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Characteristics, future trends, and transport path analysis of atmospheric formaldehyde pollution: A case study of Argentina

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Abstract: *Formaldehyde is a carcinogen and plays an essential role in photochemical reactions. Understanding its distribution and dynamics in the atmosphere significantly protects human and ecosystem health. In this paper, the temporal and spatial distribution of tropospheric formaldehyde column concentration and its influencing factors in Argentina from 2010 to 2021 are studied, and the future trend of formaldehyde is discussed. The results showed that the spatial distribution of formaldehyde gradually decreased from a high concentration in the northeast region to a low concentration in the southwest region. The concentration of formaldehyde column has fluctuated and increased in the last 12 years. The concentration of formaldehyde is highest in winter. Formaldehyde has a high correlation with NDVI (Normalized Difference Vegetation Index) and lift index but a low correlation with air pressure. The topographic factor plays an aggregation role in formaldehyde concentration during atmospheric transport. It is expected that formaldehyde in Argentina may continue to show a slight increase trend.*

Keywords: *Formaldehyde; spatial-temporal distribution; Ozone Monitoring Instrument; influencing factors; Argentina*

1. Introduction

Formaldehyde (HCHO) is the simplest volatile organic compounds (Oxygenated Volatile OrganicCompounds, OVOCs) in the atmosphere with relatively high reactivity (Boeke N. L.,et al.,2011; Atkinson R.,et al.,2003). HCHO is not only a photochemical product at atmospheric nitrate but also can participate in photochemical reactions to generate oxidizing solid pollutants such as ozone (O_3) and Peroxyacetyl nitrate (PAN) (Jiang Z.H., et al., 2016). Motor vehicle exhaust emissions and agricultural straw burning will also increase formaldehyde concentration. In indoor sources, various building materials, plywood, blankets, wooden products, flooring, decoration, and decorative materials will slowly and continuously emit formaldehyde (Liu J.S., et al.,2023).

The World Health Organization has identified formaldehyde as a carcinogen and teratogenic substance, accounting for more than 50% of the risk assessment of 187 harmful air pollutant concentrations determined by the International Agency for Research on Cancer (IARC). Indoor concentrations as high as 0.5mg/m^3 will cause human tears and abnormal eye sensitivity. The throat is uncomfortable or painful when the formaldehyde concentration is higher than 0.6 mg/m^3. It can also cause nausea, cough, chest tightness, and wheezing. Formaldehyde concentrations above 30 mg/m^3 can cause immediate death. Long-term exposure to high concentrations of formaldehyde can cause nasopharyngeal cancer and cardiovascular and cerebrovascular diseases and lead to chromosomal abnormalities and gene mutations (Scheffe R.D.,et al.,2016; Zhang Y.J., et al.,2009).The traditional formaldehyde monitoring method cannot measure the concentration of tropospheric formaldehyde in large areas, long time, and high efficiency, so it is more advantageous to use remote sensing satellite technology to monitor tropospheric formaldehyde. Currently, there are many sensors for remote sensing monitoring of air pollution, including GOME(The Global Ozone Monitoring Experiment, ERS-2

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launched in 1996), SCIAMACHY(The Scanning Imaging Absorption Spectrometer for Atmospheric Chartography, ENVISAT launched in 2002), and GOME-2 (Global Ozone Monitoring Experiment 2 ,METOP-A launched in 2007 and 2013) and OMI(Ozone Monitoring Instrument ,aboard the AURA satellite launched in 2004) (Zhang Y., 2019; Zhang X.,et al.,2019; Darreh-Shori T.,et al.,2019; Zhuang L.Y., et al.,2019), among them. OMI data has the highest inversion accuracy and the most comprehensive coverage, the high spatial resolution increases the sensitivity considering the tropospheric measurement, which can be used to study atmospheric pollution (Li C.,et al.,2010; Zhang J., et al.,2015).

