1	3 (30 min./each)	09:00 10:30
2012*	2	3 (30 min./each)	11:00 12:30
	3	3 (30 min./each)	13:00 14:30
	4	3 (30 min./each)	15:00 16:30
21 Sep 2012	1	3 (30 min./each)	08:00 09:30 ←
	2	3 (30 min./each)	10:00 11:30 ←



- PAR (Photosynthetically active radiation) and temperature correlated with Isoprene during the day.
- Some gaps associated with a reduced number of sampling conducted in 19 and 21 September



Results


Meteorological settings
Tower K-34
Classification of flight tracks
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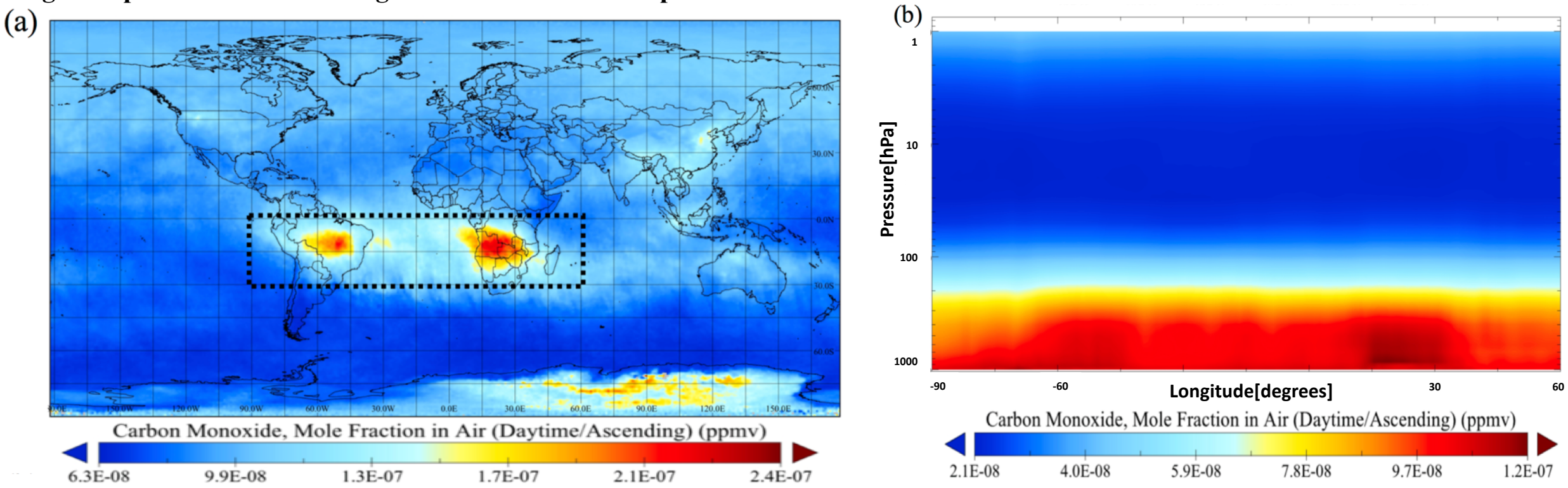
Isoprene and its oxidation ratio
Long-range transport of O_3

CLASSIFICATION METHOD OF FLIGHT TRACKS

Time averaged CO (ppmv) from AIRS onboard AQUA satellite during daytime: (a) global map at 500 hPa, and (b) cross section of longitude-pressure within the region indicated on the map.



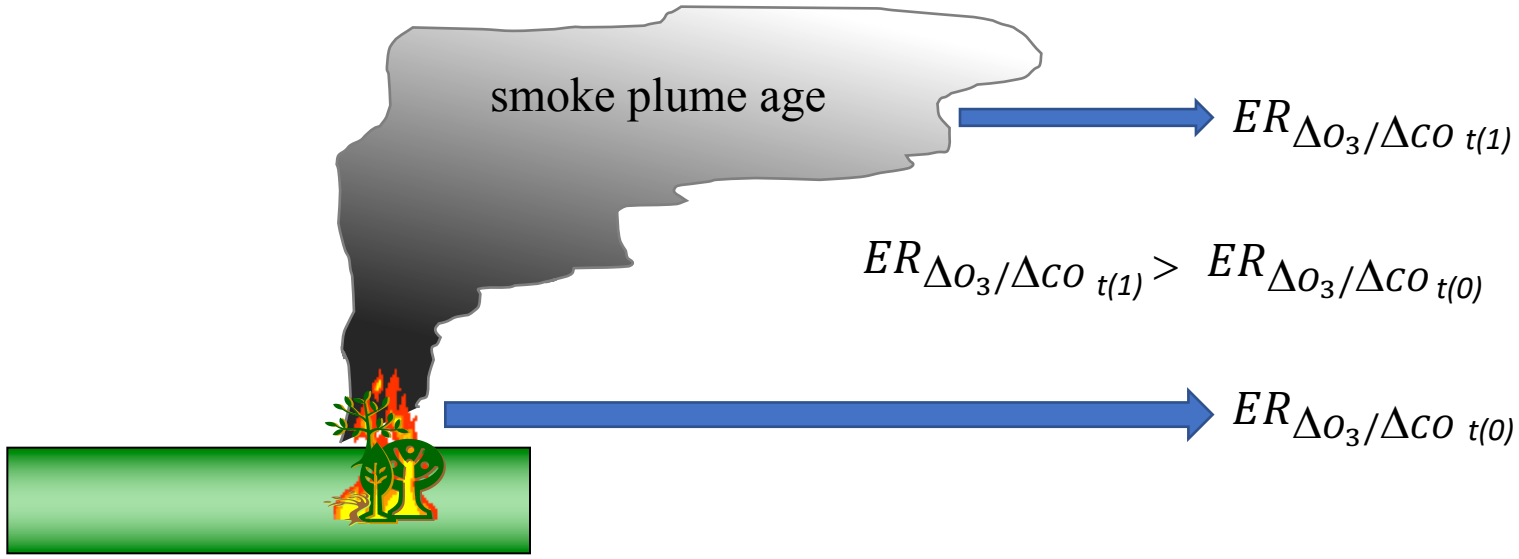
Andreae et al. (2012) and several references therein:
$[CO]_{background}$ ~100 ppbv.

CO inflow from Africa into the Amazon basin:
$[CO]_{background}$ 140 - 160 ppbv.

CLASSIFICATION METHOD OF FLIGHT TRACKS

As **O_3 is formed photochemically downwind** during smoke aging, the enhancement ratio of O_3 to CO is acceptable as a reliable indicator of the smoke plume age (PARRISH et al., 1993; ANDREAE et al., 1994).

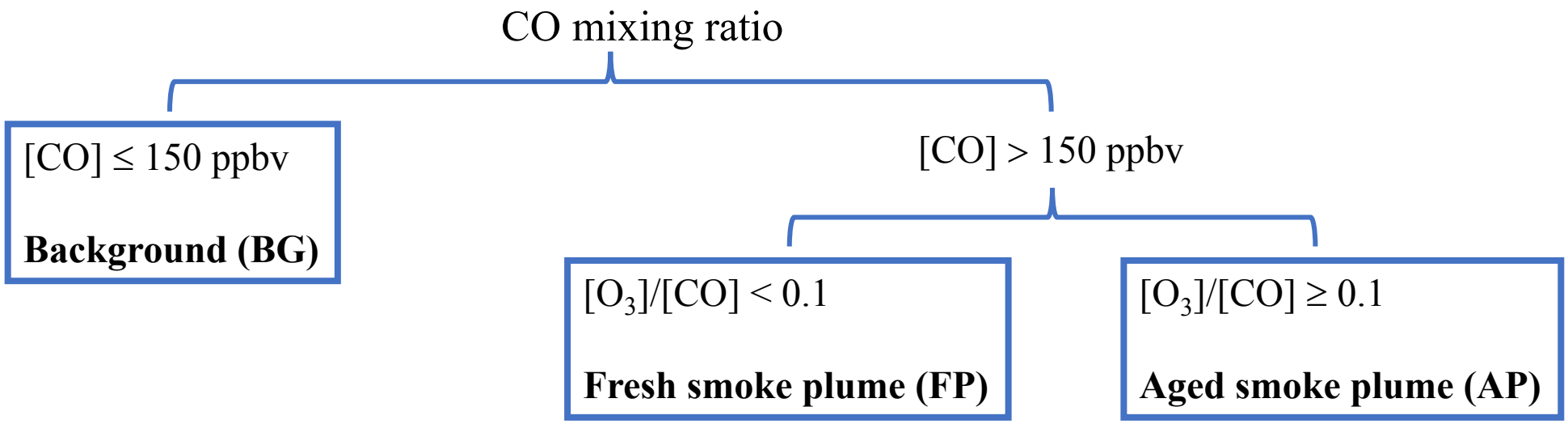
$$ER_{\Delta O_3/\Delta CO} = \frac{[O_3]_{smoke} - [O_3]_{background}}{[CO]_{smoke} - [CO]_{background}}$$



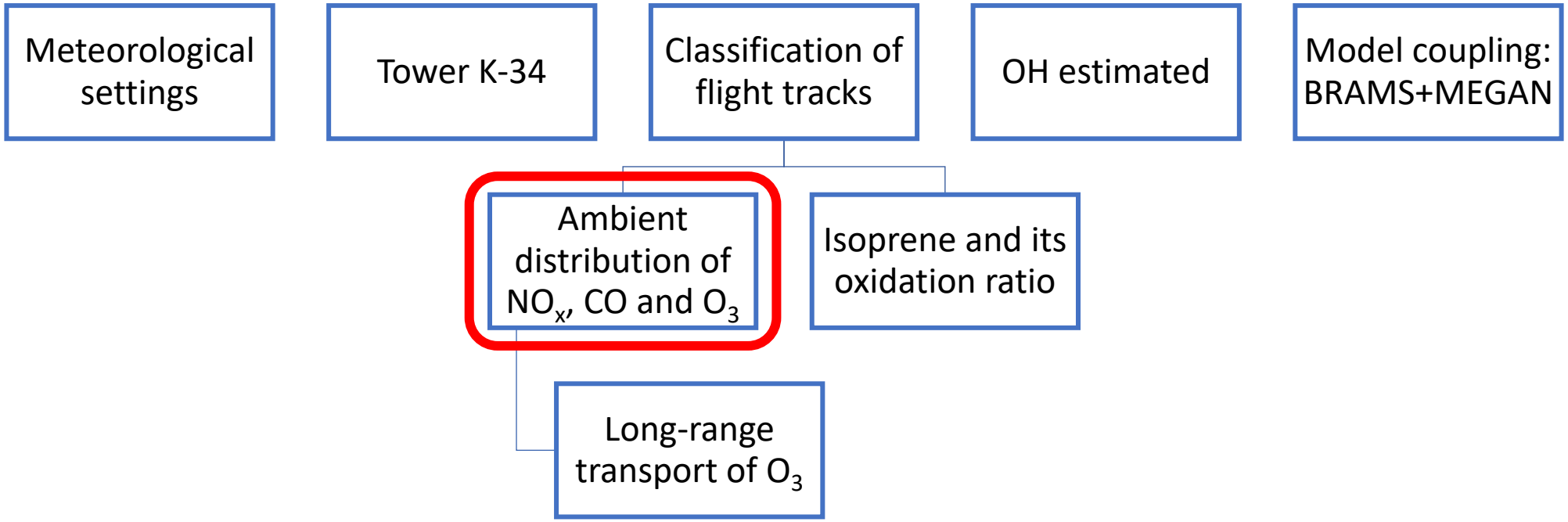
From literature:

