

HALOCOBALOXIMES CONTAINING COORDINATED PYRAZINE, PYRAZINE CARBOXYLIC ACID AND PYRAZINE CARBOXAMIDE: MICROWAVE ASSISTED SYNTHESIS, CHARACTERIZATION AND ANTIBACTERIAL ACTIVITY

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ABSTRACT

About nine cobaloximes of the type $\text{Trans} -[\text{Co}(\text{dmgH})_2(\text{B})\text{X}]$, where $\text{dmgH} = \text{Dimethylglyoximate}$, $\text{X} = \text{Cl}^-$, Br^- or I^- and $\text{B} = \text{Pyrazine}$, Pz (**1 to 3**), $\text{Pyrazine carboxylic acid}$, PzCA (**4 to 6**) or $\text{Pyrazine carboxamide}$, PzAM (**7 to 9**) were prepared and characterized by elemental analysis, TG/DTA, FABMS UV-visible, IR and NMR spectra. The ligands and their cobalt(III) complexes were screened against microorganisms for their antibacterial activity and compared against tetracycline as standard. The free ligands viz., Pz , PzCA and PzAM showed antibacterial activity in the order: $\text{Pz} > \text{PzCA} > \text{PzAM}$ whereas, the free equatorial ligand dmgH_2 was found to be inactive against all the bacteria tested. The cobaloximes were found to be more active than the corresponding pyrazine and its derivatives as axial ligand in the complexes. Bromocobaloximes are found to be more active than chloro- and iodocobaloximes with the antibacterial activity order for the axial halides as $\text{Br}^- > \text{I}^- > \text{Cl}^-$ for complexes having axial pyrazine; whereas, a slightly different trend viz., $\text{I}^- > \text{Br}^- > \text{Cl}^-$ was noticed for halocobaloximes containing axial pyrazine carboxylic acid or pyrazine carboxamide.

Key words: Cobalt(III) complexes, Cobaloximes, Pyrazine, Pyrazine carboxylic acid, Pyrazine carboxamide, Antibacterial activity of metal complexes.

INTRODUCTION

Derivatives of heterocyclic compounds such as pyridazine, pyrimidine and pyrazine¹ are valuable chemotherapeutic agents. Pyrazine ring is present in number of naturally occurring compounds in the form of pteridins and antibiotics like aspergillic acid. Benzo pyrazine, commonly called quinoxaline, has valuable diagnostic properties. Most of them occur in association with proteins, in all living cells and are constituents of chromosomes and viruses. Pyrazine ring is a part of numerous drugs. The first of them was pyrazine carboxamide (PzAM), the first-line antimycobacterial agent, for which promising antibacterial and antifungal activity have been reported². Amides³ prepared from the substituted pyrazine-2-carboxylic acid also possess antibacterial properties. Today tuberculosis (TB) is one of the major world wide public health problems^{4,5}. Pyrazine derivatives may be used as antimycobacterial drugs especially, for the treatment of multi-drug resistant tuberculosis, a major growing problem among HIV-infected patients. Metal complexes have been tested for their antibacterial activities. For example, Schiff's base complexes of nickel⁶ and thio semicarbazone complexes of copper⁷ showed moderate to good antibacterial activity against several bacteria. Our present studies aim at the use of Pz , PzCA and PzAM as effective donors for cobaloximes and as possible antibacterial active metal complexes.

EXPERIMENTAL

Materials and Methods

Cobalt(II) carbonate obtained from (Loba Chemie, Mumbai) was treated with calculated amount of aq.HBr and evaporated to get crystals of cobalt(II) bromide. Cobalt(II) chloride(NICE Chemical Pvt Ltd. Cochin) , SISCO sample of dimethylglyoxime (E.Merck) and Sigma-Aldrich samples of Pz , PzCA and PzAM were used for the preparation of the complexes. Dimethyl sulphoxide (SD Research laboratory) was dried over calcium hydride and distilled fractionally at reduced pressure. The distilled solvent was stored under molecular sieves and used.

Preparation of the cobaloximes

Preparation of chlorocobaloximes

The green colored *Trans*-Hydrogen dichlorobis(dimethylglyoximato)cobaltate(III), $H[Co(dmgH)_2Cl_2]$ was prepared by the method reported by Marzilli *et.al*⁸ Using this complex, as the starting material, the other chlorocobaloximes viz., $[Co(dmgH)_2(Pz)Cl]$, $[Co(dmgH)_2(PzCA)Cl]$ and $[Co(dmgH)_2(PzAM)Cl]$ were prepared by adopting the method reported earlier⁹ for the synthesis of the complexes having pyridine carboxamides .Hydrogen dichlorobis(dimethylglyoximato)cobaltate(III) (0.01mole) and the corresponding 0.01 mole of the substituted pyrazine & pyrazine derivatives viz., Pz , PzCA or PzAM were taken in about 60 mL of absolute alcohol and stirred for 60 min with warming, over water bath for 1hr until the green colour due to dichloro complex was discharged to get the required brown colored complexes.