Several authors have used remote-sensing satellite data to study formaldehyde. Zhu L.,et al.(2014) concluded that in Texas, under the influence of human factors, formaldehyde is mainly related to waste gas generated by the refining and petroleum industries. Marais E.A.,et al. (2012) concluded that the main factors contributing to the formaldehyde concentration in South America were the plant emissions of isoprene and the burning of biomass. Schedlbauer A.,et al. (2011) used OMI's tropospheric formaldehyde column concentration data and the GEOS-Chem model to study the spatial and temporal distribution of tropospheric formaldehyde column concentrations in different regions worldwide. Based on the formaldehyde daily product data of Aura-OMI sensor L2-V003, Li L.,et al. (2020) showed that the interannual mean value of formaldehyde column concentration in Shaanxi Province, China, from 2010 to 2018 showed a fluctuating upward trend. The spatial distribution decreased to the north and south in the upper Guanzhong region, while the stability of formaldehyde concentration decreased to the south along the Qinling Mountains. Xian L.,et al. (2018) showed that the formaldehyde concentration in the Pearl River Delta was (13.11-15.14) $\times 10^{15}$molec$\cdot$cm^{-2}, and the total energy consumption and industrial waste gas emissions accounted for a large proportion. Wang F.,et al. (2021) took INTEX-B anthropogenic sources, FINNv1 biomass combustion sources, and MEGAN biological sources as the benchmark sources. The study showed that anthropogenic sources increased by 1.51 times, 1.87 times, and 1.93 times, respectively, in the three urban agglomerations of Beijing-Tianjin-Hebei, Yangtze River Delta, and Pearl River Delta in China, which are currently the most polluted. Biomass combustion sources increased by 12.2 times, 6.15 times, and 2.27 times in the three regions, and biological sources increased by 1.66 times, 1.31 times, and 1.21 times in the three areas, respectively[1-6].

There is little research on atmospheric formaldehyde pollution. Therefore, this paper used OMI formaldehyde remote sensing satellite data from 2010 to 2021, combined with vegetation and meteorological data, and HYSPLIT to study atmospheric formaldehyde pollution in Argentina to provide a basis for environmental protection in the study area.

2. Study area

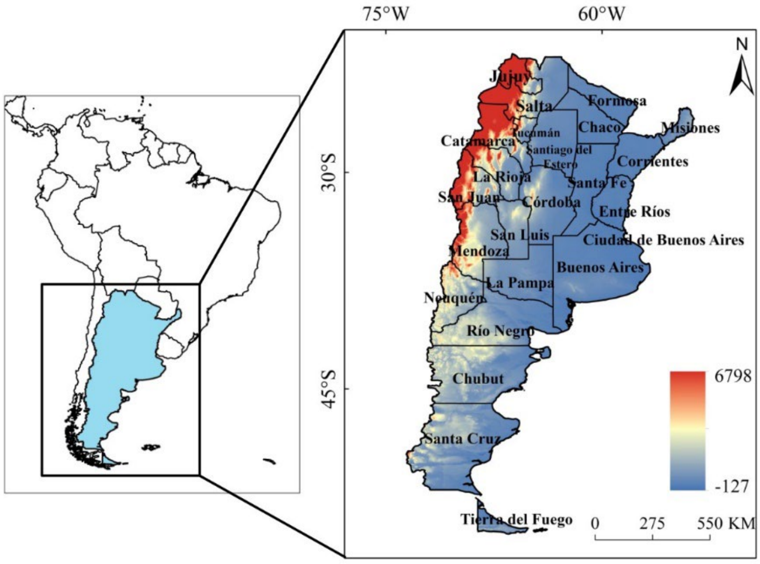

Figure 1: Diagram of the study area

Argentina (73°W~54°W,22°S~54°S) is in the southern part of South America (Fig.1), bordered by the Atlantic Ocean to the east, the Andes Mountains to the west with Chile, and Bolivia, Paraguay, Brazil, and Uruguay to the north and east. The Andes Mountains in the west, of which the mountainous area accounts for 30.64% of the country's size, the Pampas grassland in the east and central, the Patagonia plateau in the south, Argentina's overall terrain is high in the west and low in the east. Argentina has a diverse climate, with the main climate types being savanna, subtropical monsoon, and temperate continental. Fig.1 is the diagram map of the research area. The colors represent altitude, and red means high altitude[7-15].

3. Datas and methods

3.1 Data source

The formaldehyde data in this paper come from the data monitored by the Ozone Monitoring Instrument carried by NASA's AURA satellite. This sensor has the characteristics of high accuracy and wide range. The working principle is to obtain information by observing the backscattered radiation of the Earth's atmosphere and Earth's surface. Its orbital scanning amplitude is 2600km, the spatial resolution is 13km×24km, and the spectral resolution is about 0.45nm (Boeke et al., 2011; Li et al., 2013).