$ER_{\Delta O_3/\Delta CO}$ = 0.1 (approx. 2 hours)

CLASSIFICATION METHOD OF FLIGHT TRACKS



Results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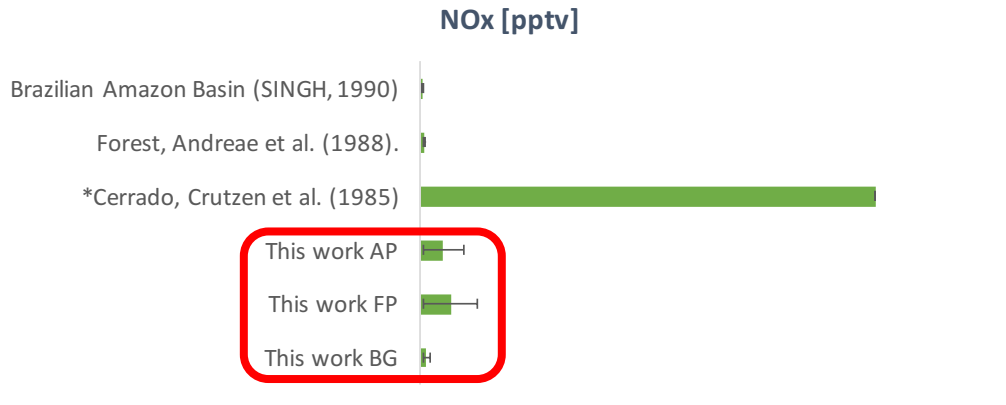
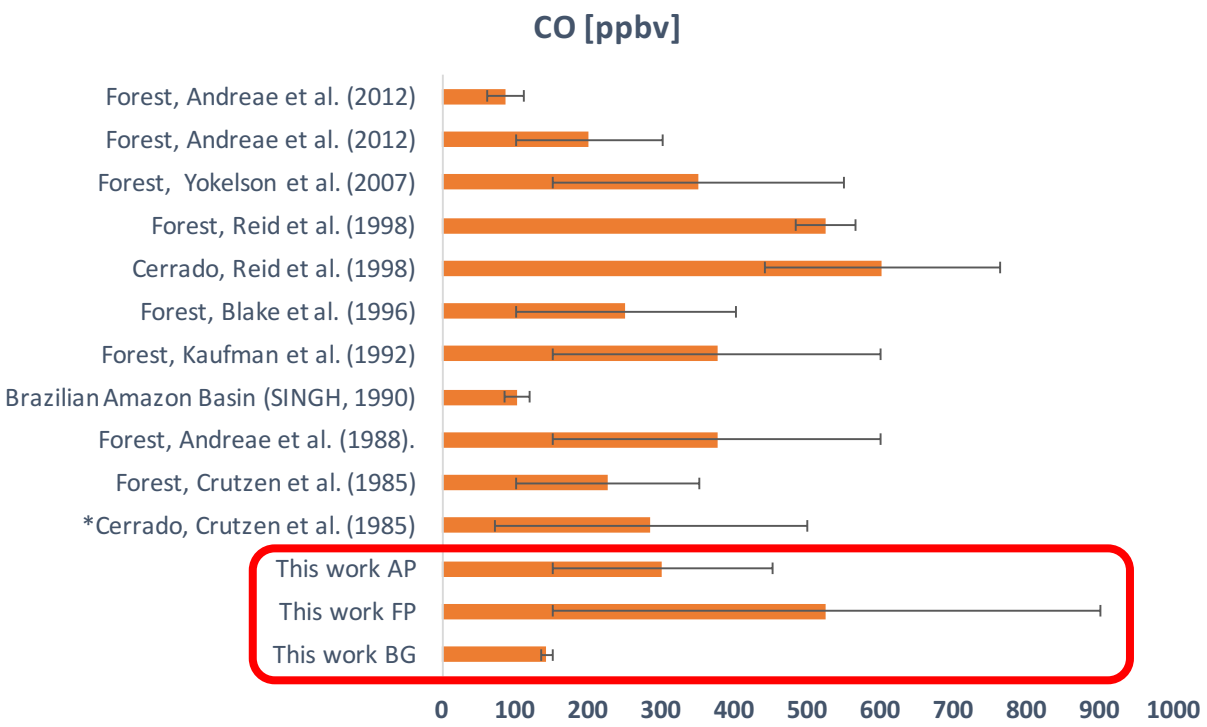
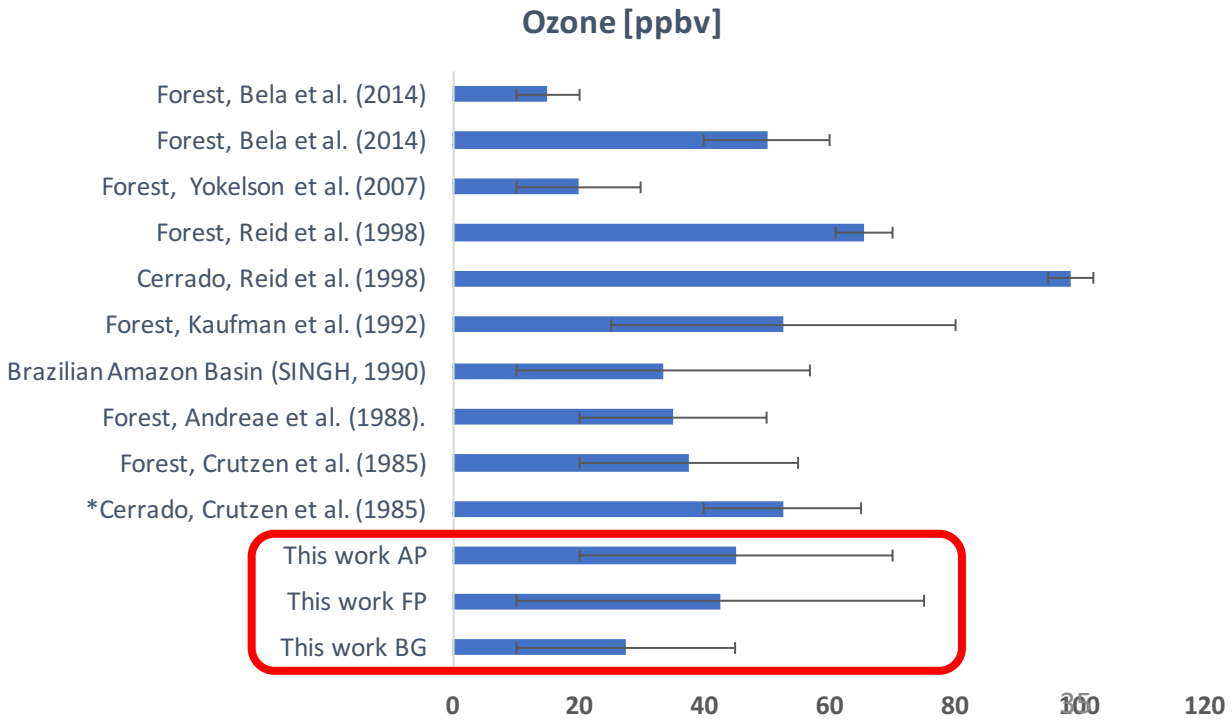
Ambient distribution of NO_x, CO and Ozone



- Low NOx regime for SAMBBA field campaign < 1 ppbv
- High BG level for CO (limited in 150 ppbv)
- Fresh plume presented an CO average ~550 ppbv
- Different pattern for CO and O_3 in FP and AP
- Difference of 20 ppbv for O_3 between BG and AP





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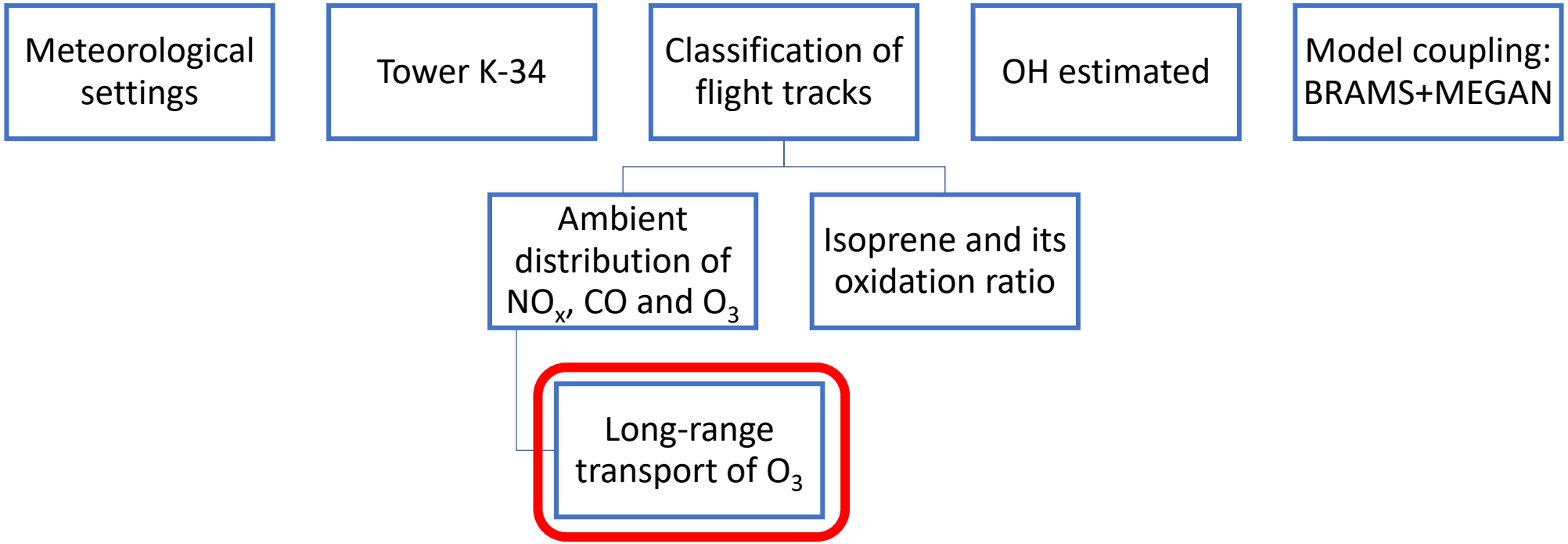
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Long-range transport of Ozone