Preparation of bromocobaloximes

Using cobalt(II) bromide and dimethylglyoxime , the green colored *Trans*-Hydrogen dibromobis(dimethylglyoximato)cobaltate(III), $H[Co(dmgH)_2Br_2]$ and the other complexes derived from it viz., $[Co(dmgH)_2(Pz)Br]$, $[Co(dmgH)_2(PzCA)Br]$ and $[Co(dmgH)_2(PzAM)Br]$ were prepared by the same methods as explained above.

Preparation of iodocobaloximes

Hydrogen dichlorobis(dimethylglyoximato)cobaltate(III), (0.01mole) was mixed with 30 mL of water and exposed to microwave irradiation for 3 min at regular intervals of 30 s each time after cooling down to room temperature. The completion of the reaction was indicated by the color change from green to light brown indicating the formation of aquochlorobis(dimethylglyoximato)cobalt(III), $[Co(dmgH)_2(H_2O)Cl]$. The aquochloro complex, viz., $[Co(dmgH)_2(H_2O)Cl]$ thus obtained, was filtered and treated with 0.01mole of KI in 30 mL of water and subjected to microwave irradiation as above till the solution turned dark brown¹⁰ . The brown colored product, aquoiodobis(dimethylglyoximato)cobalt(III) viz., $[Co(dmgH)_2(H_2O)I]$ crystallised when allowed to stand over night . The aquoiodocobaloxime thus obtained was mixed separately with the equimolar amounts of Pz or PzCA or PzAM and subjected to microwave irradiation for 3 min as above until the light brown color turned dark brown. The iodocobaloximes , viz., $[Co(dmgH)_2(Pz)I]$, $[Co(dmgH)_2(PzCA)I]$ or $[Co(dmgH)_2(PzAM)I]$ crystallized as dark brown crystals on standing overnight.

Characterization of the complexes

The complexes were characterized by elemental, thermal analysis, FABMS (CDRI, Lucknow), UV-visible, IR and NMR. The cobalt in the complexes was estimated by Kitson method¹². The UV-visible spectra were recorded on LAMBDA-125 spectrophotometer in ethanol using matched quartz cells of path length 1 cm, IR spectra were obtained on KBr pellet on PERKIN ELMER IR Spectrum-1 spectrophotometer and the NMR spectra were recorded in DMSO-d₆ using JOEL 400 MHz NMR spectrophotometer. The thermograms (TG/ DTA) were recorded on Perkin Elmer Model Pyris Diamond at a heating rate of 10°C min⁻¹ with argon gas purged at the rate of 12 L hr⁻¹. The cyclic voltograms of some cobaloximes on the surface of a Pt disk electrode in acetonitrile solution were recorded in the potential range + 1.6 to -1.6 V using CH instrument model No-620 B Electrochemical analyzer.

Antibacterial activity studies

The human pathogens viz., *Escherichia coli* (ATCC-25922), *Proteus vulgaris* (ATCC-1771), *Proteus mirabilis* (ATCC49565) and *Pseudomonas aeruginosa* (ATCC-27853) were obtained from Christian medical college, Vellore. The cultures were maintained on Nutrient Agar (NA) slant at 4°C. The antibacterial activity of the compounds (ligands and the cobaloximes) were tested against the above human pathogens using agar diffusion method¹¹. The bacterial cultures were grown in Mueller Hinton broth (MHB) medium under shaken condition at 37 °C. Inoculum of 1 mL drawn from each test pathogen was added to molten Mueller Hinton Agar (MHA) medium and poured into sterile petri plates under aseptic conditions. After solidification, a 5 mm well was made in the center of each plate using a sterile cork borer. Each compound was dissolved in DMSO to get different concentrations (2.5, 5.0, 7.5 and 10mg/ml). In each well, 50 µL solution of each compound was added separately and the plates were incubated at 37°C for 24 hrs with DMSO used as negative control. After incubation period the appearance of inhibition zone around the well was observed.

The minimum inhibitory concentration (MIC) for each pathogen was determined according to the standard reference method¹³. The compounds were dissolved in water + 2% DMSO (v/v). The initial concentration of compound was 10 mg/ml. The initial test concentration was serially diluted two-fold. Each well was inoculated with 5 µl of suspension containing 10⁸ CFU/ml of bacteria. Tetracycline was included in the assays as positive control. The plates were incubated for 24 hrs at 37°C. After incubation period, 5 µl of tested broth was placed on the sterile MHA plates and incubated at respective temperature. The MIC was determined as the lowest concentration of the compound inhibiting the visual growth of the test cultures on the agar plate. Three replications were performed.