Normalized Difference Vegetation Index (NDVI) is a standardized index that reflects plant growth and vegetation coverage. The NDVI used in this paper is MODIS Level 3 product from 2010 to 2021. From NASA (https://code.earthengine.google.com/).It quantifies vegetation by measuring the difference between near-infrared (strongly reflected by vegetation) and red light (absorbed by vegetation). The product coverage degree, high precision, light, and resolution are 1000 m.

The oil and coal data, energy consumption, and electricity consumption used in this paper are from the World Development Bank database. Land cover data from the National Center for Basic Geographic Information (http://wwww.ngcc.cn) include natural and artificial land cover, which can broadly reflect the synergistic effects of human activities and natural factors on the atmosphere (Wei et al., 2021).

The HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory model) model used in this paper is a professional model developed by the Air Resources Laboratory of the National Oceanic and Atmospheric Administration (NOAA) of the United States and the Australian Bureau of Meteorology (https://www.ready.noaa.gov/archives.php) in the past 20 years to calculate and analyze the transport and diffusion trajectory of air pollutants[16-28].

3.2 Data processing method

3.2.1 OMI data processing

In this paper, daily data of troposphere L2-v003HCHO in Argentina from 2010 to 2021 are obtained, and the data is read by HDF-view software. ArcGIS10.2 software was applied to format conversion, kriging interpolation, masking, and calculation of the data, as well as the spatial distribution maps of the annual quarterly. The spatial distribution of formaldehyde column concentration's annual, seasonal, and monthly mean values were obtained.

3.2.2 Spatial computation method

This paper uses the Pearson correlation analysis method (Mu S.J.,et al.,2012), which can measure whether the two data groups show a linear trend index, and the correlation coefficient range is [-1,1]. This method analyzes the relationship between formaldehyde column concentration and water content, air temperature, pressure, and rise index. The MODIS data from 2010 to 2021 downloaded from NASA's official website are selected for NDVI data processing. The calculation formula is:

$$R_{xy} = \frac{\sum_{i=1}^{n}[(x_i-\bar{x})(y_i-\bar{y})]}{\sqrt{\sum_{i=1}^{n}(x_i-\bar{X})^2 \sum_{i=1}^{n}(y_i-\bar{y})^2}} \tag{1}$$

In the formula, R_{xy} represents the correlation coefficient between two variables x and y, n is the number of samples, x_i represents the average value of HCHO column concentration in the year i, and y_i represent the value of water content, air temperature, air pressure, rise index, and vegetation coverage in year i. $\bar{x}$ represents the multi-year mean of HCHO,$\bar{y}$ represents the multi-year mean of other data, and i represents the year.

3.2.3 HYSPLIT model data processing

This paper used the HYSPLIT model (Yang H.,et al.,2017) to simulate pollution and analyze the source of formaldehyde transport. This method has been widely used at home and abroad to study pollutants' source and transport path (Draxler R.R.,et al.,1997). The principle of the HYSPLIT model is as follows: it is assumed that the moving trajectory of particles is floating with the wind field, and the moving trajectory of particles is a vector integral in space and time. The specific calculation formula is as follows:

$$p(t + \Delta t) = p(t) + 0.5[v(p,t) + v(p', t + \Delta t)]\Delta t \quad (2)$$

$$p'(t + \Delta t) = p(t) + v(p,t)\Delta t \quad (3)$$

In the formula, p is the initial position of the particle; p' is the first guess position; v is wind speed; t is the location $p(t + \Delta t) = p$ time; Δt is the step length, the change range is (1min~1h), and the movement of the air mass must meet the $v_{max}\Delta t < 0.75$ grid distance.

3.2.4 Slope trend analysis

In this paper, David Freedman's single linear regression trend analysis method was used to fit the variation trend of each formaldehyde column concentration pixel over on a spatial scale (Hovland H.J.,et al.,1977; Li F.S., et al.,2020) studied the interannual spatiotemporal variation of formaldehyde in Argentina from 2012 to 2021. The calculation formula is:

$$\theta_{slope} = \frac{n \times \sum_{i=1}^{n}(i \times HCHO_i) - \sum_{i=1}^{n} i \sum_{i=1}^{n} HCHO_i}{n \times \sum_{i=1}^{n}(i^2) - \left(\sum_{i=1}^{n} i\right)^2} \quad (4)$$

In the formula, θ_{slope} is the slope of the HCHO concentration value in each pixel, n is the cumulative number of years in the study period, i is the year, and $HCHO_i$ is the formaldehyde column concentration value in the year i. $\theta_{slope}>0$ indicates that the formaldehyde column concentration shows an increasing trend with the increase of years, and $\theta_{slope}<0$ indicates that the formaldehyde column concentration shows a decreasing trend with the increase of years. The greater the absolute value of θ_{slope}, the greater the variation range of the HCHO column concentration value (Huang Y.Y., et al.,2019).