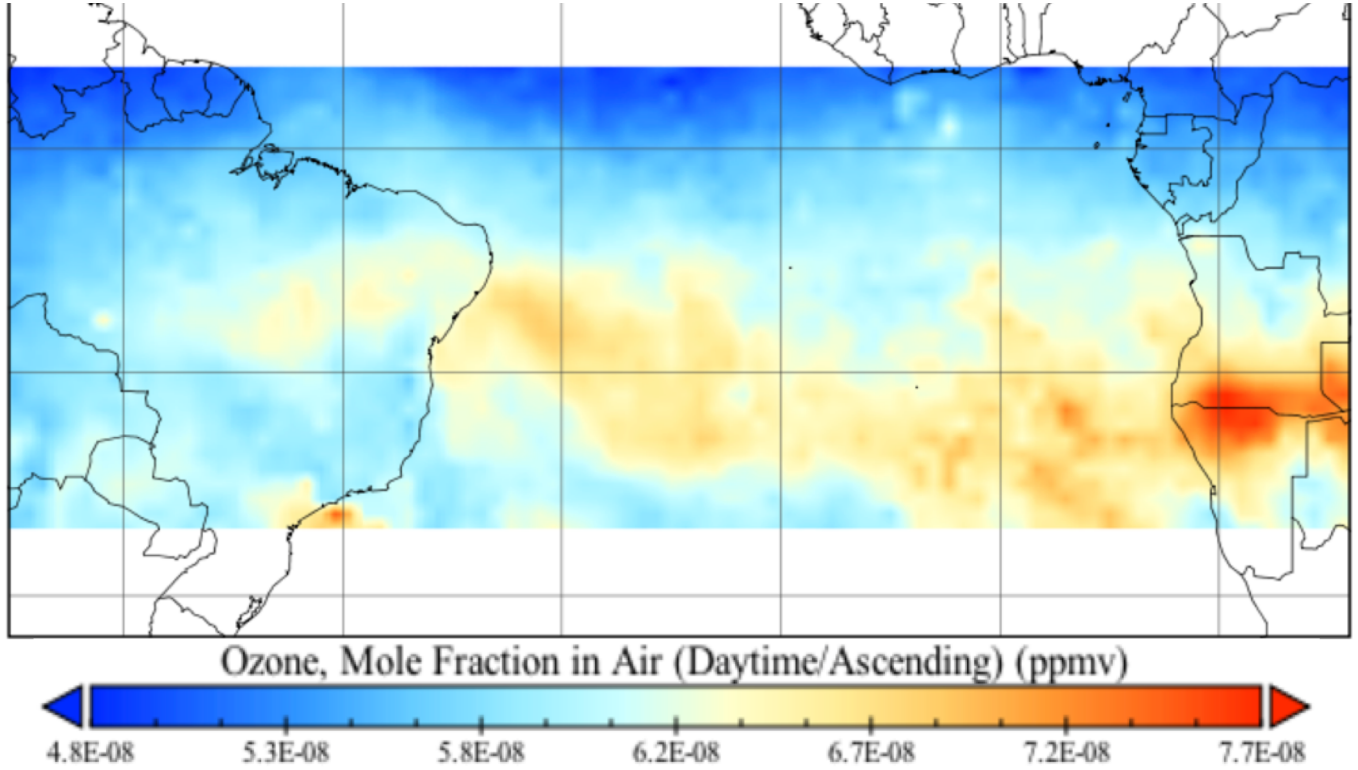
Ozone vertical distribution for all SAMBBA flights following the methodology proposed by Greenslade et al. (2017).

- excluding the first 0.5 km of altitude to avoid noise levels of ozone (ex. biomass burning).

- filter the data ignoring high O_3 events within 0.5 km of the tropopause (reduce uncertainties in lower stratospheric and upper tropospheric layer variations) - ~8 km.

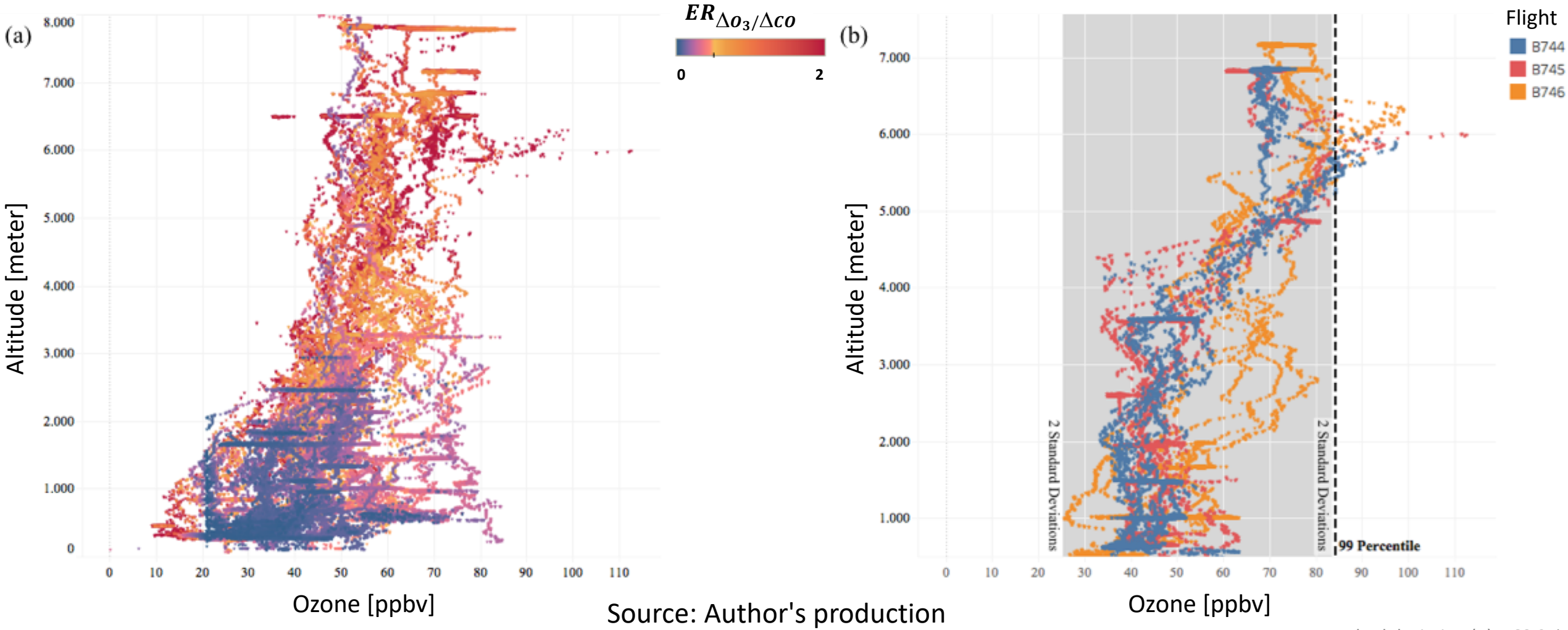
- calculate the 99^{th} percentile perturbation value.

Time averaged Ozone (ppmv) from September 24 to 29 2012 retrieved from AIRS onboard AQUA satellite during daytime at 500 hPa, with 1º x 1º spatial resolution.



Source: Adapted from AIRS.

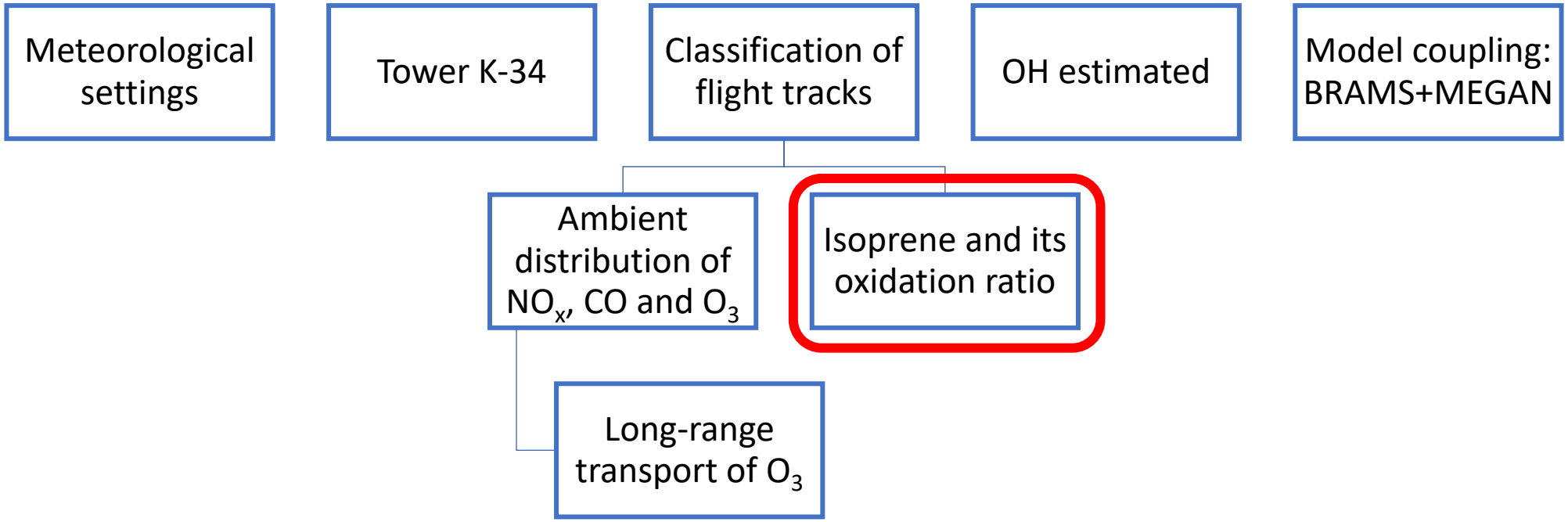
Long-range transport of Ozone



Source: Author's production

1 standard deviation (σ) ~ 68.2%
2 standard deviations (σ) ~ 95.4%
3 standard deviations (σ) ~ 99.7%

Results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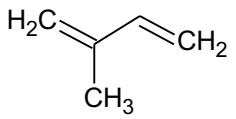
ISOPRENE AND ITS OXIDATION RATIO

Information about the isoprene transport and chemistry can be derived from the isoprene abundance in the atmosphere and the ratio of its oxidation products.

$$\frac{[MVK + MACR + ISOPOOH]}{Isoprene}$$

- distance from the emission source "transport time"
- photochemical degradation
- availability of OH