RESULTS AND DISCUSSION

Dichloro- and dibromocobaloximes are intense green crystalline compounds. They turn brown on treatment with one mole of heterocyclic donors like pyridine or pyridine carboxylic acids or pyridine carboxamides¹⁴. Hence, Pz, PzCA and PzAM should also react in the same manner towards dihalocobaloximes. In fact, the color change and the nature of the reactivity of the Pz, PzCA and PzAM towards dihalocobaloximes were found to be much similar to pyridine, 4-pyridine carboxylic acid and 4-pyridine carboxamide.

Elemental analysis

The elemental analysis data, obtained by analytical methods agree well with the theoretical data proposed for the complexes (Table-1).

Thermal data

The data of thermal studies of the cobalt(III) complexes suggests that the axial halogen dissociates at 230° C, followed by the dissociation of pyrazine¹⁵ based axial ligand around 250° C. The remaining residual complex viz; [Co(dmgh)₂] might be stable up to 340° C, after which it may be decomposed to Co₂O₃ a final residue¹⁶.

FAB Mass spectra

The molecular ion peak of the fast atom bombardment mass spectra (FABMS) of the cobaloximes confirm their molecular weights. [Co(dmgh)₂(PzCA)Cl]: m/z = 448.5 and [Co(dmgh)₂(PzAM)Cl] m/z = 447

UV-Visible spectra

The UV spectrum of pyrazine, pyrazine carboxamide and pyrazinic carboxylic acid show an absorption around 260 nm, which may be attributed to $\pi \rightarrow \pi^*$ transition of the pyrazine ring. This absorption band remains unchanged in their complexes. The band around 245 nm may be due to $n \rightarrow \pi^*$ of Pz, PzCA and PzAM. A shoulder around 330 nm may be due to the ligand to metal charge transfer (LMCT). This LMCT peak was found to be disappearing upon reduction of the cobalt(III) by Fe(II) or Cr(II)^{17,18}. The weak transition around 570 nm for the complexes may be attributed to d⁶ low spin cobalt(III) spin allowed d-d transitions viz., ¹A_{1g}, ¹T_{1g} with the other high energy spin allowed d-d transition viz., ¹A_{1g}, ²T_{1g} occurring in the range 370-385 nm.

IR spectra

The C=N stretching of oxime in its complexes was observed in the range 1550-1600 cm⁻¹ and the intra molecular hydrogen bonded -OH around 3400 cm⁻¹. A moderate intense peak around 1240 cm⁻¹, observed for all the complexes, may be assigned to the =N-O- stretching of the oxime in the complexes. The peaks at 3320 and 3440 cm⁻¹ confirm the free -NH₂ group of the amide group of PzAM. The peak around 520 cm⁻¹ observed for all the cobaloximes could be attributed to cobalt(III)-nitrogen stretching.

NMR Spectra

The methyl protons of dimethylglyoxime, in all the cobaloximes, appear as a sharp singlet at $\delta = 2.3$ ppm (12H, s)¹⁹⁻²⁰ whereas the hydrogen bonded hydroxyl protons of the oxime resonate at $\delta = 8.7$ ppm (2H, s). The pyrazine protons in the complexes of the type: [Co(dmgh)₂(Pz)X] appear as doublet at $\delta = 8.0$ (d, 4H). The pyrazine carboxylic acid protons in its complexes, viz., [Co(dmgh)₂(PzCA)X] show three series of lines at $\delta = 8.6$ (s, 3H, 1H); $\delta = 8.2$ (d, 5H, 1H) and $\delta = 7.4$ (d, 6H, 1H) due to three different aromatic protons with the carboxyl protons appearing at $\delta = 0.8$ (s, 1H). Similarly, the complexes of the third type, viz., [Co(dmgh)₂(PzAM)X] also exhibited three series of lines at $\delta = 8.3$ (s, 3H, 1H); $\delta = 8.2$ (d, 5H, 1H) and $\delta = 8.0$ (d, 6H, 1H) due to the three different aromatic protons with the amide -NH₂ protons appearing at $\delta = 3.4$ (s, 2H). The oxime carbons appear at 153 ppm followed by the pyrazine carbons appearing at 145 ppm. The methyl carbons of the equatorial oxime resonate at 12 ppm.

Cyclic Voltametry

The cyclic voltagrams of the cobaloximes exhibited two well defined cyclic responses at -0.9 V and -1.4 V corresponding to Co(III) / Co(II) and Co(II) / Co couples, respectively²¹.

Antibacterial studies

The results of antibacterial activity of tested compounds showed significant activity in Pz, PzCA and PzAM. The zone of inhibition recorded in these cobaloximes ranged from 18 mm and 30 mm as against 2 mm to 29 mm for tetracycline¹² (Table-2). The maximum zones of inhibitions were noticed for the complexes containing PzCA than those containing Pz or PzCAM. Pyrazinecarboxylicacidiodocobaloxime(III) showed significant activity against *Escherichia coli* at 10 mg/ml (29 mm), *Proteus vulgaris* at 10mg/ml (29 mm), *Proteus mirabilis* at 10 mg/ml (29 mm) and *Pseudomonas aeruginosa* at 10mg/ml (29 mm). In general the cobaloximes showed more activity than the respective free ligands.