3.2.5 Hurst index

The Hurst index can reflect the future change trend of a time series and is generally calculated using the R/S analysis method (Nan, 2021; Lixia et al., 2020). The trend strength can be judged according to the Hurst index value. When H=0.5, the change is relatively random. When 0<H<0.5, the future trend is expected to decrease, showing an anti-sustainability. The future performance is sustained when 0.5<H<1 (Xiaowei and Rui, 2015). The future trend corresponding to the Hurst index value interval can be subdivided into five levels: weak anti-sustainability, strong anti-sustainability, random change, weak sustainability, and strong sustainability[29-33].

4. Results and discussion

4.1 Temporal and spatial characteristics of formaldehyde column concentration and its trend analysis

4.1.1 Interannual spatial variation of formaldehyde column concentration

Fig.2 shows Argentina's interannual spatial distribution of formaldehyde column concentrations from 2010 to 2021. To analyze the spatial variation rules and characteristics of Argentine formaldehyde, the concentration of HCHO was divided more clearly into six grades according to its maximum value (17.72×10^{15}molec·cm^{-2}) and minimum value (8.07×10^{15}molec·cm^{-2}). The results showed that the average formaldehyde concentration in Argentina was 13.01×10^{15}molec·cm^{-2}, with prominent spatial distribution characteristics. The high concentration was concentrated in the northeastern region, while the concentration was low in the southern region, showing a descending spatial distribution pattern from the northeast to the southwest. The second, third, and fourth levels were the main concentrations, and the first and second levels were concentrated in the western and southern regions. This may be related to the topography and land and sea location, and the west is the Andes mountains and the southern coast. The air diffusion rate is fast, and formaldehyde is challenging to gather.

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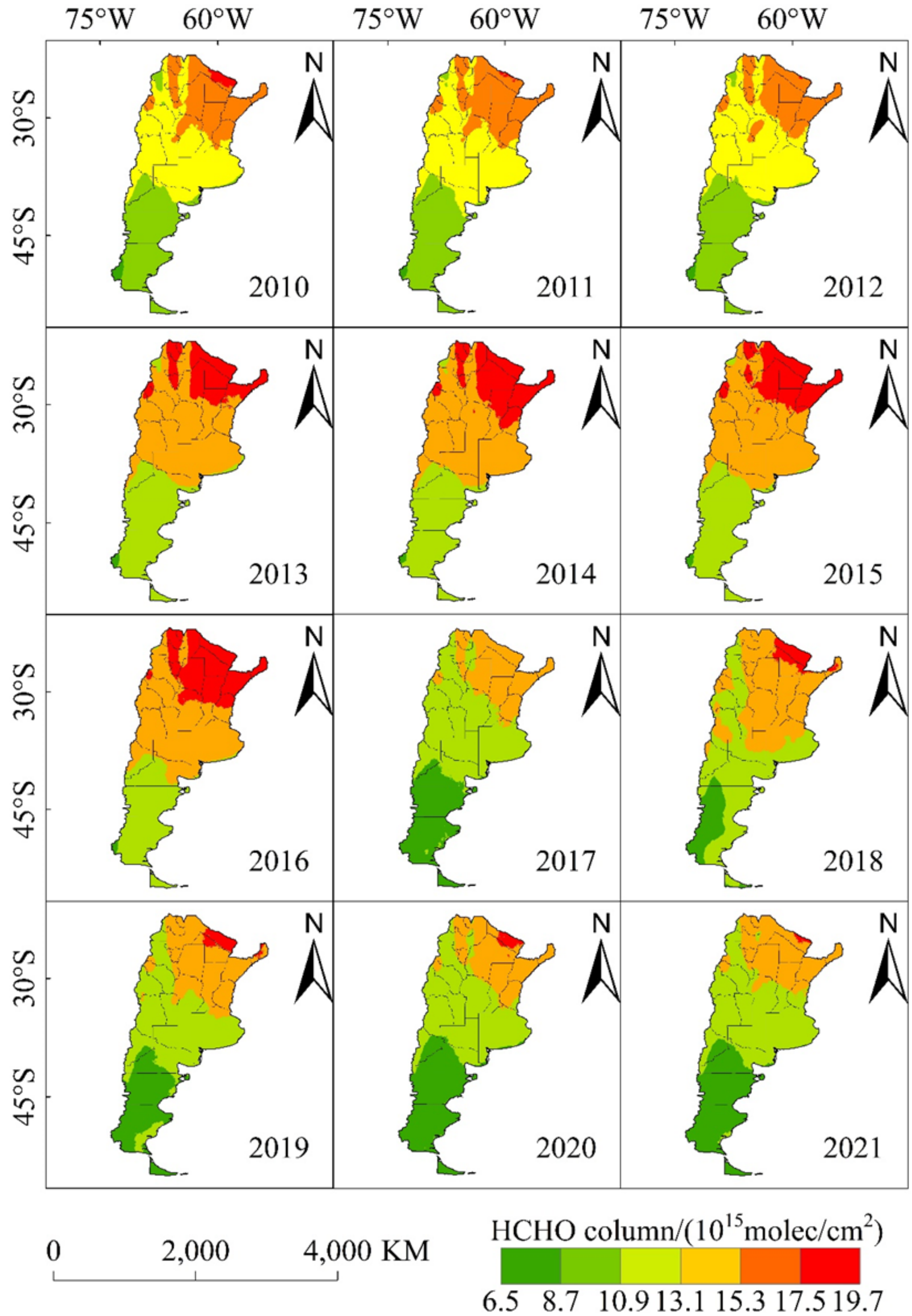