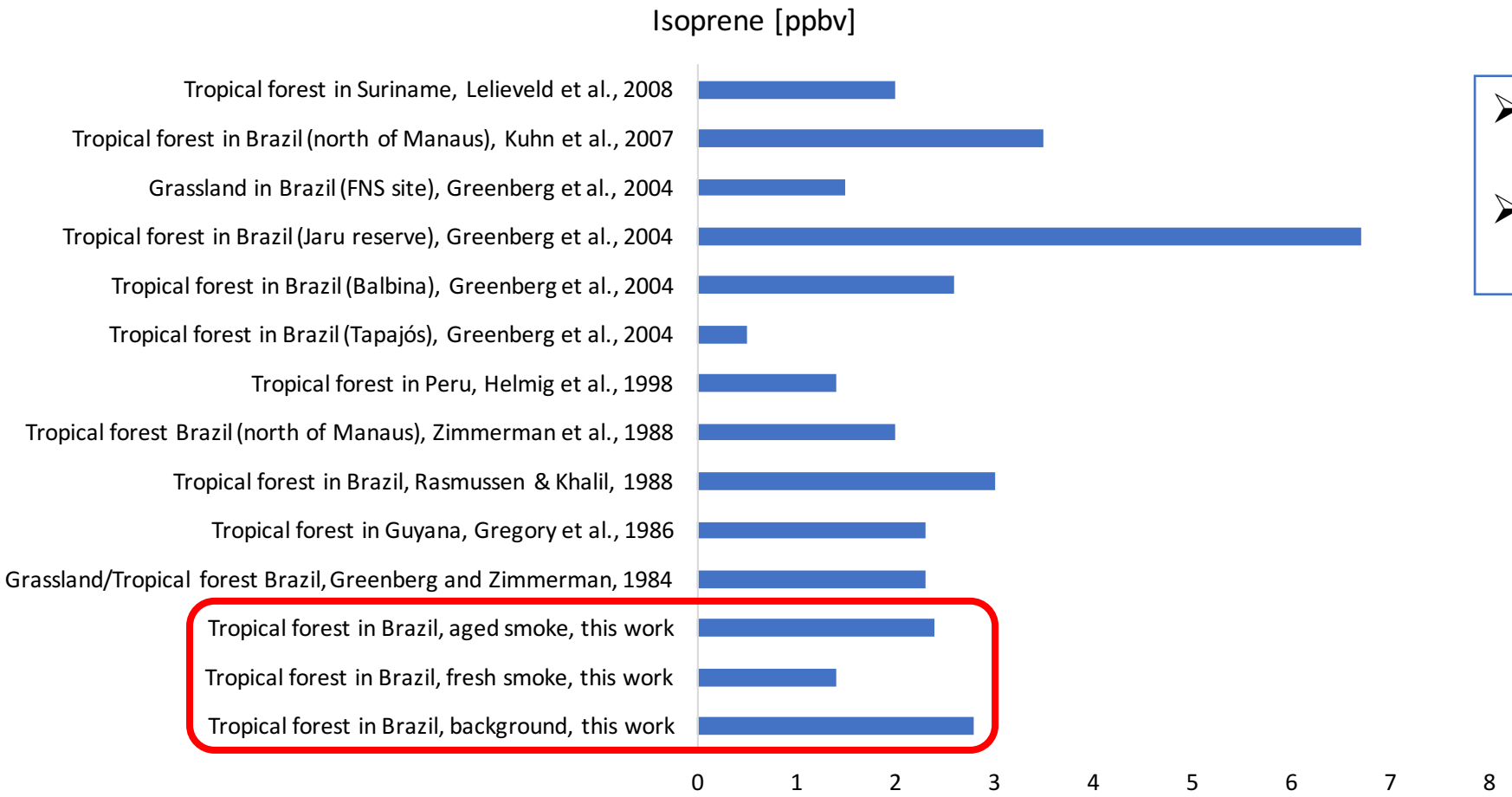
During day time



Isoprene + OH ⟶ MVK + MACR + ISOPOOH

ISOPRENE AND ITS OXIDATION RATIO

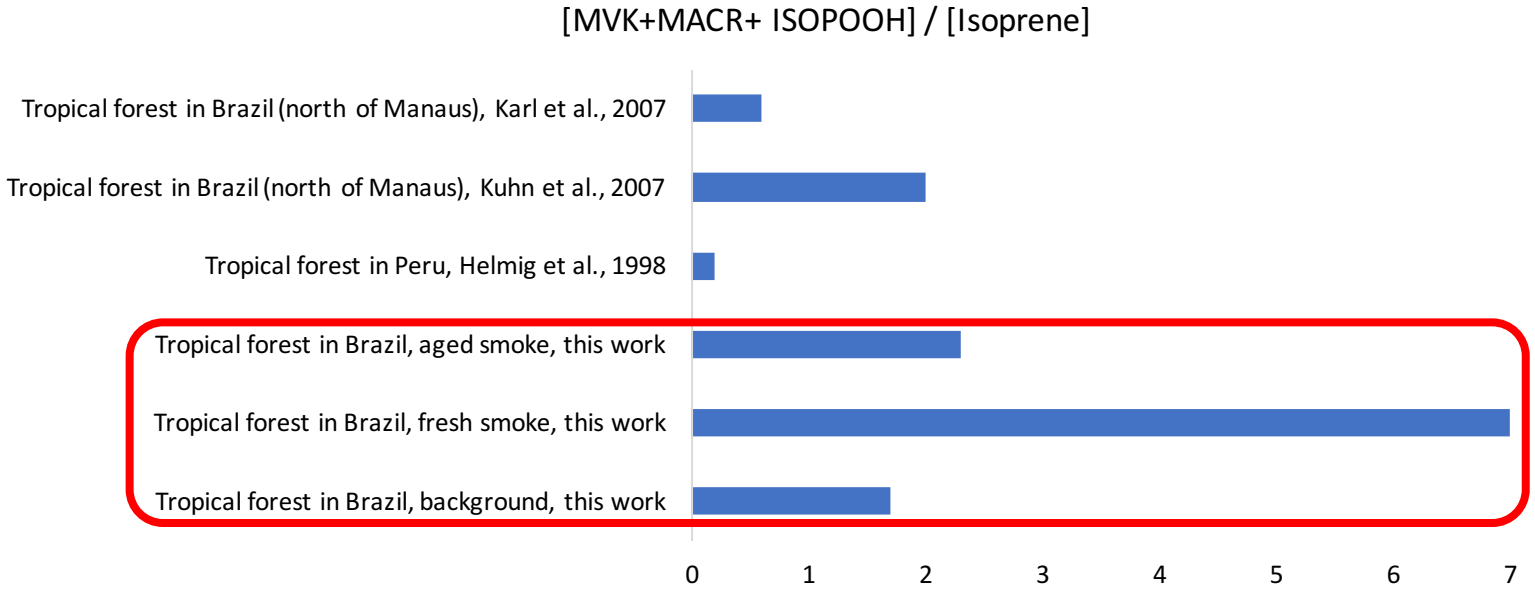
Isoprene mixing ratio in different forest sites. The values correspond to an average in PBL



- Values in agreement with other forest sites
- Fresh smoke plumes presenting low values of Isoprene

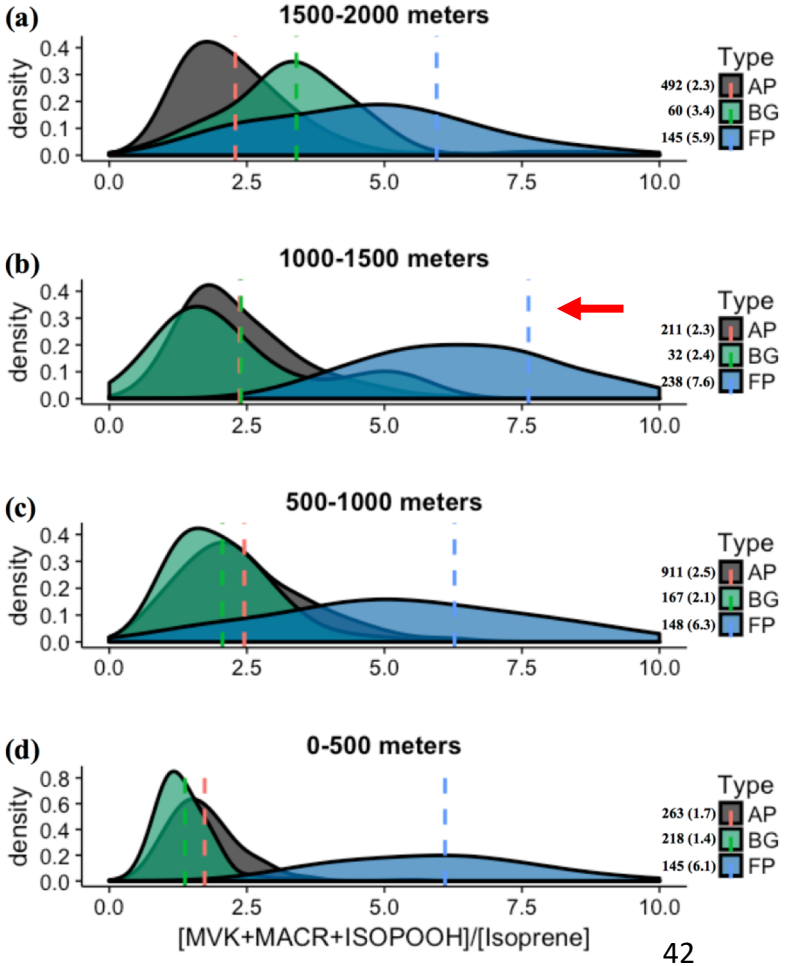
ISOPRENE AND ITS OXIDATION RATIO

[MVK+MACR+ISOPOOH]/[Isoprene] ratio in different forest sites. The values correspond to an average in PBL



- High level of oxidation for Isoprene in fresh smoke plumes
- Increasing [MVK+MACR+ISOPOOH]/[Isoprene] along the altitude
- High level of oxidation in cloud layer for fresh smoke plumes

Density distributions of [MVK+MACR+ISOPOOH]/[Isoprene] at different altitude layers.



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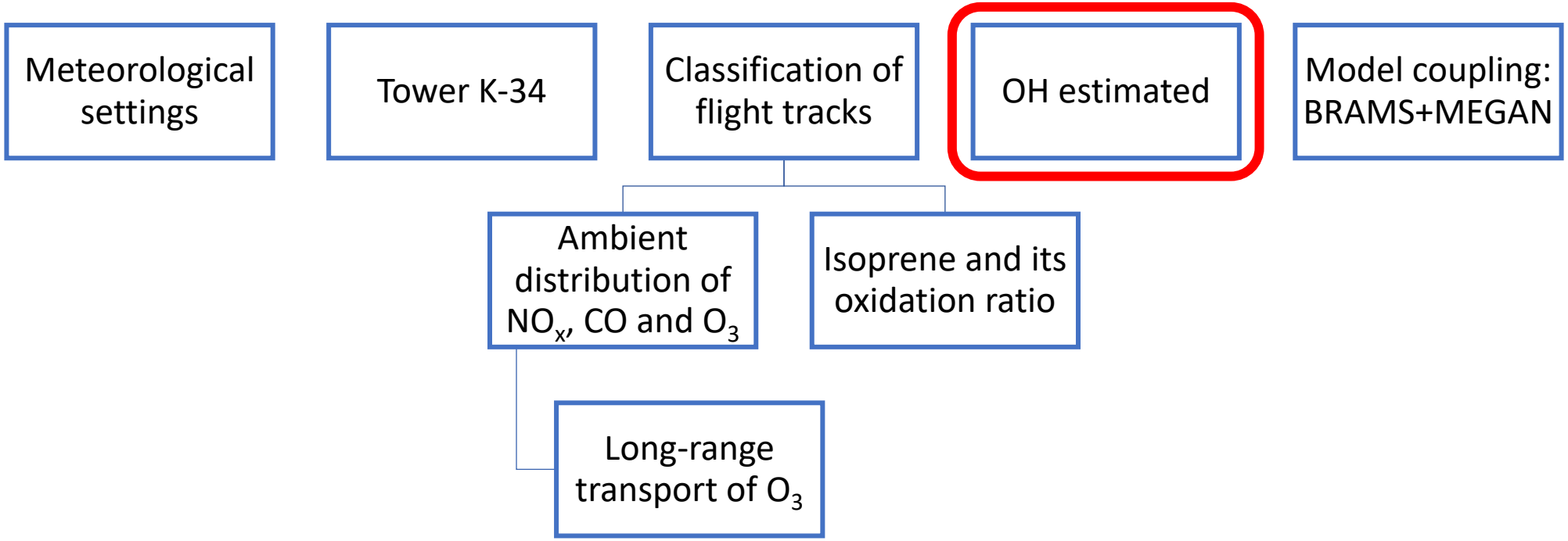
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OH ESTIMATED USING THE SEQUENTIAL REACTION APPROACH

$$[OH] = \left[\ln\left(1 + \left(\frac{\frac{[MVK+MACR+ISOPOOH]}{[Isoprene]} * (K_{iso} - K_{prod})}{K_{iso} * 0.55}\right)\right)\right] \Big/ \left((K_{iso} - K_{prod}) * t\right)$$

Total yield of ***0.55*** of MVK+MACR+ISOPOOH from:

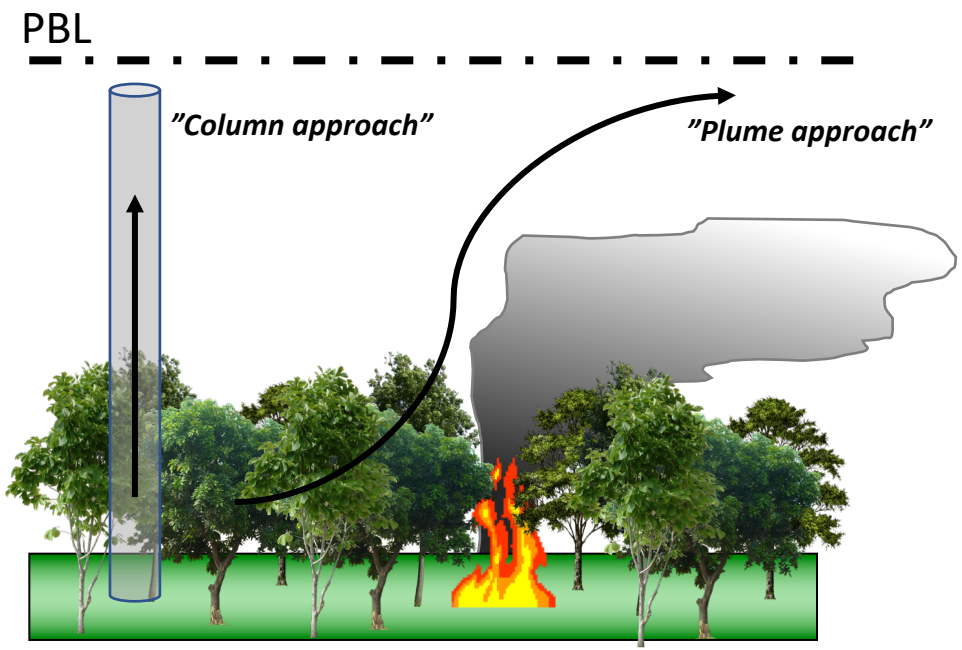
Isoprene + OH → MVK + MACR + ISOPOOH

k_{iso} reaction rate constants of Isoprene + OH

1.1 x 10^{-10} cm^3 $molecules^{-1}$ sec^{-1}

k_{prod} reaction rate constants of [MVK+MACR+ISOPOOH] + OH

6.1 x 10^{-11} cm^3 $molecules^{-1}$ sec^{-1}



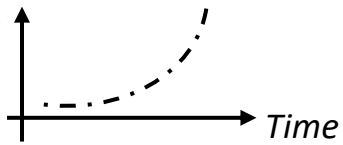
$t = \frac{z_i}{w^*}$

z_i is the PBL depth (m)

w^* is the convective velocity scale (m/s)

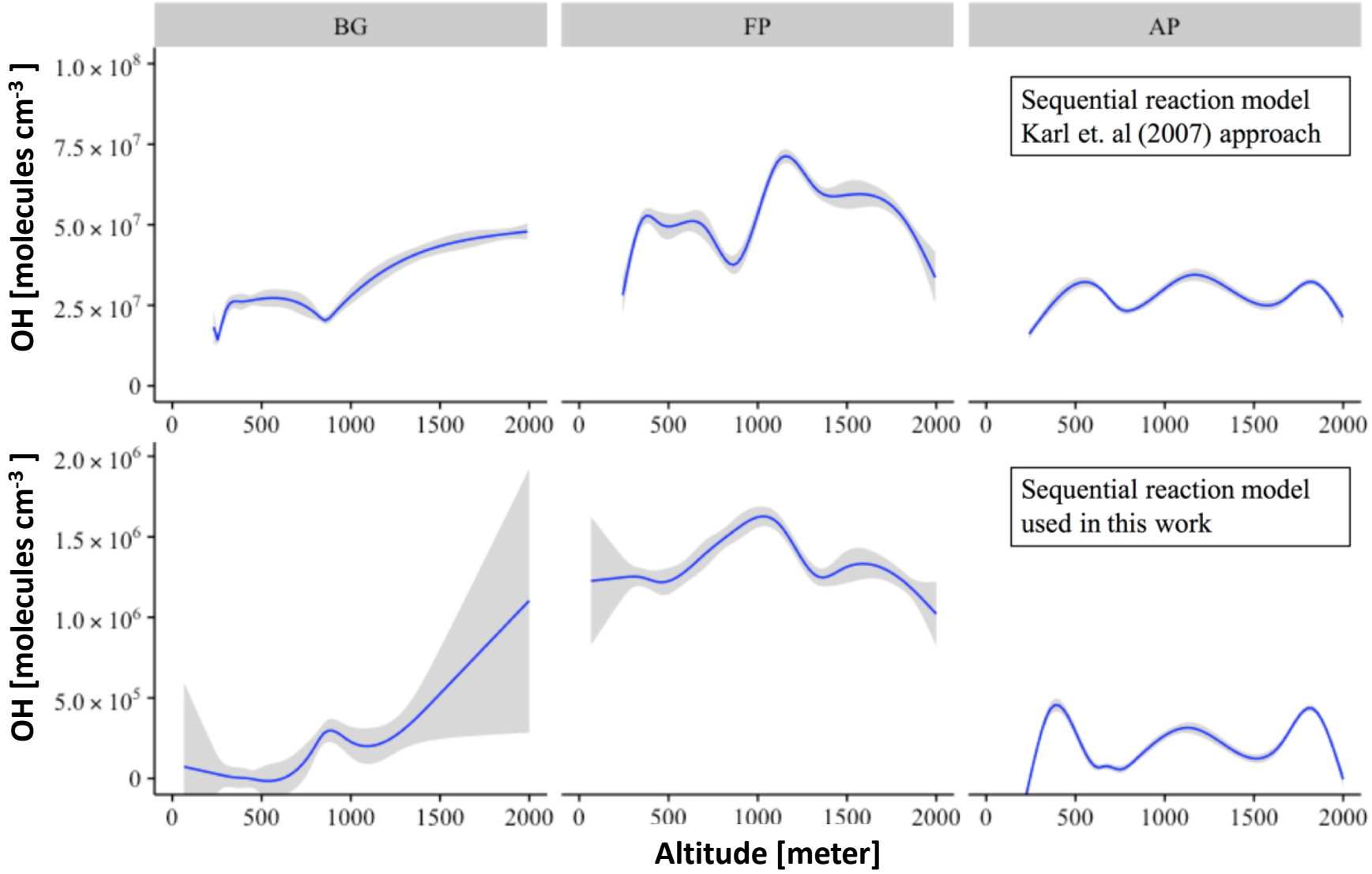
"Column approach" Karl et al. (2007)

t estimated the processing time as $t = 5.3e^{5.4 * ER_{[\Delta O_3]/[\Delta CO]}}$



"Plume approach" this study

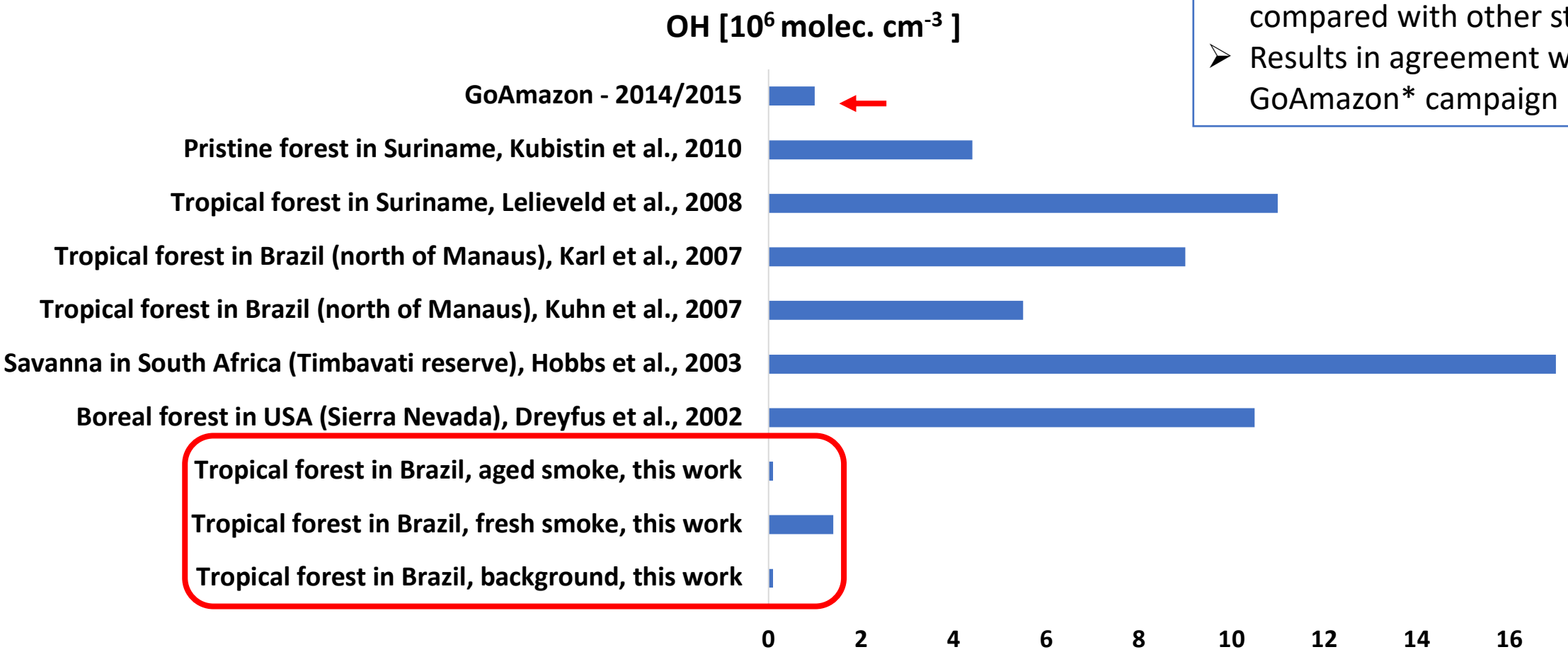
OH ESTIMATED USING THE SEQUENTIAL REACTION APPROACH



- Fresh plume presented high values in both methods
- Maximum at ~1000m (”cloud layer effect”)
- New approach reduced the OH estimated previously by Karl et. al (2007)

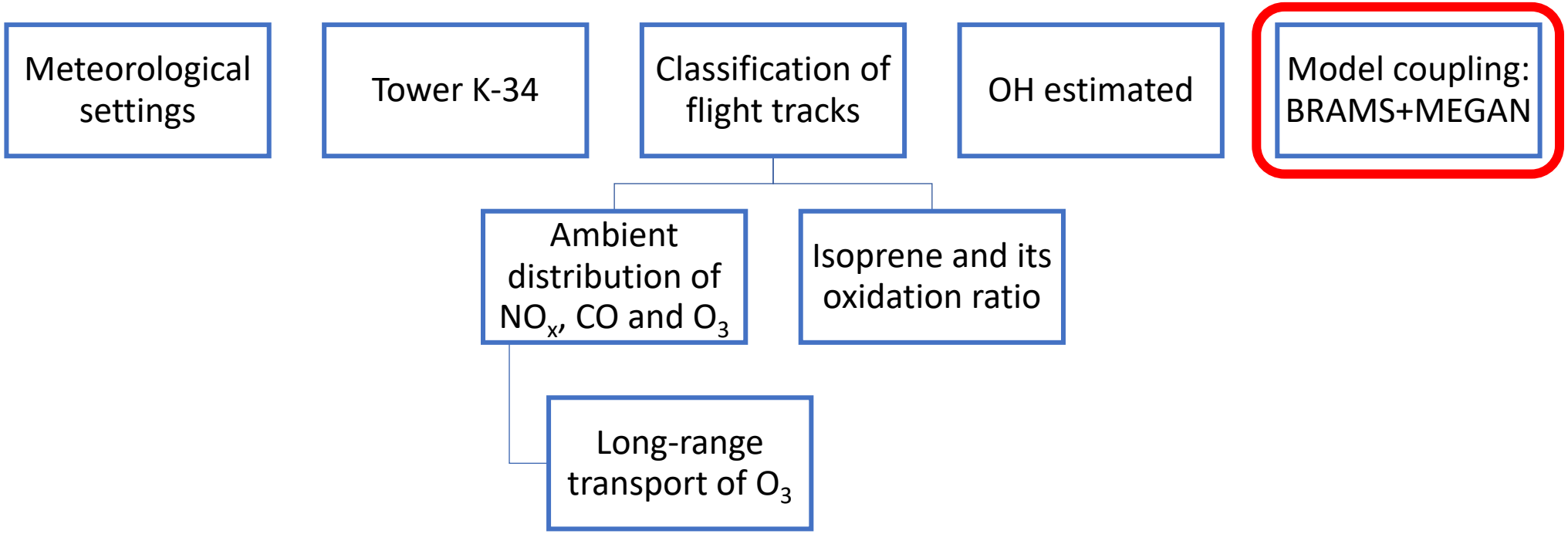
OH ESTIMATED USING THE SEQUENTIAL REACTION APPROACH

OH mixing ratio in different forest sites. The values correspond to an average in PBL.