The antibacterial activities of the cobaloximes were found to be dose dependent and were remarkable at higher concentrations. In majority of the cobaloximes the antibacterial activity was higher than the commercial antibiotic, tetracycline in certain concentrations. The over all analysis on the antibacterial activity revealed that among all the tested cobaloximes, chloro cobaloximes are less active against the pathogens tested. Iodo cobaloximes carrying substituted pyrazine have superior activity than the corresponding bromocobaloximes. A different trend was noticed for pyrazinecobaloximes. Pyrazinebromocobaloxime was found to be more active than pyrazineiodocobaloxime and they were always superior over tetracycline in all the tested concentrations (Table-2). The general trend for halopyrazinecobaloximes was $\text{Br}^- > \text{I}^- > \text{Cl}^-$; whereas, a slightly different trend viz., $\text{I}^- > \text{Br}^- > \text{Cl}^-$ was observed for halocobaloximes containing pyrazine carboxylic acid or pyrazine carboxamide. Similar results were noticed for the MIC measurements (Table-3)

CONCLUSIONS

The halogenocobaloximes with axial Pyrazine (Pz), Pyrazine carboxylic acid, (PzCA) or Pyrazine carboxamide (PzAM) were prepared and characterized. The free ligands: Pz, PzCA and PzAM showed antibacterial activity in the order: $\text{Pz} > \text{PzCA} > \text{PzAM}$ whereas, the free equatorial ligand dmgH_2 was inactive against all of the bacteria tested. The cobaloximes were found to be more active than the corresponding pyrazine and its derivatives as axial ligand in the complexes. Bromocobaloximes are found to be more active than chloro- and iodocobaloximes with the antibacterial activity order for the axial halides as $\text{Br}^- > \text{I}^- > \text{Cl}^-$ for a given other axial pyrazine type of ligand; whereas, a different trend viz., $\text{I}^- > \text{Br}^- > \text{Cl}^-$ was noticed for halocobaloximes containing Pz or PzCA or PzAM.

ACKNOWLEDGEMENTS

The authors are thankful to Rev.Fr.A.Albert Muthumali, S.J., *Principal*, Loyola College (*Autonomous*), Chennai-34 for providing the necessary facilities, The director, LIFE, Loyola College (*Autonomous*), Chennai-34 for the supply of thermal data, The Head, SAIF, IIT, Madras for recording NMR spectra and Dr. K.P. Madhusudanan, Head, SAIF, CDRI, Lucknow, for the supply of the elemental & FABMS data.

Table-1: Elemental data for cobaloximes

Code	Complex	C		H		N		Co	
		Exp	Cal	Exp	Cal	Exp	Cal	Exp	Cal
1	[Co(dmgh) ₂ (Pz)Cl]	42.15	42.35	5.24	5.29	24.73	24.70	17.25	17.35
2	[Co(dmgh) ₂ (Pz)Br]	37.35	37.40	4.61	4.67	20.94	21.01	15.26	15.32
3	[Co(dmgh) ₂ (Pz)I]	33.38	33.41	4.14	4.17	19.41	19.48	13.59	13.68
4	[Co(dmgh) ₂ (PzCA)Cl]	33.79	34.82	4.00	4.02	18.66	18.75	13.15	13.17
5	[Co(dmgh) ₂ (PzCA)Br]	31.62	31.64	3.59	3.65	16.10	17.04	11.87	11.97
6	[Co(dmgh) ₂ (PzCA)I]	28.76	28.89	3.29	3.33	15.52	15.56	10.78	10.93
7	[Co(dmgh) ₂ (PzAM)Cl]	34.81	34.82	4.12	4.02	21.88	21.92	13.20	13.19
8	[Co(dmgh) ₂ (PzAM)Br]	31.65	31.70	3.2	3.65	19.79	19.91	11.82	11.99
9	[Co(dmgh) ₂ (PzAM)I]	28.88	28.94	3.0	3.33	18.03	18.18	10.76	10.94

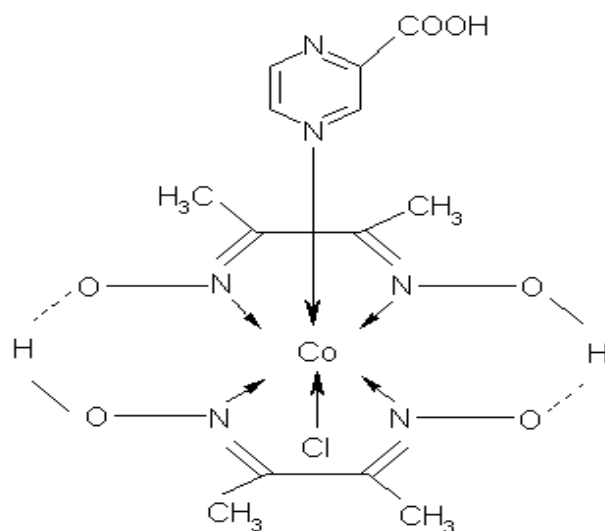
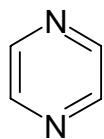
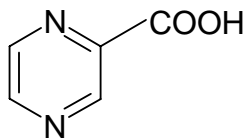