Figure 2: Interannual spatial distribution of formaldehyde column concentration in Argentina from 2010 to 2021

Combined with the inter-annual variation grade diagram of formaldehyde column concentration in Argentina from 2010 to 2021 (Fig. 3), it was found that the formaldehyde column concentration had slight fluctuation and showed an overall upward trend, among which the formaldehyde column concentration was the highest in 2018 (14.16×10^{15}molec·cm^{-2}) and the lowest in 2013. The concentration of formaldehyde column was 12.47×10^{15}molec·cm^{-2} from 2010 to 2013, which showed an overall downward trend and reached the lowest concentration value in 2013. In Argentina, the relevant legislation in that year required the path of sustainable development, the appropriate sustainable management of the environment, and the strengthening of the control of waste pollutants from industrial and service activities.

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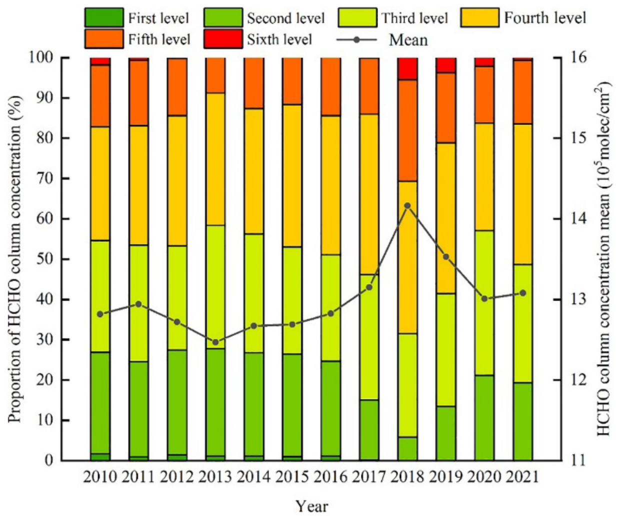

Figure 3: Annual variation trend of formaldehyde column concentration in Argentina from 2010 to 2021

4.1.2 Monthly spatial variation of formaldehyde column concentration

Fig.4 shows the spatial distribution of Argentina's monthly mean column concentration of formaldehyde. It can be seen from the figure that the formaldehyde concentration decreases from northeast to southwest in general. From January to March, the area of Grade 5 formaldehyde column concentration in the northeast region shows a decreasing trend. From July to December, the concentration of formaldehyde column shows a fluctuation trend, first decreasing and then increasing.