- Low values for OH estimated when compared with other studies
- Results in agreement with GoAmazon* campaign

Results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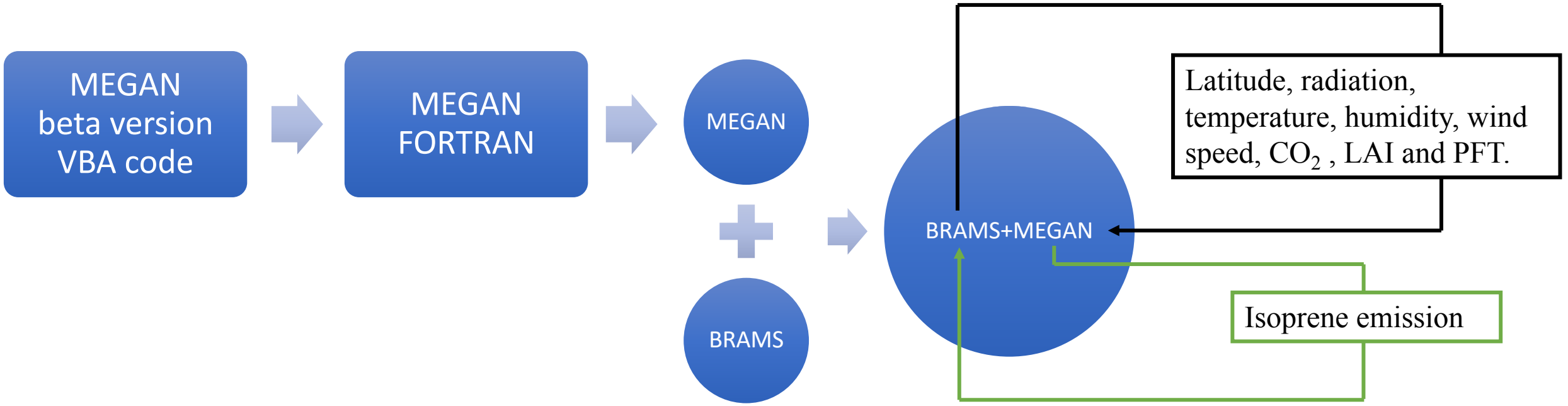
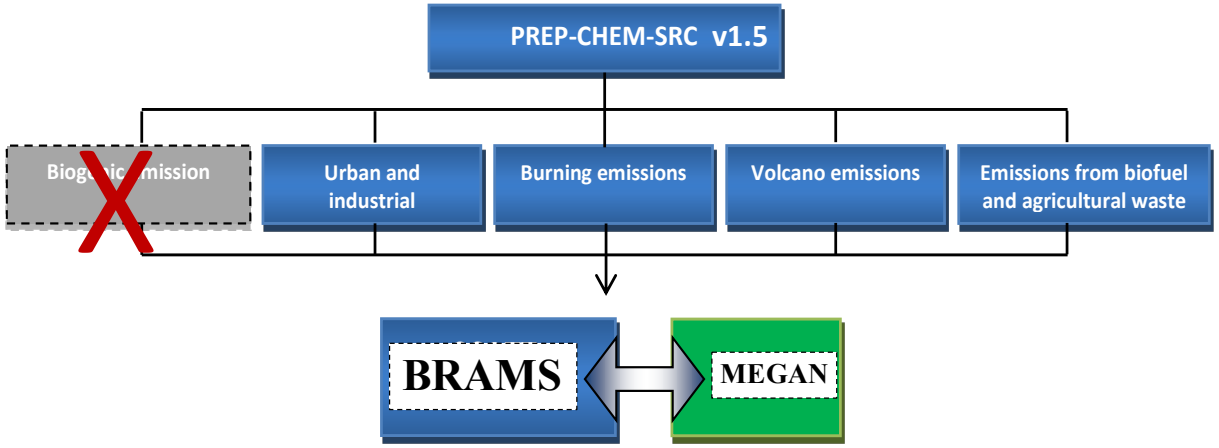
MODEL COUPLING: BRAMS+MEGAN

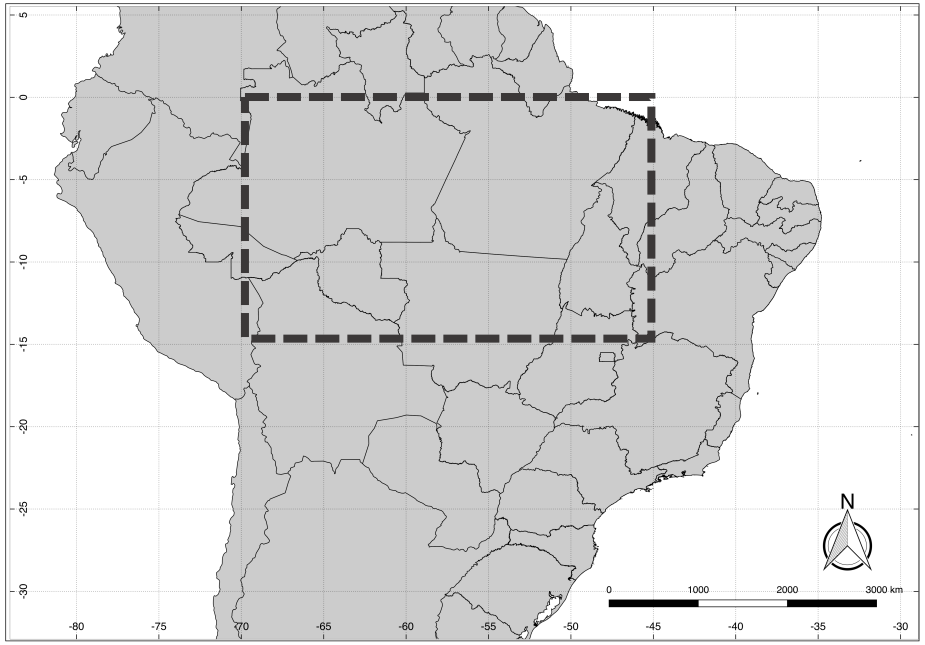




Emissions (PREP-CHEM-SRC) based on ~~climatological data~~ :

Latitude, radiation, temperature, humidity, wind speed, CO_2 , LAI and PFT

MODEL COUPLING: BRAMS+MEGAN

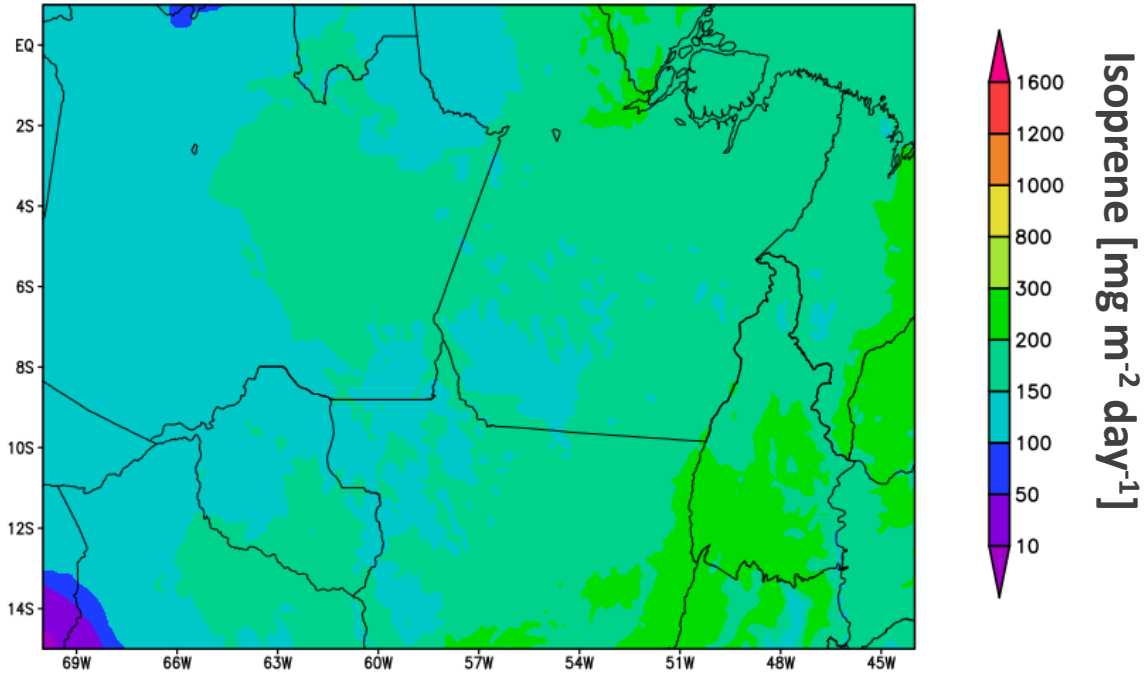


	BRAMS
Grid resolution	10 km
Period of simulation	14 Sep – 4 Oct 2012
Trace gas chemistry	RACM, (STOCKWELL et al., 1997)
Initial chemistry and boundary conditions	MOCAGE
Short/longwave radiation	Based on CARMA
Photolysis	FAST-TUV
Shallow and deep convection	GRELL and DÉVÉNYI (2002)
PBL parameterization	(MELLOR; YAMADA, 1982)

BRAMS-MEGAN offline X BRAMS-MEGAN online

MODEL COUPLING: BRAMS+MEGAN

Average isoprene emissions rate from the model simulations during the SAMBBA period, from 14 September to 3 October 2012: BRAMS-MEGAN online.