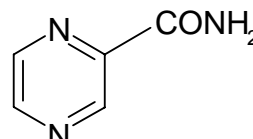
Fig: 1-Structures of the ligands and a typical cobaloxime

[Co(dmgh)₂(PzCA)Cl]

Pz



PzCA



PzAM

Table-2 : Antibacterial activity of Pz , PzCA & PzAM and their cobaloximes against human pathogens

Compound	[§] Zone of inhibition (mm)															
	<i>E. coli</i>				<i>P. vulgaris</i>				<i>P. mirabilis</i>				<i>P. aeruginosa</i>			
	Conc. of the compound (mg/mL) [*]				Conc. of the compound (mg/mL) [*]				Conc. of the compound (mg/mL) [*]				Conc. of the compound (mg/mL) [*]			
	2.5	5.0	7.5	10	2.5	5.0	7.5	10	2.5	5.0	7.5	10	2.5	5.0	7.5	10
Pz	10	13	16	18	8	10	12	15	8	12	15	19	9	13	16	21
[Co(dmgh) ₂ (Pz)Cl]	12	16	20	23	10	12	16	22	6	10	16	18	12	15	19	22
[Co(dmgh) ₂ (Pz)Br]	21	23	25	27	22	24	26	27	18	20	24	27	18	22	24	26
[Co(dmgh) ₂ (Pz)I]	17	20	22	25	14	16	20	24	14	16	18	22	14	18	21	23
PzCA	8	11	14	16	4	6	10	16	2	8	10	14	0	0	12	14
[Co(dmgh) ₂ (PzCA)Cl]	13	15	18	20	10	14	18	22	12	18	22	24	14	15	16	19
[Co(dmgh) ₂ (PzCA)Br]	15	17	19	22	14	18	22	24	18	22	25	26	18	22	24	25
[Co(dmgh) ₂ (PzCA)I]	22	24	26	28	18	22	26	27	21	24	27	28	20	23	25	27
PzAM	4	6	8	12	2	4	10	14	2	6	10	18	0	0	0	0
[Co(dmgh) ₂ (PzAM)Cl]	14	16	19	22	8	16	20	24	4	10	10	16	12	14	18	20
[Co(dmgh) ₂ (PzAM)Br]	19	22	23	26	10	19	24	26	6	15	20	24	16	19	23	26
[Co(dmgh) ₂ (PzAM)I]	22	25	27	28	22	24	27	29	18	20	26	29	20	23	25	29
Tetracycline	2	14	20	26	8	20	24	27	16	18	24	29	16	28	29	29

[§]Values are mean of three replicates.