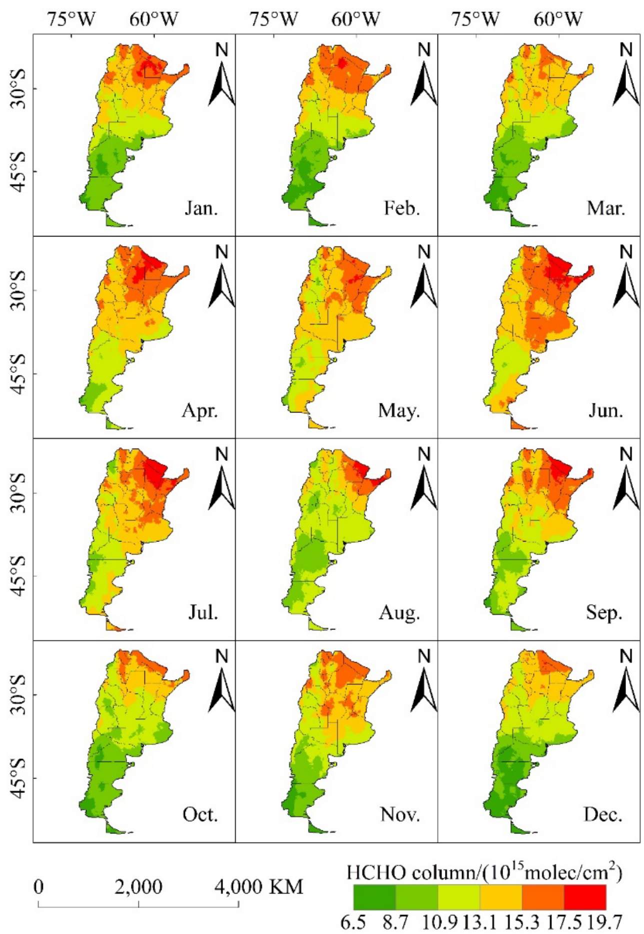

Figure 4: Monthly mean distribution of formaldehyde column concentration in Argentina from 2010 to 2021

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4.1.3 Trend analysis of formaldehyde column concentration

The Slope and Hurst index can be used to analyze the past and future trends of formaldehyde column concentration in Argentina (Liang Z.H., 2022; Huang R.R.,2022; Duan J.L.,2022). Table 1 is the interval table of Slope and Hurst grades. Hurst is also divided into five levels, namely weak sustainability (0<H<0.25), strong sustainability (0.25<H<0.5), random variation (H=0.5), weak sustainability (0.5<H<0.75), and strong sustainability (0.75<H<1).

Combined with the Hurst spatial distribution map in Argentina, the Hurst value ranges from 0.55 to 0.76, with an average value of 0.63. The area of formaldehyde column concentration showing weak persistence (0.5<H<0.75) in the future reaches more than 98%, and the distribution is uniform. The vital persistence area (0.75<H<1) is only 2%, scattered in Argentina, and is not considered. The results showed that the slight increase in formaldehyde column concentration in Argentina was the primary trend in the future, and the overall change was not significant, as shown in Figure 5.

Table 1: Slope and Hurst grade interval table

Level	Slope	Past trend	Hurst	Future trend
1	$-0.16 < \theta_{slope} < -0.08$	Significantly decreased	0<H<0.25	Weak discontinuity
2	$-0.08 < \theta_{slope} < -0.01$	Slightly decreased	0.25<H<0.5	Strong discontinuity
3	$-0.01 < \theta_{slope} < 0.06$	Basically unchanged	H=0.5	Random variation
4	$0.06 < \theta_{slope} < 0.14$	Slightly increased	0.5<H<0.75	Weak persistence
5	$0.14 < \theta_{slope} < 0.21$	Significantly increased	0.75<H<1	Strong persistence

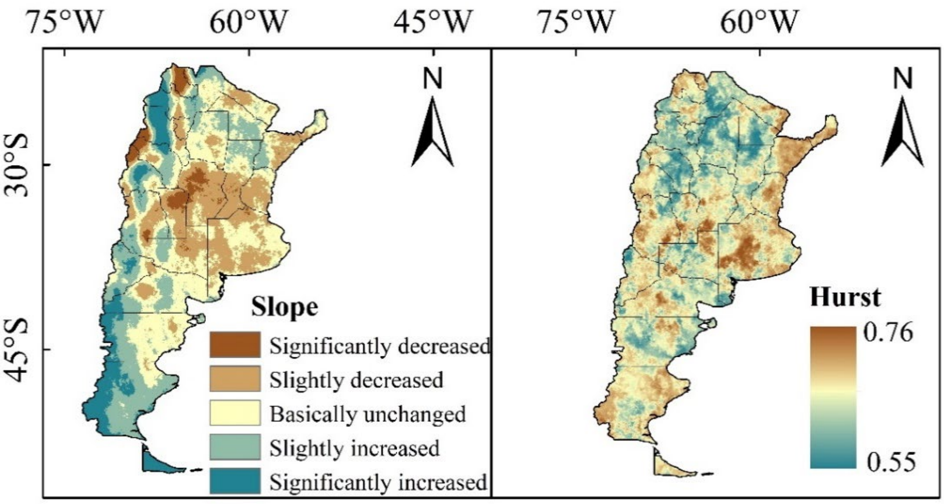

Figure 5: Trend analysis of formaldehyde column concentration in Argentina in 2010—2021