BRAMS-MEGAN online in agreement with Gu et al. (2017)

- 200 mg m^{-2} day^{-1} ≈ 8 mg m^{-2} h^{-1}

MODEL COUPLING: BRAMS+MEGAN

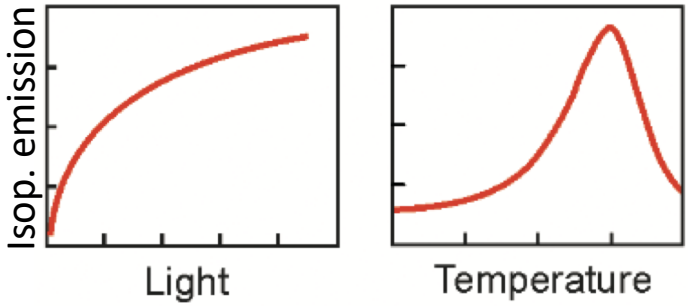
Isoprene	Pearson correlation coefficient		
	CO_2	Surface temperature	Short-wave radiation
BRAMS-MEGAN offline	0.37	-0.34	-0,16
BRAMS-MEGAN online	-0.42	0.62	0.95

BRAMS_MEGAN online
Positive correlation:
- ✓ short-wave radiation (0.95)
- ✓ surface temperature (0.62)

Negative correlation:
- ✓ with CO_2.(-0.42).

In agreement with

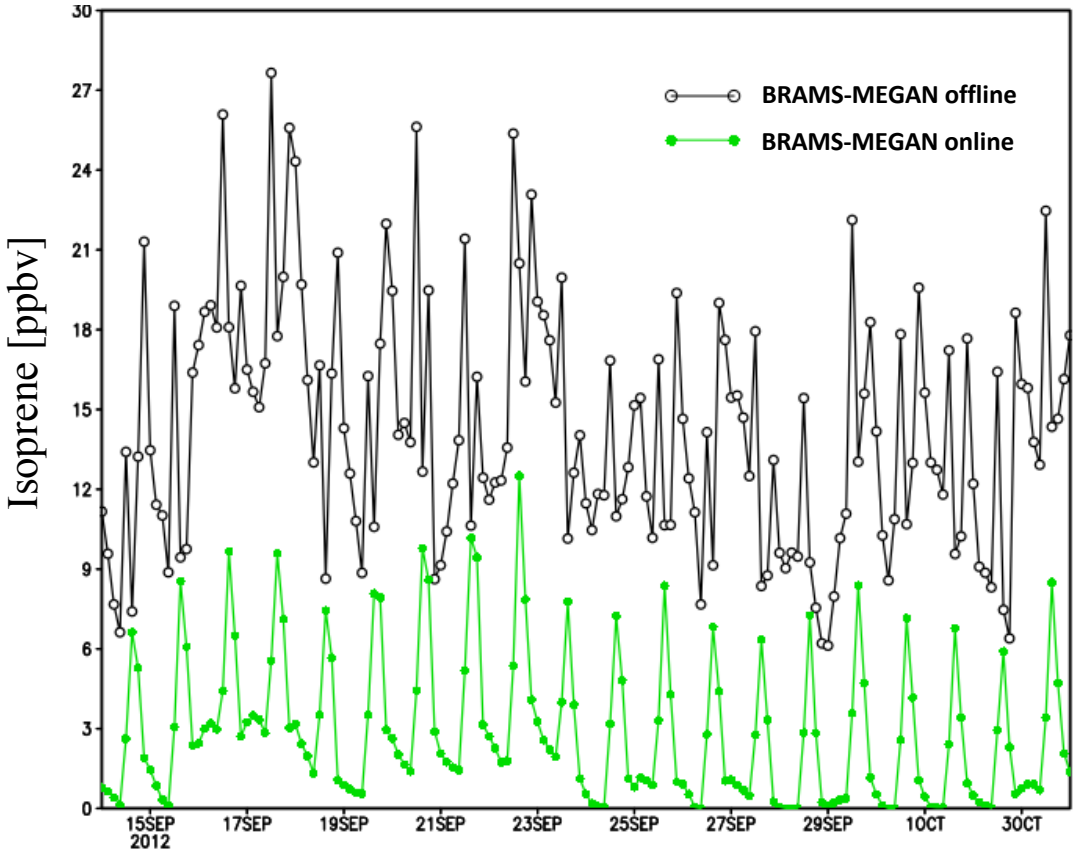
- Inhibition response for CO_2 (Wilkinson et al., 2009)
-

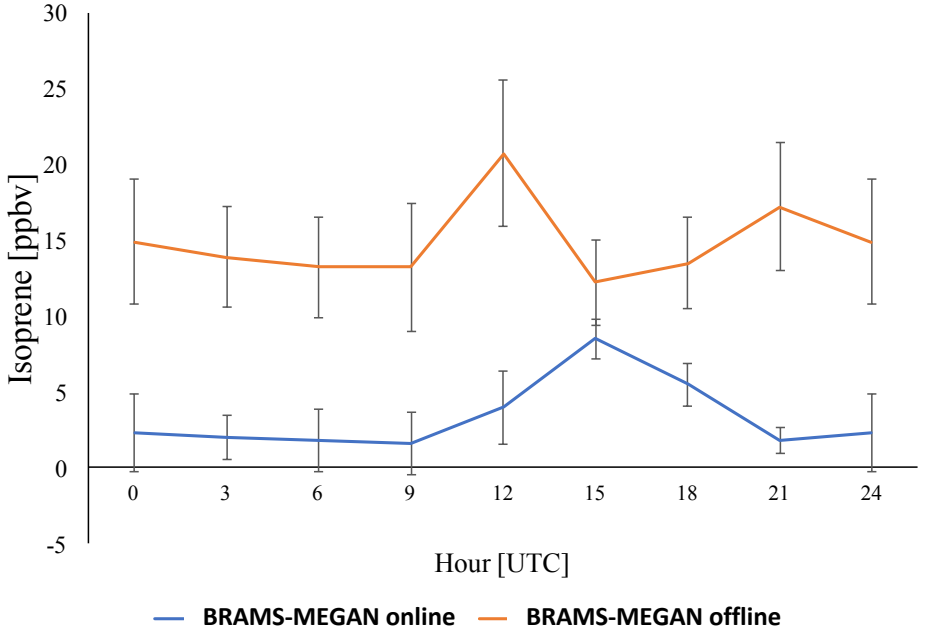


Laothawornkitkul et al. (2009)

MODEL COUPLING: BRAMS+MEGAN

Time series of the isoprene mixing ration in the tower K-34 site at the first model vertical layer (~40 m).

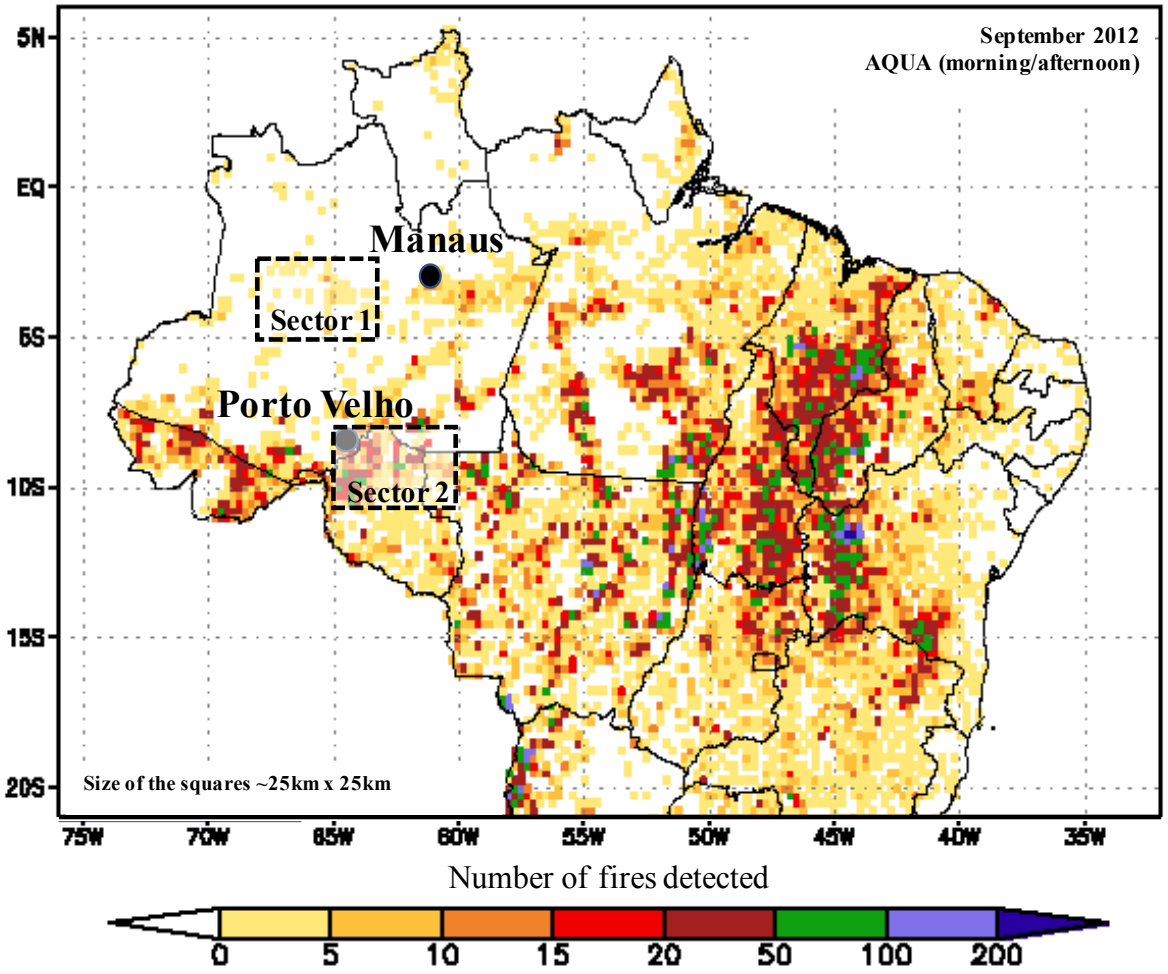




- ✓ Diurnal cycle varying from 1 to 12 ppbv, in agreement with the magnitude of the isoprene at the K-34 tower and previous studies.
- ✓ During nighttime , BRAMS-MEGAN offline isoprene is up to five times higher than the values typically observed in Amazonia.
- ✓ The maximum value of Isoprene during the day adjusted in BRAMS-MEGAN online

MODEL COUPLING: BRAMS+MEGAN

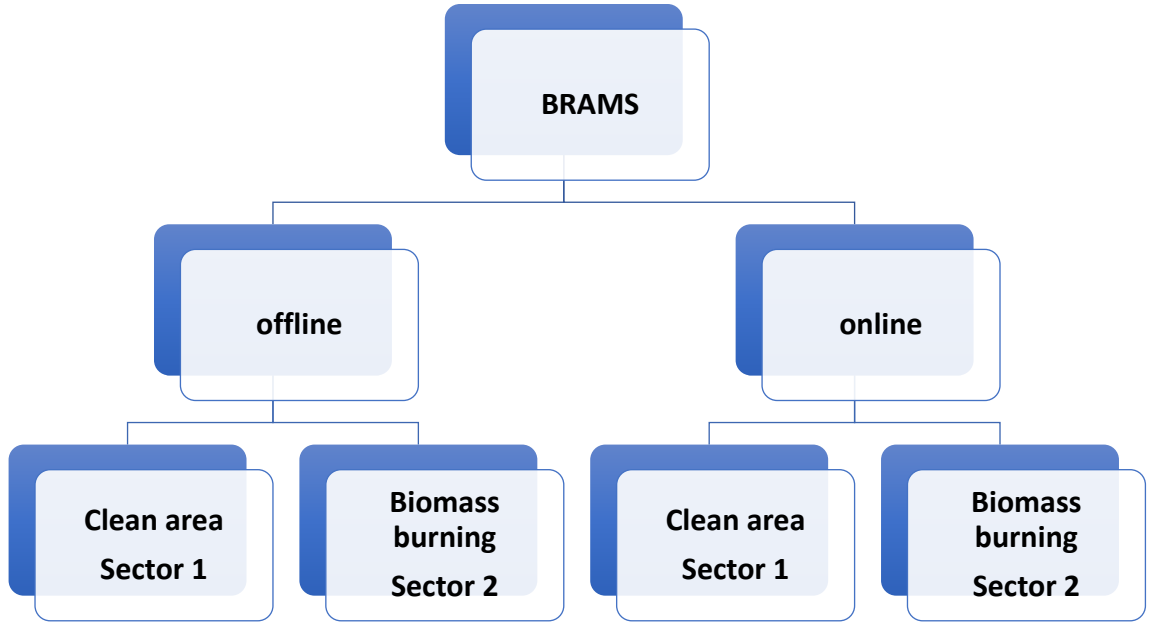
Selected sectors based on number of fires detected from MODIS onboard AQUA during SAMBBA campaign in 2012



Fires detected from MODIS onboard AQUA during SAMBBA field campaign.