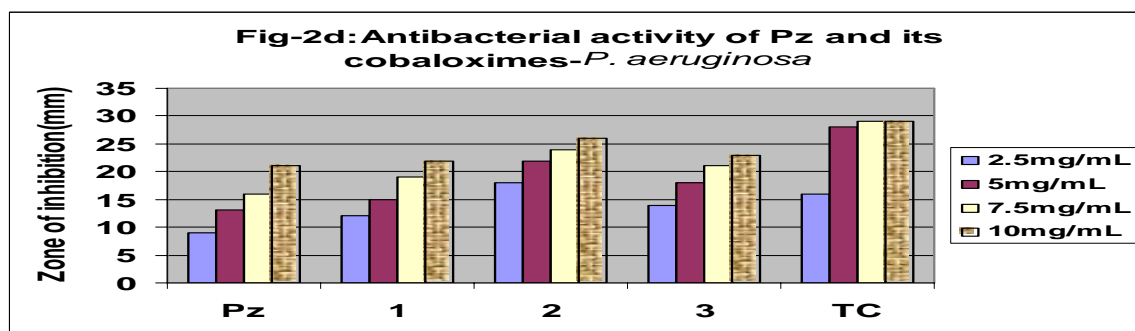
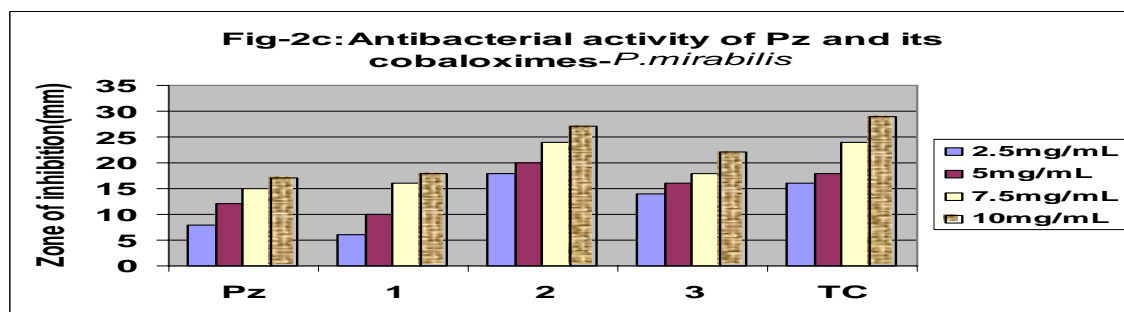
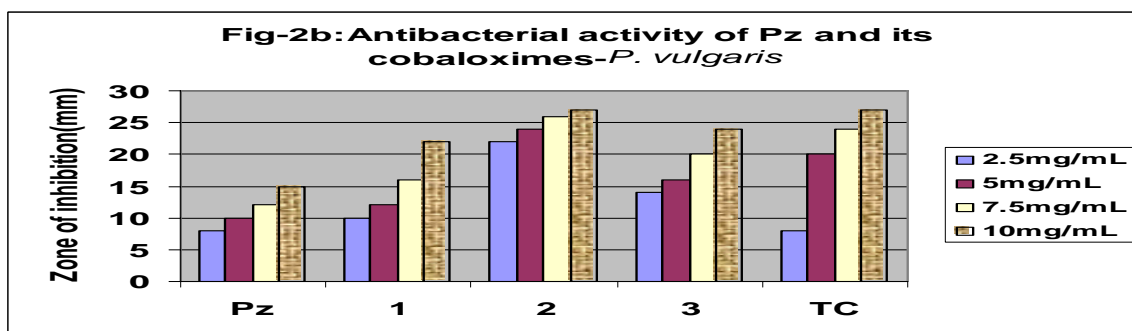
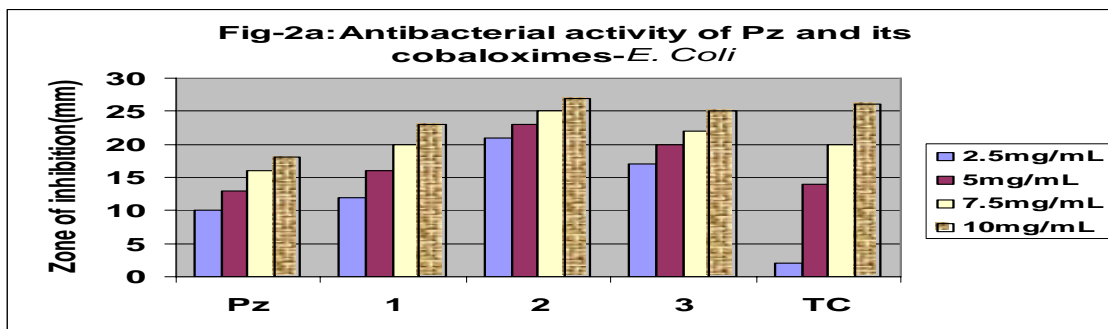
^{*} Volume each solution = 50 μL