4.2 Analysis of influencing factors of formaldehyde column concentration

To better analyze the factors affecting the formaldehyde column concentration in Argentina, this paper is divided into human and natural factors.

4.2.1 Human activity factor

Fig.6 shows mean formaldehyde column concentration and spatial distribution of DMSP/OLS night light. DMSP/OLS is an important branch of remote sensing application, which can indirectly reflect the characteristics of urbanization level and population activities. It is one of Argentina's two most polluted provinces, and the high concentration area is consistent with the high-value area of the light data. At the same time, the coincidence degree of these two parts is high in the figure, indicating that human activities are an essential factor affecting the concentration of formaldehyde columns[34-40].

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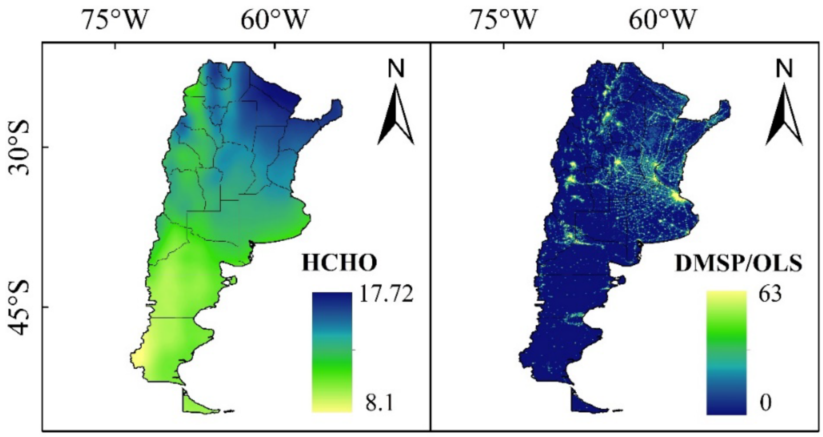

Figure 6: Mean formaldehyde column concentration and spatial distribution of DMSP/OLS night light

4.2.2 Vegetation factor

As shown in Fig.7, the regions with high correlation are mainly distributed in the central and western regions and most of the coastal areas of the central and southern regions. The released isoprene was the main factor for the increase in formaldehyde concentration. In this region, formaldehyde column concentration significantly correlated with NDVI, with the correlation coefficient ranging from -0.64 to 0.73 and the positive correlation area reaching 61.51%. LAI (Leaf area index) was a comprehensive index representing the canopy structure and community growth status of vegetation. Leaf area and leaf age have been widely used in studies on changes in HCHO (Trostdorf et al., 2004), and the correlation between Argentine formaldehyde column concentration and LAI ranged from -0.84 to 0.81.

BVOC emissions contribute significantly to local formaldehyde consistency (Barkley et al., 2008).

Therefore, vegetation is another crucial factor affecting the formaldehyde concentration value.

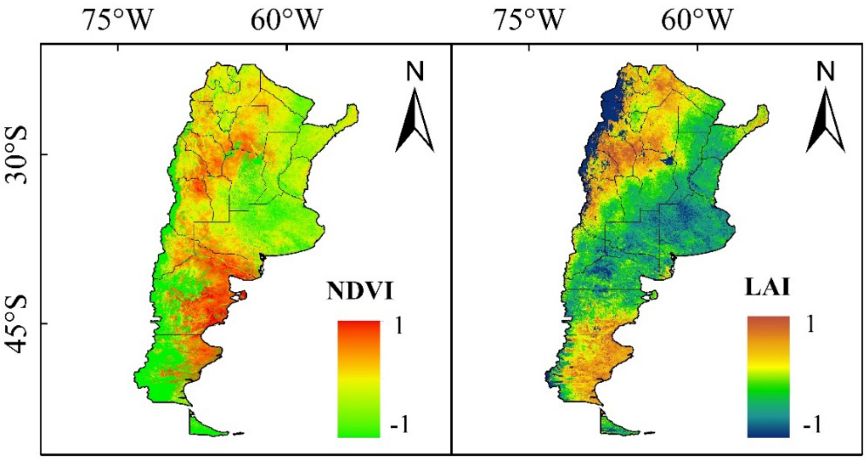

Figure 7: Correlation graph between formaldehyde column concentration and NDVI and LAI