Sector 1 → Representative of a clean area

Sector 2 → Impacted by biomass burning emissions



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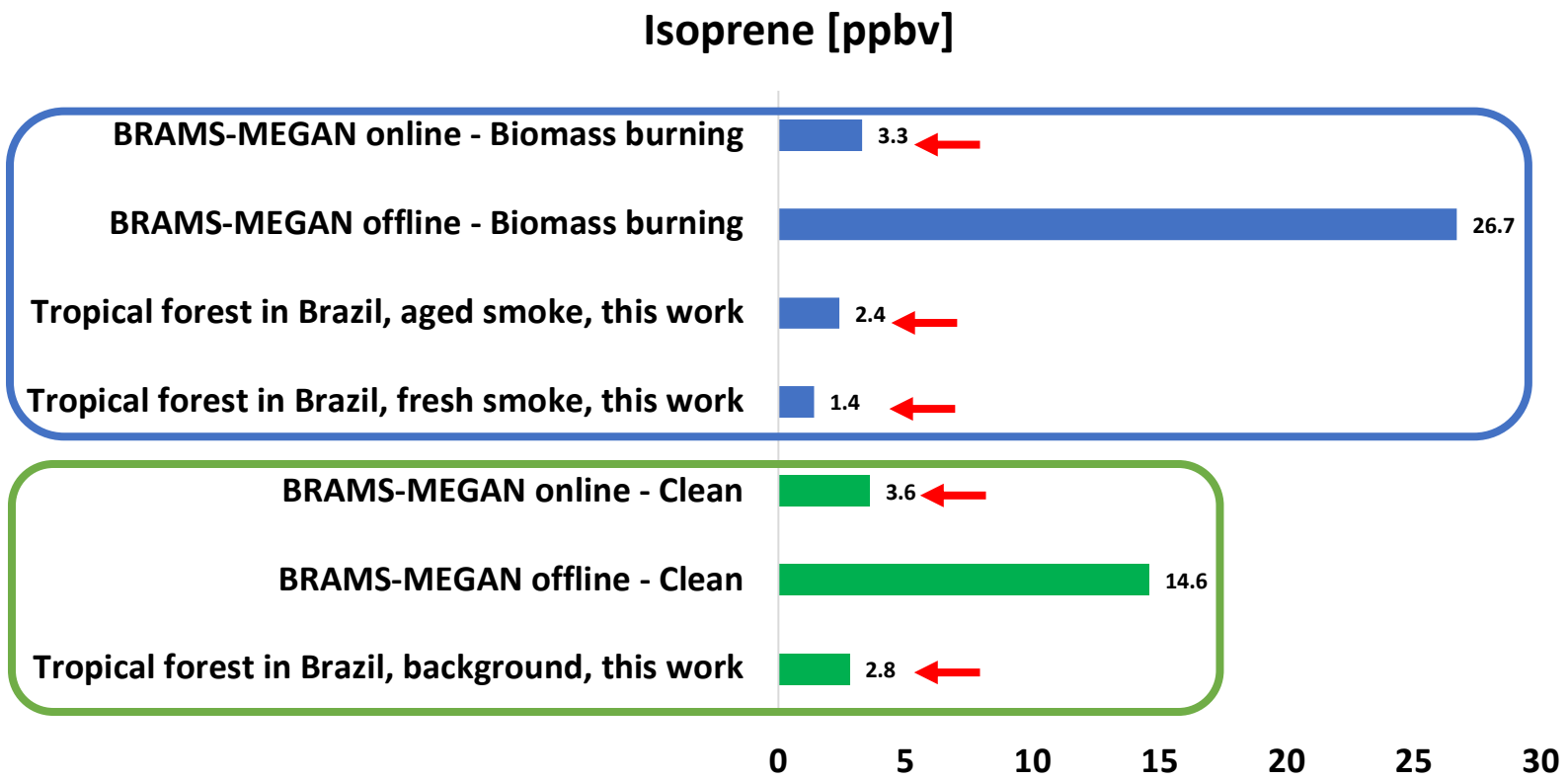


BIOMASS BURNING

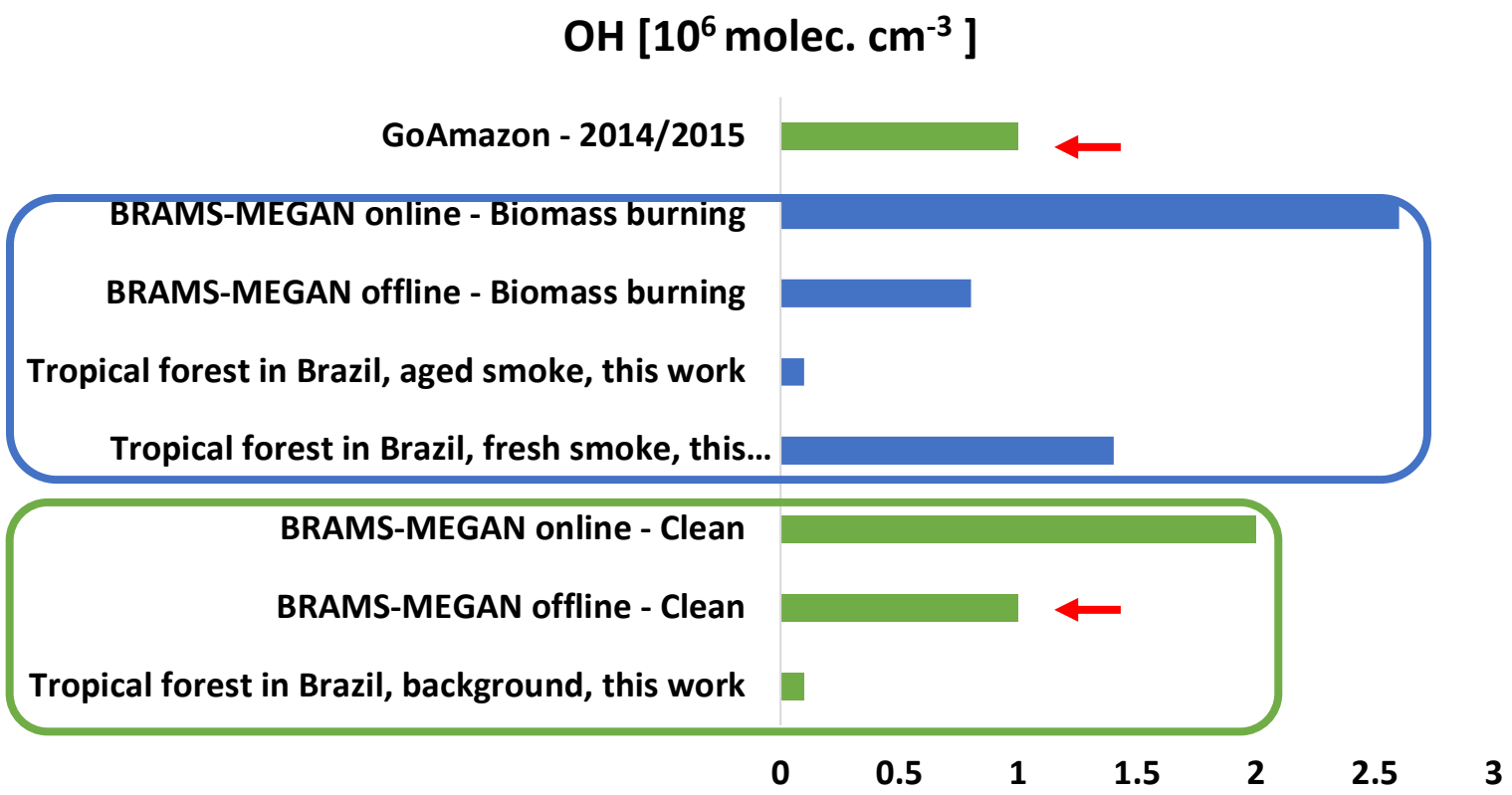
- BRAMS-MEGAN online in agreement with SAMBBA observation
- BRAMS-MEGAN offline overestimate up to 10 times

CLEAN AREA

- BRAMS-MEGAN online in agreement with SAMBBA observation
- BRAMS-MEGAN offline overestimate up to 5 times

- BRAMS-MEGAN online with Isoprene mixing ratio higher in clean than polluted area

MODEL COUPLING: BRAMS+MEGAN



BIOMASS BURNING

- BRAMS-MEGAN online and offline overestimate the OH in aged smoke
- Fresh smoke with higher OH level than aged smoke

CLEAN AREA

- BRAMS-MEGAN online and offline in disagreement with OH estimated in this study.

CLEAN AREA

- BRAMS-MEGAN offline in agreement with GoAmazon.

Conclusion

Conclusions

Observations:

➢ Clean area more exposed to radiation and polluted region with high temperatures

➢ Wind circulation pattern carries pollutants from the southeastern part of the forest to clean regions (north-west and central), besides the influence of long-distance transport of O3 that was also detected during the experiment

➢ Isoprene levels found in the K-34 tower are in agreement with the literature

➢ Chemical classification method was useful in identifying different phases of biomass burning - "chemical regimes"

➢ SAMBBA experiment occurred in low levels of NOx, but with high values of O3 and CO in the regions affected by biomass burning.

➢ Intense oxidation in FP (~ <2 hours) through high levels of [MVK + MACR + ISOPOOH] / [Isoprene]

➢ OH estimated with the new "plume approach" changed the OH values to lower levels, in accordance with the results obtained during GoAmazon - 2014/2015

➢ Intensification of OH levels by 1,000-1500 m, confirming the cloud layer effect.

Conclusions

BRAMS-MEGAN online

- improved the isoprene emission levels, in agreement with observations
- agree with MEGAN, with a positive correlation with temperature and radiation, and an inhibition response for CO_2
- Isoprene mixing ratio in the first model vertical layer (~40 m) agree with observation and K-34 tower measurements
- Nighttime isoprene mixing ratios returning to the background values
- maximum value of Isoprene during the day adjusted
- increase the isoprene mixing ratio from biomass burning to clean regime in agreement with SAMBBA field campaign
- overestimate the OH in clean and biomass burning regime

Future work

Suggestions for future work

- Improve the OH estimation developed in this study and use as a possible diagnosis tool for chemical mechanisms
- More tests to adjust the currently coupling, expanding to other biogenic compounds and increasing the MEGAN model complexity (past 24/240 hours)
- Emphasis on fundamental studies that may assist the correct prediction of aerosols in numerical models, with a synergist approach between observations and numerical models