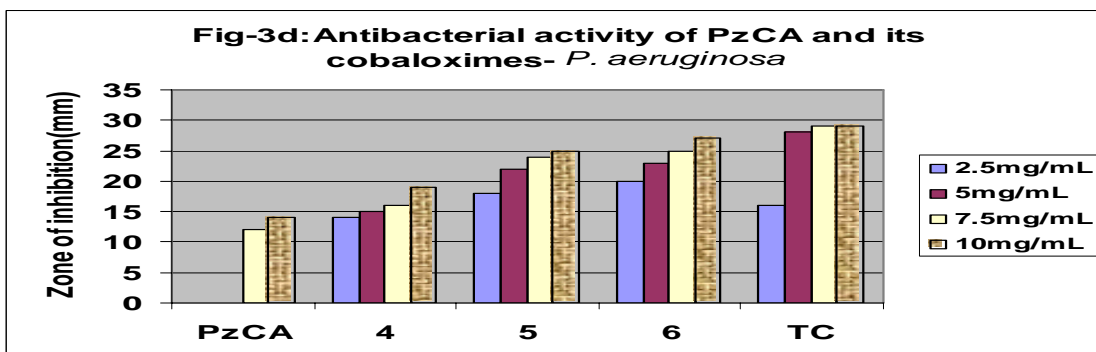
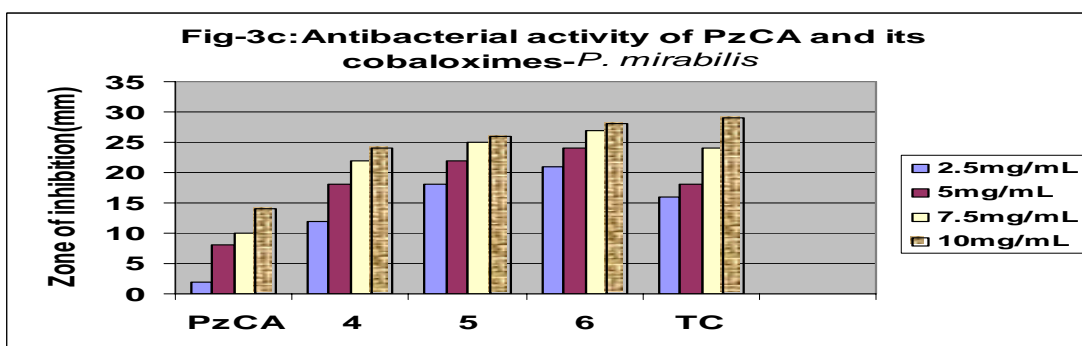
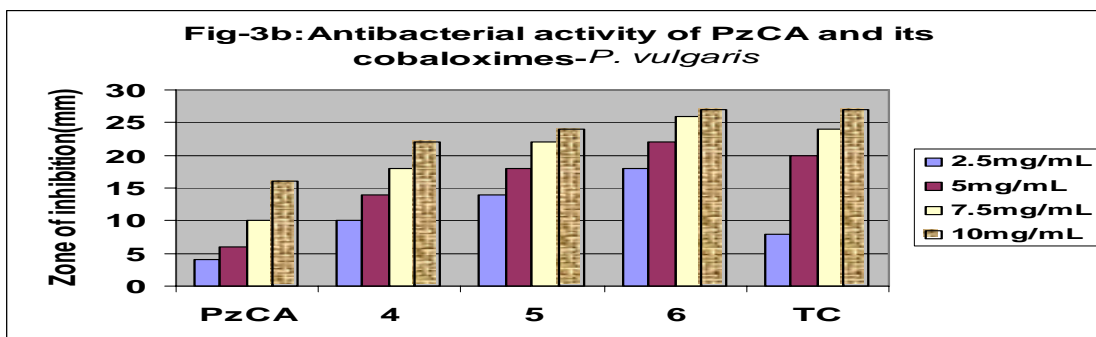
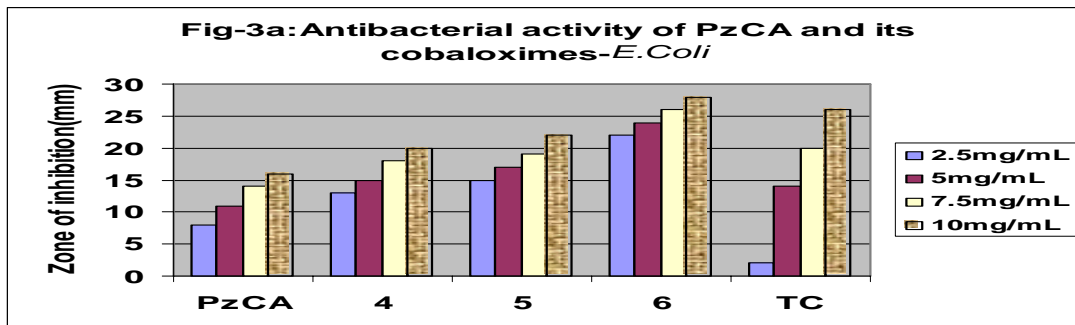
Table-3: *In vitro* antibacterial activity (Minimum inhibitory concentrations, $\mu\text{g/ml}$)