4.2.3 Climatic factor

Atmospheric water content can reflect the abundance of atmospheric water vapor, an essential factor affecting climate and weather due to its uneven distribution in space, rapid change in time, and three-state transformation. Studies have shown that 90% of biogenic VOC emissions in the Southern Hemisphere occur between the equator and 25 degrees south latitude, while isoprene follows a similar latitudinal pattern. It is mainly driven by temperature (Guenther et al., 1995). The uplift index is a convective instability index. It represents the difference between the actual temperature of the 500hpa space and the temperature of an air block that starts from the observation ground, rises to the lifting condensation height along the dry adiabatic line, and then rises along the wet adiabatic line to the temperature of the 500hPa space (Liu J.W. et al., 2005), as shown in Figure 8.

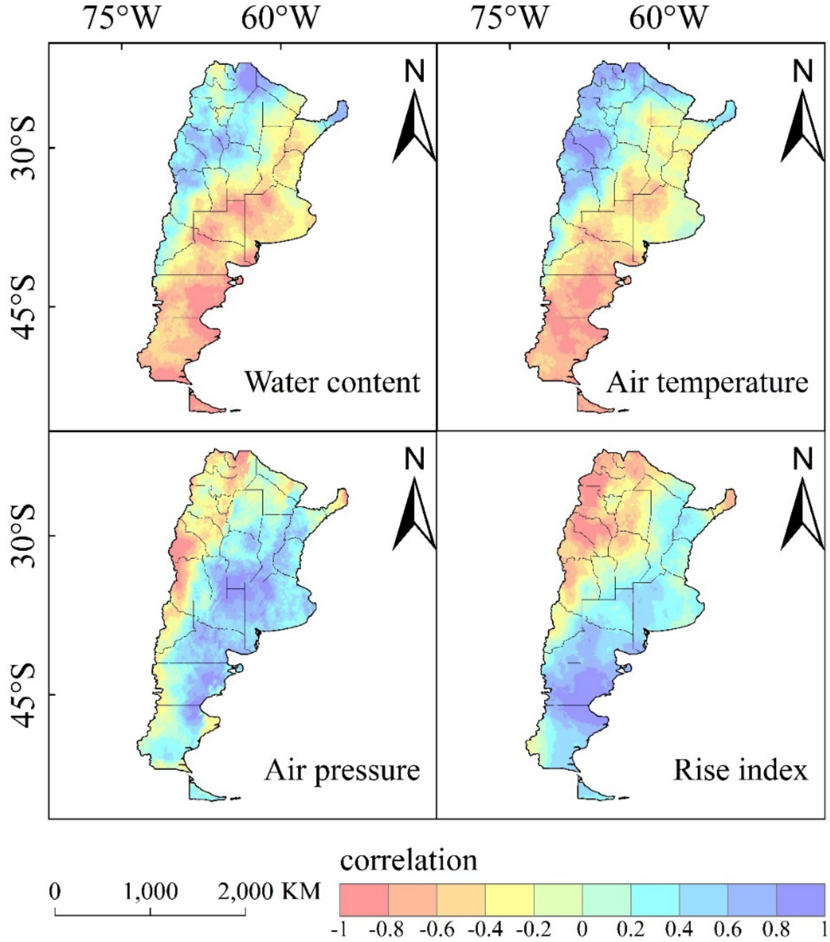

Figure 8: Spatial correlation of formaldehyde column concentration distribution with climatic factors

5. Conclusions

(1) The interannual spatial distribution of formaldehyde column concentration in Argentina is apparent, decreasing gradually from northeast to southwest in general, and the high-value areas are mainly concentrated in Formosa and Misiones provinces.

(2) From the perspective of influencing factors, this paper analyzes the influence of vegetation and meteorological factors on formaldehyde in Argentina. There was a positive correlation between formaldehyde and NDVI, indicating that vegetation greatly influenced formaldehyde.

(3) According to the Slope and Hurst index, formaldehyde in 59.14% of Argentina shows an unchanged trend from 2010 to 2021, while 32.17% of Argentina shows a slight increase.

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