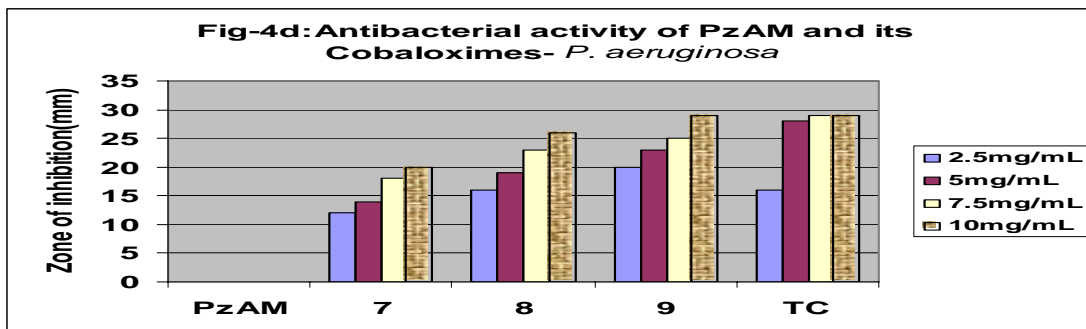
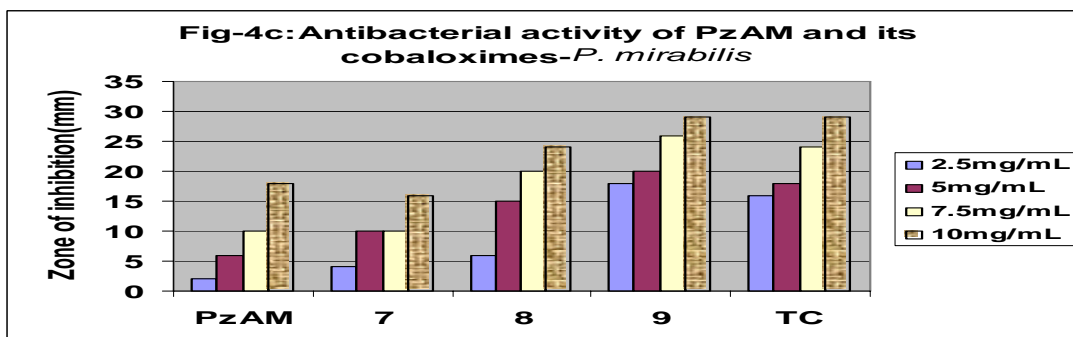
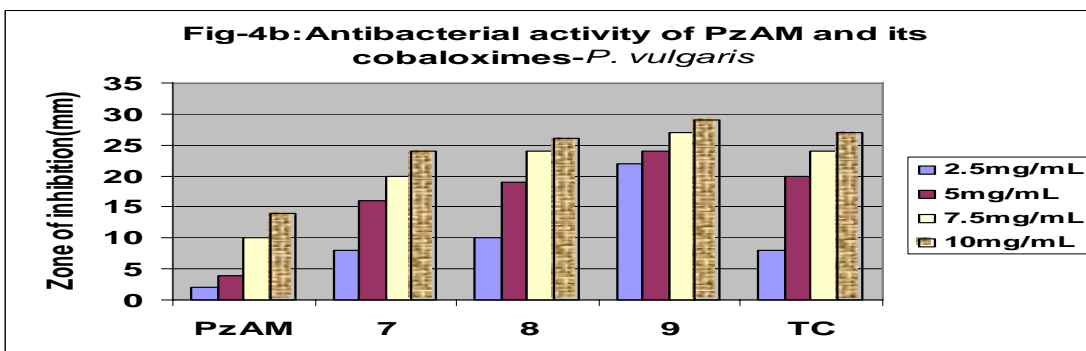
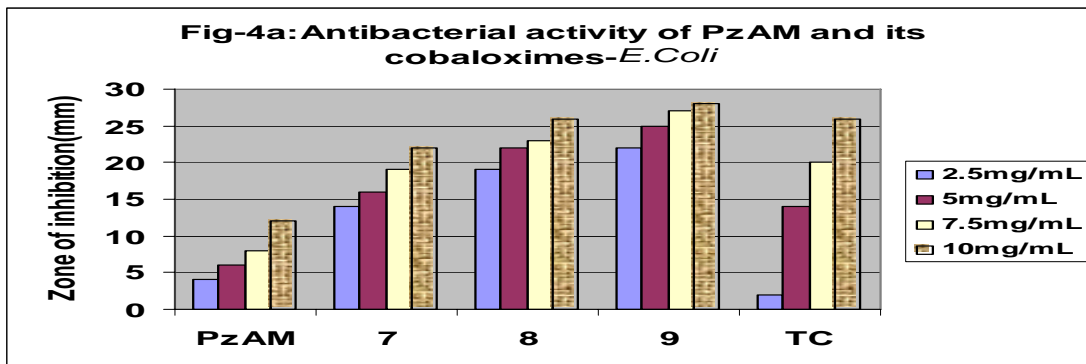
Code	Compound	<i>E. coli</i>	<i>P. vulgaris</i>	<i>P. mirabilis</i>	<i>P. aeruginosa</i>
dmgH₂	Dimethyl glyoxime	-	-	-	-
Pz	Pyrazine	25	30	45	50
1	[Co(dmgH) ₂ (Pz)Cl]	25	25	35	40
2	[Co(dmgH) ₂ (Pz)Br]	15	15	15	25
3	[Co(dmgH) ₂ (Pz)I]	20	20	30	35
PzCA	Pyrazinecarboxylic acid	30	45	50	55
4	[Co(dmgH) ₂ (PzCA)Cl]	40	35	45	35
5	[Co(dmgH) ₂ (PzCA)Br]	35	25	40	30
6	[Co(dmgH) ₂ (PzCA)I]	30	20	30	25
PzAM	Pyrazinecarboxamide	60	75	70	-
7	[Co(dmgH) ₂ (PzAM)Cl]	55	50	50	65
8	[Co(dmgH) ₂ (PzAM)Br]	40	45	45	35
9	[Co(dmgH) ₂ (PzAM)I]	35	30	35	30
TC	Tetracycline	35	20	50	35
	Control(2% aq DMSO(v/v))	-	-	-	-

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(Received:6 April 2008

Accepted: 25 April 2008

RJC-167)

**International Conference of Physical Chemistry,
ROMPHYSCHEM13
Bucharest (Guadeloupe)
3-5 September 2008
Email: vtpopa@icf.ro
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